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A hybrid modeling approach to resolve pollutant concentrations in an urban area

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Abstract

A modeling tool that can resolve contributions from individual sources to the urban environment is critical for air-toxics exposure assessments. Air toxics are often chemically reactive and may have background concentrations originated from distant sources. Grid models are the best-suited tools to handle the regional features of these chemicals. However, these models are not designed to resolve pollutant concentrations on local scales. Moreover, for many species of interest, having reaction time scales that are longer than the travel time across an urban area, chemical reactions can be ignored in describing local dispersion from strong individual sources making Lagrangian and plume-dispersion models practical. In this study, we test the feasibility of developing an urban hybrid simulation system. In this combination, the Community Multi-scale Air Quality model (CMAQ) provides the regional background concentrations and urban-scale photochemistry, and local models such as Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) and AMS/EPA Regulatory Model (AERMOD) provide the more spatially resolved concentrations due to local emission sources. In the initial application, the HYSPLIT, AERMOD, and CMAQ models are used in combination to calculate high-resolution benzene concentrations in the Houston area. The study period is from 18 August to 4 September of 2000. The Mesoscale Model 5 (MM5) is used to create meteorological fields with a horizontal resolution of $1 \times 1\,km^2$. In another variation to this approach, multiple HYSPLIT simulations are used to create a concentration ensemble to estimate the contribution to the concentration variability from point sources. HYSPLIT simulations are used to model two sources of concentration variability; one due to variability created by different particle trajectory pathways in the turbulent atmosphere and the other due to different flow regimes that might be introduced when using gridded data to represent meteorological data fields. The ensemble mean concentrations determined by HYSPLIT plus the concentrations estimated by AERMOD are added to the CMAQ calculated background to estimate the total mean benzene concentration. These estimated hourly mean concentrations are also compared with available field measurements.
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1. Introduction

Hazardous air pollutants, that include hundreds of chemical species, contribute to a wide variety of

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human health and ecological effects. Due to the large number of air toxics, the relatively small number of pollutants measured, and the sparse nature of routine monitoring networks, many regulatory agencies rely on air quality models to estimate ambient concentrations and compare results to monitoring data where available to evaluate model performance. Air quality models are useful tools for assessing baseline ambient concentrations, analyzing the relative importance of various emission sources, and testing emission reduction strategies. These assessments typically involve the application of different models depending on program objectives—national, regional, urban, or local scale (Fig. 1).

Air toxics emitted in significant quantities from isolated sources may have important local impacts. Air toxic pollutants, such as benzene vapors and diesel fine 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. Resolving these "hot spots" is critical for air-toxics exposure assessments, for model evaluation studies, and also for air quality regulatory applications.

There are several available modeling approaches capable of assessing pollutant concentration gradients at a fine resolution (Touma et al., 2006) and these can be categorized into two major types of air quality models: source-based dispersion models and Eulerian grid-based chemical transport models.

Source-based dispersion models use either plume, puff or particle 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 and their range of application is from a few hundred meters to a few kilometers from the source. The temporal resolutions range from an hour to an entire year (annual average). In general, they are not very computationally demanding. However, the computational burden can become excessive if local-scale models are applied to urban-scale domains (e.g., $100 \times 100\,km^2$) involving thousands of emission sources and receptors. For some of the pollutants, background concentrations must

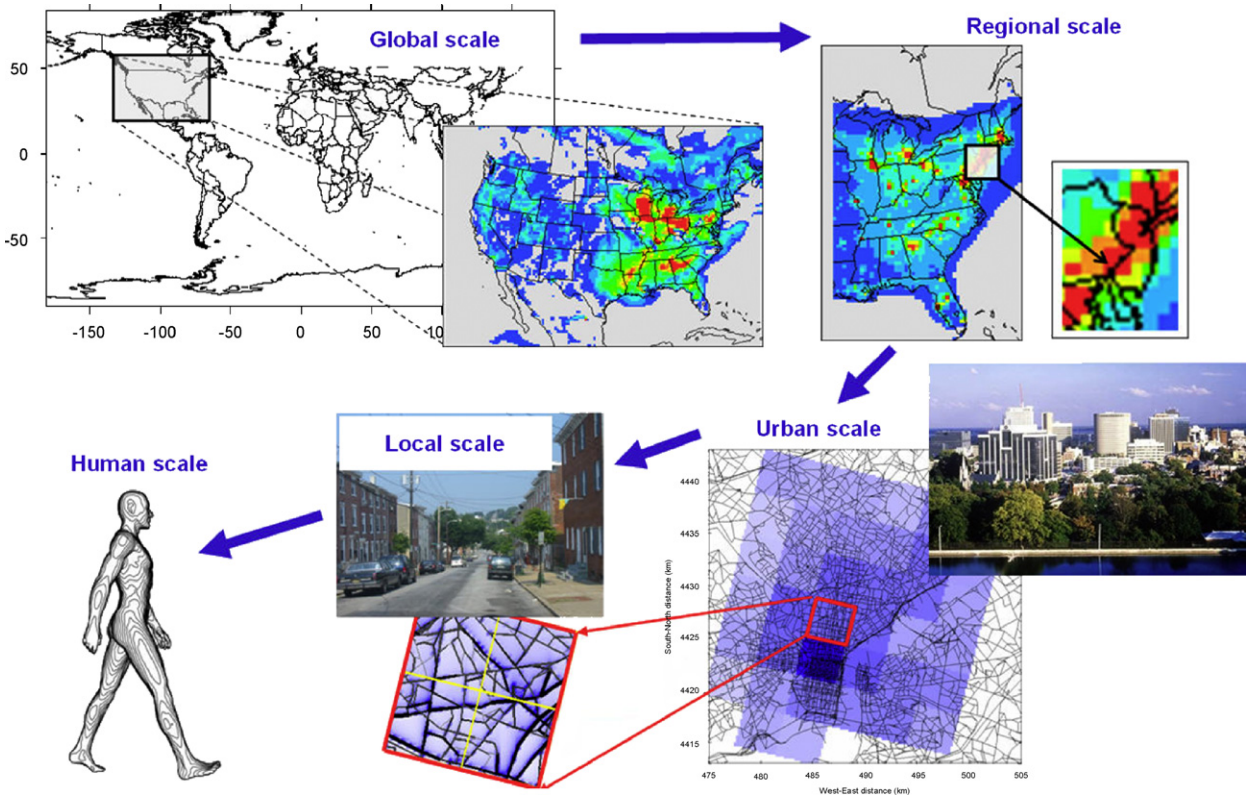

Fig. 1. Multiple scales in air quality modeling.

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 and may include contributions from long-range transport of air toxics from distant sources (i.e., located outside the local modeling domain), non-inventoried anthropogenic emissions, and natural emissions.

On the other hand, Eulerian grid-based models (such as the Community Multi-scale Air Quality (CMAQ)) are used to simulate the transport and formation of ozone, 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 applied over hundreds of kilometers using an array of grid cells with a horizontal grid resolution of several kilometers up to tens of kilometers. These 3-dimensional grid models require considerable computational resources and are typically applied only for several multi-day periods over the duration of an entire year to represent long-term averages. 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 their 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 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 neither within the source grid cell nor on any of the nearby cells. Reducing grid cell size to correspond to the size of the area of interest is possible (Jacobson and Seinfeld, 2004); however it could become computationally very demanding and currently there are technical limitations to reducing grid size below about a kilometer. For many pollutants, there is evidence of significant spatial variability at scales <1 km (Weijers et al., 2004).

It would be desirable to combine the capabilities of chemical grid and source-based dispersion models into one coupled modeling system, but this is as yet an evolving area of research and development. One viable option is a hybrid approach (Isakov and Venkatram, 2006; Isakov et al., 2007a), where a regional grid model and local-scale plume models are run independently and later combined. In this paper, we demonstrate this technique using an example application of the hybrid modeling approach in Houston, Texas and compare results with available measurements from monitoring sites.

2. Methodology

A grid-based model is the tool of choice for the simulation of atmospheric chemistry and fate of airborne pollutants. However, increasing its resolution to solve local features presents technical limitations and very long computation times. On the other hand, there are various transport and diffusion models developed to simulate the fate of relatively chemically inert airborne pollutants that can provide detailed resolution of the spatial variations in hourly average concentrations of airborne pollutants with shorter computation times. To date, local-scale dispersion models have been relied upon to provide the desired detailed description of the concentration pattern but they cannot properly treat photochemical effects. Some of the air toxic pollutants are identified as having a photochemical origin or being affected by photochemical processes. Therefore, chemical species that are either non-reactive or react slowly, such as benzene, or even relatively fast reactive pollutants that can be assumed to decay as a first-order process can be modeled using these local-scale dispersion models. An estimate of the background concentration levels can be provided by the grid-based models.

A hybrid approach (Isakov et al., 2007a) is a logical and efficient way to combine regional grid and local plume models. In this approach, the regional grid model provides the regional background concentrations and urban-scale photochemistry, and the local plume-dispersion model provides the air-toxics concentrations due to local emission sources assuming non-reactive chemistry on subgrid scales. Then, the results of both model simulations are combined to provide the total ambient air pollutant concentrations.

The hybrid approach uses the appropriate modeling tools to describe different types of sources, making its application computationally efficient. Furthermore, since local dispersion models are not resource intensive, this methodology allows the study of local concentration variability in the local plume simulation, helping to gain confidence in the

simulation results by encompassing a range of model outcomes. This constitutes a clear advantage of the hybrid approach, since performing a local concentration variability estimation using a nested grid model alone would be an impractical task. The descriptions of each of the modeling components of the hybrid approach are presented in the following sections.

2.1. Regional background

The CMAQ modeling system (Byun and Schere, 2006) was selected for simulation of regional transport and photochemical transformations in this hybrid modeling application. CMAQ constitutes a state-of-the-science tool for regional-scale simulations of photochemical smog, visibility, toxics, and fine particulates. It is a three-dimensional Eulerian chemical transport model that accounts for horizontal and vertical advection, eddy diffusion, gas-phase chemical transformations, emissions, cloud mixing, aqueous-phase chemical reactions, and aerosol processes.

2.2. Stationary sources

Stationary sources have both local and long-range impact on air quality. In order to characterize the impact of stationary sources, we selected the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) to simulate the emission, transport, and dispersion of pollutants in the troposphere. The model simulates the pollutant emission by releasing a fixed number of Lagrangian particles (point masses) having a mass determined by the source strength. Once in the atmosphere, these particles are moved by a wind having mean and random components. Furthermore, the pollutant concentration is calculated by dividing the sum of the particle masses of the pollutant by the volume of the corresponding user-defined concentration grid cell in which the particles reside. A detailed model description can be found in Draxler and Hess (1997, 1998).

One of the major advantages of using HYSPLIT in the hybrid modeling application is its capability to simulate the transport of pollutants in a complex flow pattern. In addition, HYSPLIT can be used to estimate the uncertainty of a particular modeling application. In order to estimate the uncertainty in modeled concentrations, HYSPLIT is used as an ensemble modeling application in which multiple HYSPLIT simulations are conducted to create the concentration ensembles used to estimate the variability in modeled concentrations from stationary sources. This is especially important in applications for air toxics, where the emission inventory is uncertain for source locations and stack parameters. Two sources of concentration variability have been investigated in this work. The first one represents the variability due to different flow regimes that are introduced when using gridded data to represent meteorological data fields. The second type of variability simulated arises from different, and all equally valid, particle trajectory pathways created in the turbulent atmosphere due to the trajectory changes introduced by the random component of the particle diffusion.

2.3. Near-road impacts

Several studies have found that concentrations of mobile source related pollutants are significantly higher near busy roadways compared to the urban background (Sapkota and Buckley, 2003; Skov et al., 2001). Current approaches for characterizing ambient air toxic concentrations near roadways rely on developing a detailed emissions inventory and applying a dispersion model, such as the AMS/EPA Regulatory Model (AERMOD) (Touma et al., 2006). AERMOD is a steady-state plume-dispersion model for air quality assessments of inert pollutants that are directly emitted from a variety of sources (Cimorelli et al., 2005; Perry et al., 2005; US EPA, 2004a). Based on an advanced characterization of the atmospheric boundary layer turbulence structure and scaling concepts, AERMOD is applicable to rural and urban areas, flat and complex terrain, surface and elevated releases, and multiple sources (including point, area, or volume sources). The model employs hourly sequential preprocessed meteorological data to estimate concentrations at receptor locations for averaging times from 1 h to 1 year. AERMOD incorporates both dry and wet particle and gaseous deposition.

AERMOD assumes concentrations at all distances during a modeled hour are governed by the set of hourly meteorology inputs, which are held constant. AERMOD constructs vertical profiles of required meteorological variables based on measurements and extrapolations of those measurements using similarity (scaling) relationships. Vertical profiles of wind speed, wind direction, turbulence, and temperature are estimated using all

available meteorological observations. However, using NWS observations could pose problems because observation sites can be located tens or even hundreds of kilometers from the location at which AERMOD is being applied. Also, upper air meteorological data needed to estimate mixing heights are usually not collocated with the surface observations. Thus, these data may not be representative of the application site. Furthermore, because the data have to be quality controlled and archived by the National Climatic Data Center (NCDC), they might not be available for months after they are collected. One possible way of solving this problem with meteorological inputs is to use comprehensive meteorological models to provide estimates of the boundary layer variables required by AERMOD at the site of interest. There is an extensive history of coupling numerical weather prediction models and dispersion models (e.g., Yamada et al., 1992; Draxler and Hess, 1998; Draxler, 2003; Isakov et al., 2007b). Outputs from prognostic models run by the National Oceanic and Atmospheric Administration (NOAA) are available in near-real-time over a 12 km grid system that covers the United States. Thus, a model user has ready access to the most recent meteorological data, which can be used in air quality simulations with little further processing. In addition, meteorological models can provide reasonably representative inputs where representative measurements are not available. However, these inputs need to be carefully evaluated prior to using them as inputs for AERMOD (Isakov et al., 2007b).

In this study, we combined CMAQ, HYSPLIT, and AERMOD in a hybrid modeling application, where CMAQ provides the regional background concentrations and urban-scale photochemistry, HYSPLIT provides the spatially resolved concentrations due to selected point emission sources, and AERMOD simulates mobile emissions from selected major roads. Furthermore, multiple HYSPLIT simulations are used to generate a concentration ensemble to estimate the concentration variability from point sources.

3. Example of application: Houston, TX case study

To illustrate an application of the hybrid modeling approach, we focus on a 36 km × 36 km area in Houston, TX. Benzene was chosen for this exercise because its reaction time scales are longer than the travel time across the urban area under study, further justifying the use of HYSPLIT and AERMOD. However, since benzene's lifetime in the lower troposphere is approximately 12 days (Seinfeld and Pandis, 1998) background concentrations need to be estimated with a photochemical model such as CMAQ. Thus, benzene is appropriate for use in the HYSPLIT/CMAQ/AERMOD modeling application. The hybrid-modeling domain is shown in Fig. 2. This study area includes several thousand sources emitting small amounts of benzene, mostly at ground level from roadway traffic, but also from some very large stationary sources such as refineries.

Houston was chosen for this project because of the diversity and magnitude of air pollution sources and their impact on the nearby community, the availability of field measurement data for benzene, and because this geographical area was also a part of the Texas 2000 air quality study (http://www.utexas.edu/research/ceer/texaqs/).

Fig. 2 also shows the location of the six monitor sites available in 2000. Monitor data provide both hourly and 24-h average concentrations for multiple pollutants. However, for the study period, most of the stations measure 24-h average benzene concentrations. Monitors 1 and 3 are located in a suburban industrial environment. Monitors 2, 4, 5, and 6 are located in residential zones. Monitor 2 corresponds to an urban environment while stations 4, 5, and 6 are located in suburban areas (Reiss, 2006).

3.1. Emissions

The inventories for the case study were developed from the 2002 draft National Emissions Inventory (NEI) (US EPA, 2004b). The total emissions inventory is contained in four separate inventories: point, non-point, on-road, and non-road.

The point inventory contains major, area and other sources that have reasonably known location coordinates. The top six point sources have been modeled using the HYSPLIT model. Table 1 describes the main features of these sources. It should be taken into account that there are multiple stacks in each facility and the emissions inventory for these sources is self-reported, making the stack parameters very uncertain. Consequently, conducting an ensemble modeling analysis that includes horizontal and vertical variation of the location of the sources relative to the meteorological data fields is essential.

The non-point inventory contains county-level area and other emissions by Source Classification

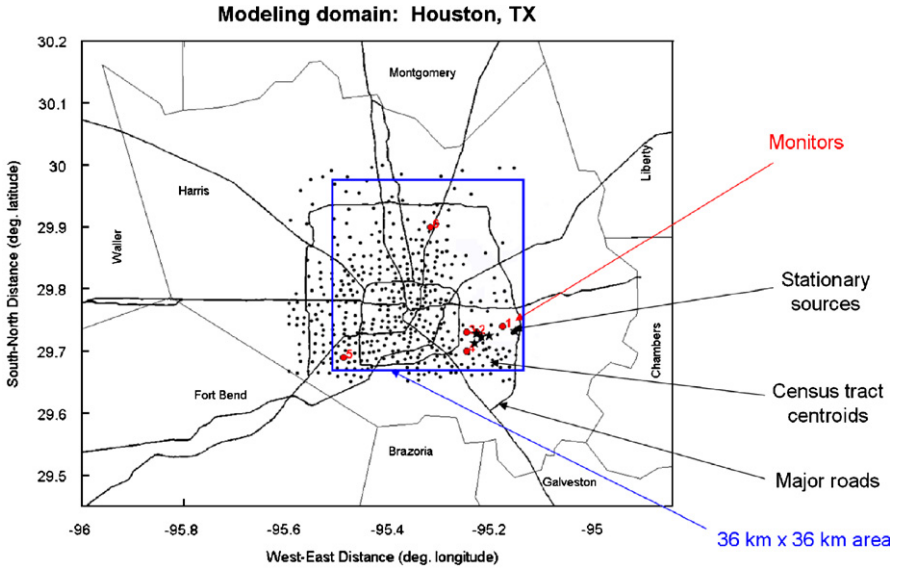

Fig. 2. Geographical location of the $36 \times 36\,km^2$ area under study (blue box). Also shown in the figure the distribution of monitors (red dots), point sources (black stars), major roads (black lines), and census tract centroids (black dots).

Table 1
Description of the characteristics of the top six point emissions in the area under study

Latitude (°)	Longitude (°)	Emission (tons year^{-1})	Description
29.712	−95.234	48.1	Petroleum refining
29.732	−95.155	9.84	Industrial organic chemicals
29.722	−95.22	7.94	Sewerage systems
29.724	−95.206	7.61	Petroleum refining
29.727	−95.231	6.1	Petroleum bulk stations and terminals
29.722	−95.254	3.78	Petroleum refining

Code (SCC) that are spatially allocated to census tracts or airports during emissions processing to further refine where the emissions are located. Emissions related to airports such as aviation gasoline distribution are allocated to airports locations. Other non-point emissions are allocated to the census tracts. The on-road inventory contains link-based emissions where the links represent road segments. Using link-based emissions allows the user to have more accurate locations of the on-road emissions as opposed to using spatial allocation to census tracts. The non-road inventory contains non-road mobile emissions at the county and SCC level that are spatially allocated to the census tracts or airports in similar fashion as the non-point emissions.

On-road mobile sources were simulated using a seasonal-hourly link-based emissions inventory. Link-based emissions have been processed using the Emissions Modeling System for Hazardous Air Pollutants (EMS-HAP) to create area sources inputs to AERMOD. The EMS-HAP Modeling System consists of a series of SAS-based programs that process emissions inventories for input into subsequent air quality modeling (US EPA, 2004c). In order to create the AERMOD input, the link-based emissions from EMS-HAP were normalized and scaled to the total benzene emissions from on-road mobile sources in Harris County according to the NEI emissions inventory.

3.2. Model setup

The CMAQ modeling system (version 4.5) was applied in a one-way nested mode for a set of domains exhibiting 36, 12, 4, and 1 km grid cell resolutions (Fig. 3). Of relevance to this effort, the regional modeling domain with a 36 km grid spacing encompassing the southern United States and a portion of the Gulf of Mexico consisted of 45×46

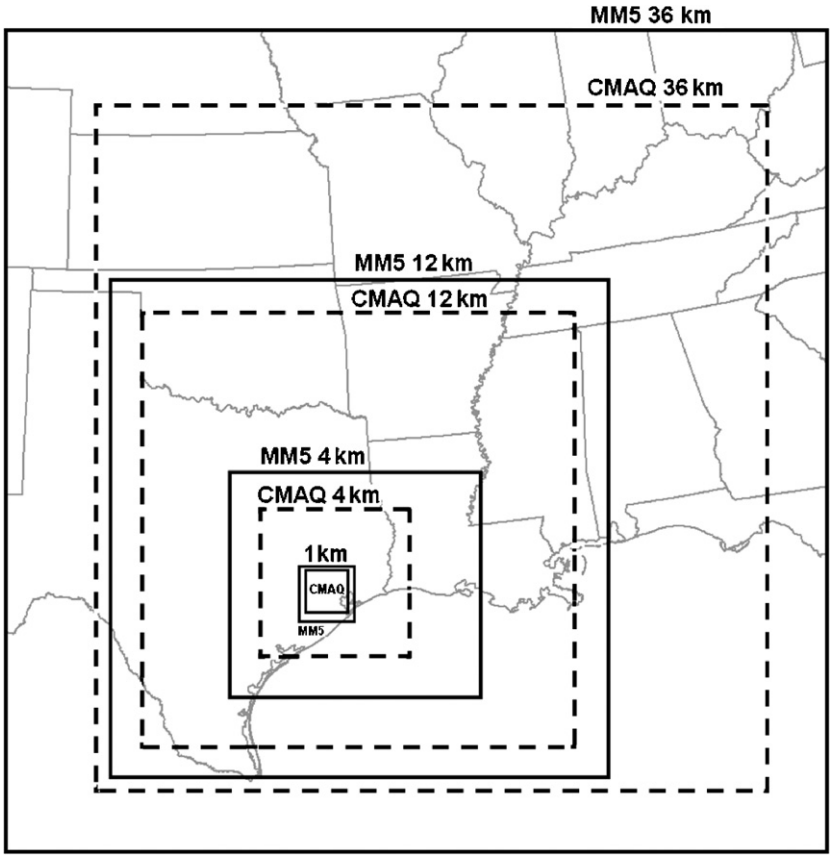

Fig. 3. Regional modeling domain showing locations of MM5 and CMAQ grids.

horizontal grid cells. In particular, a 36 km grid cell (16,14) in this domain encompassed the greater Houston metropolitan area and its gridded concentrations served as the regional background values in this hybrid approach. The model simulation of each finer nested domain was performed in a sequential manner. The initial conditions and lateral boundary concentrations for the regional domain were defined by a time-invariant set of tropospheric background values. However, boundary concentrations for each subsequent nested domain were provided by results from the immediate coarser domain. The modeling period extended from 18 August through 4 September of 2000 plus three additional days prior to this simulated period used as model spin-up. The vertical structure consisted of 24 layers extending from the surface to over 15 km on a sigma-pressure, terrain-following coordinate system.

The same CMAQ chemical transport model (CTM) executable code was used to perform the simulation for each modeling domain. It employed a modified version of the SAPRC-99 gas-phase chemical mechanism which contained additional air-toxics species, including benzene (Luecken et al., 2006). A computationally efficient Euler backward iterative (EBI) chemistry solver was customized for this set of photochemical reactions. The model was also configured to solve horizontal and vertical advection with the piece-wise parabolic method and turbulent diffusion in the horizontal and vertical with the eddy coefficient method. Moreover, aqueous-phase chemistry, cloud effects on photolysis rates, and dry and wet deposition processes were included in the calculation. The details of theoretical formulations and numerical algorithms employed to treat these physical and chemical processes incorporated in the CMAQ/CTM model are described in Byun and Schere (2006).

Meteorological fields were generated by the Penn State/NCAR fifth-generation Mesoscale Model (MM5; Grell et al., 1995). The MM5 model (version 3.6.3) was applied in a non-hydrostatic mode and included a four-dimensional data assimilation (FDDA) technique using analysis nudging on the

36 and 12 km domain applications by incorporating available observed wind, temperature, and moisture data to provide more accurate three-dimensional modeled fields. The CMAQ Meteorology-Chemistry Interface Processor (MCIP v3.1) program was exercised to extract and reformat the MM5 output from 34 levels and prepared data sets containing the hourly two-dimensional and three-dimensional meteorological parameter fields for the 24 vertical layers utilized in the CMAQ simulations. The thickness of layer 1 is about 40 m.

The three-dimensional emission data sets for CMAQ were generated by the comprehensive Sparse Matrix Operator Kernel Emissions (SMOKE version 2.2; http://www.smoke-model.org) processing system. Anthropogenic emissions from the 1999 NEI were used to generate surface area and elevated point source emissions. Natural surface emissions of NO_x, isoprene, and other biogenic VOC species were computed by the Biogenic Emissions Inventory System (BEIS version 3.13; Pierce et al., 2002). The MOBILE6 model (http://www.epa.gov/otaq/m6.htm#m60) was applied to use projections of vehicle-miles-traveled (VMT) and fleet factors to develop gridded motor vehicle emissions on each domain of the modeling period. The SMOKE point source processing programs computed plume rise with stack parameters and meteorological fields in order to allocate all point source emissions into the proper vertical layers for each domain application. Benzene emissions from the sources used in HYSPLIT and AERMOD were excluded from the Houston 36 km grid cell to avoid a double-counting effect. Fig. 4 compares the spatial distribution of daily emissions for the HYSPLIT and AERMOD models and the CMAQ model. The location of mobile sources is similar in both approaches; however they differ in magnitude. Furthermore, the emissions corresponding to the CMAQ model show a maximum in the downtown area.

The AERMOD model requires values of meteorological variables. The data is derived from a combination of a selected surface station and upper air station. The NWS meteorological station at George Bush Intercontinental Airport (IAH), located north of Houston, has been chosen as the representative station for 2000. The Lake Charles upper air station was considered the most representative of the city. The AERMOD Meteorological Preprocessor (AERMET) (US EPA, 2004a) was used to process the NWS data for 2000. The AERMET preprocessor reads surface and upper air data and creates files of meteorological variables needed by AERMOD. AERMET has three stages. The first reads the surface and upper air data files from the user and performs several quality assurance checks of the data for missing values and values considered out of range by the user. The second stage merges the surface and upper air data into one dataset while the third stage reads the merged data, calculates necessary boundary layer variables and creates the surface and profile files used in AERMOD. For a detailed discussion of the stages and boundary layer calculations, see the AERMET User's Guide (US EPA, 2004a).

The HYSPLIT model has been set to describe the emission, transport, dispersion, and deposition of benzene originated from six major point sources in the area under study. Emissions are represented by continuously releasing three-dimensional Lagrangian particles from the location of the point sources. A total of 5000 particles per hour have been released for this work. The transport and dispersion of these particles is driven by the same meteorological fields used by CMAQ as derived from the Mesoscale Model Version 5 (MM5) output. The horizontal dimensions of the concentration grid have been set equal to those of the meteorological model. Although the meteorological grid contains 24 layers, HYSPLIT's concentration grid only consists of one 100 m layer at the ground surface. Thus, transport, dispersion, and deposition processes are based on the full vertical data structure of the meteorological model while benzene concentrations are calculated only for one layer near the ground to represent the measurement data layer.

3.2.1. *HYSPLIT ensemble modeling*

The assumption behind the ensemble is that errors in the downwind plume position are primarily a function of the accumulation of initial errors in the particle trajectories. These errors are especially significant just downwind of the source, when the size of the pollutant plume is much smaller than the meteorological grid size. This is analogous to the basis of meteorological ensemble forecasts, where differences in initial conditions may lead to quite different results. Each ensemble member of the dispersion forecast is computed from the same pollutant source location, but during the calculation the meteorological grid is offset. The rationale of shifting the meteorological grid to determine initial transport errors is that the meteorological data field,

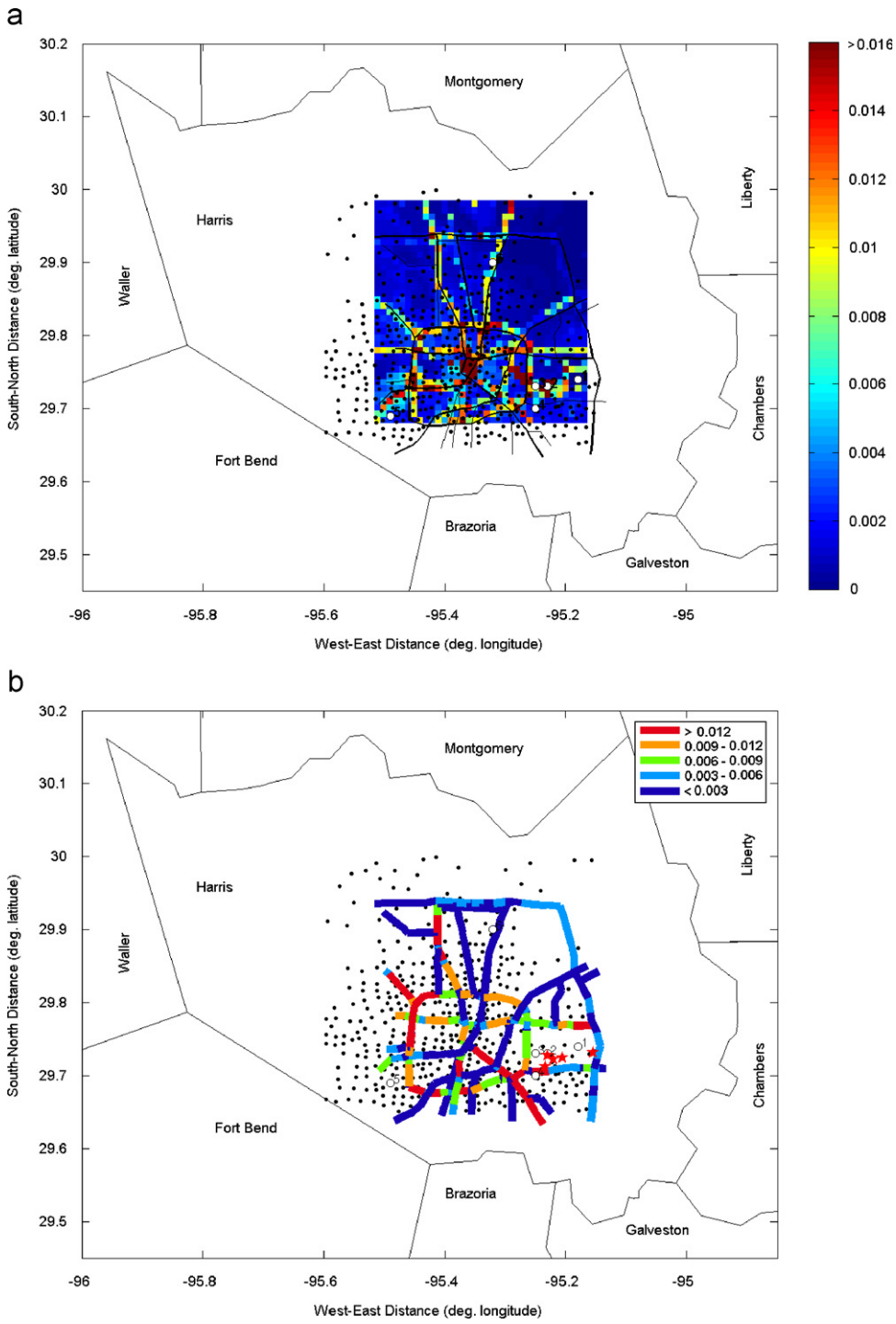

Fig. 4. Spatial distribution of benzene emissions for (a) CMAQ at $1 \times 1\,km^2$ resolution, and (b) for HYSPLIT and AERMOD in tons day^{-1}.

limited in spatial and temporal resolution, is only an approximation of the true flow field, which is continuous in space and time. Only features that are several times larger than the grid spacing are resolved by the meteorological data fields. The first series of ensemble runs created 18 members by

shifting the meteorological field. In the first nine members, the meteorological field was displaced by ± 1 grid point in the horizontal. The other nine members also had a ± 1 grid-point displacement in the horizontal, but the starting height was set to $+150$ m (approximately) from the original starting height.

The second series of ensemble runs examines the variability introduced by the turbulence by varying the initial seed of the random number generator used to simulate the dispersive component of the motion of each particle. In a normal simulation, a sufficient number of particles should be released so that the downwind concentrations are no longer sensitive to particle number. By reducing the particle number release rate (while correspondingly increasing the pollutant mass on each particle) to the point where air concentrations again become sensitive to particle number, and at the same time, changing the random seed with each simulation, insures that the results of each simulation will be different, while the ensemble average simulation will give the same concentration result as the high particle number simulation. In this study, the dispersion ensemble consisted of 27 members, each with a different seed number to calculate the random component of the particle diffusion.

4. Results and discussion

Figs. 5 and 6 show an example of the spatial distribution of benzene in the area of Houston as calculated by the hybrid approach and CMAQ only (at $1 \times 1\,km^2$ grid resolution) for two different days. The benzene concentrations as estimated by the hybrid approach closely follow the geometrical distribution of the mobile sources. Also, the concentrations from the hybrid modeling system show higher values downwind of the point sources. In general, the benzene geographical distribution estimated by the sum of the concentrations provided by the hybrid model ($36 \times 36\,km^2$ km CMAQ, HYSPLIT, and AERMOD) shows sharper gradients than those calculated with the 1 km nested CMAQ model. These modeling results illustrate the fundamental differences between the CMAQ 1 km model and the hybrid approach. In a grid model such as CMAQ, emissions get diluted into a $1 \times 1\,km^2$ grid and the transport and dispersion of the pollutants between adjacent cells takes place following the concentration gradient tending to smooth out the concentration distribution. On the other hand, Lagrangian models such as HYSPLIT or AERMOD maintain the sub-grid structure by emitting the pollutants at a point or a line conserving sharper gradients at the scales of relevance for this study.

Fig. 7 shows a time series of the modeled benzene concentrations for the six measurement stations in the area under study and the 24 h averaged measured benzene values when available. The intermittent nature of the measured time series complicates the analysis of the model performance. Also, the level of detail (temporal and spatial allocation) in the emissions inventory is not sufficient for individual stacks. The NEI inventory is self-reported and typically provides information on facility level, not for individual stacks. In this study, representative stacks were selected for each facility, as shown in Table 1. The results indicate that both the $1 \times 1\,km^2$ CMAQ and the hybrid approach reasonably compare with observations. However, the $1 \times 1\,km^2$ CMAQ approach predicts many more high transient events than the hybrid model at monitors 4 and 5. These simulated high transient events cause the $1 \times 1\,km^2$ CMAQ to overpredict the observed Benzene concentrations at monitors 4 and 5. On the other hand, the hybrid approach does not show this transient behavior giving somewhat better model performance for those monitor stations. This is mainly due to the accuracy in the geographical distribution of the emissions input used in the hybrid approach. At monitors 1 and 6 both models produce comparable results and consistently underestimate the measurements. For monitor 1, this difference can be explained by a possible wind direction bias. Also, monitor 1 is very close to the source; therefore the uncertainty in source parameters such as stack location, height, and exit velocity can explain this underestimation. For monitor 6, the underestimation might be due to the impact from a nearby source not reported in the inventory. At monitors 2 and 3, the hybrid approach seems slightly better in reproducing the frequency and range of the measured concentrations. Table 2 presents a suite of statistical measures to evaluate the model performance that includes temporally and spatially paired observed and modeled benzene concentrations. These statistical results are similar to the ones estimated in other applications (Raw Bias $1.4\,\mu g\,m^{-3}$, Normalized Raw Bias 36.64%, and Normalized Gross Error 45.53% from Luecken et al. (2006)). In general, the hybrid

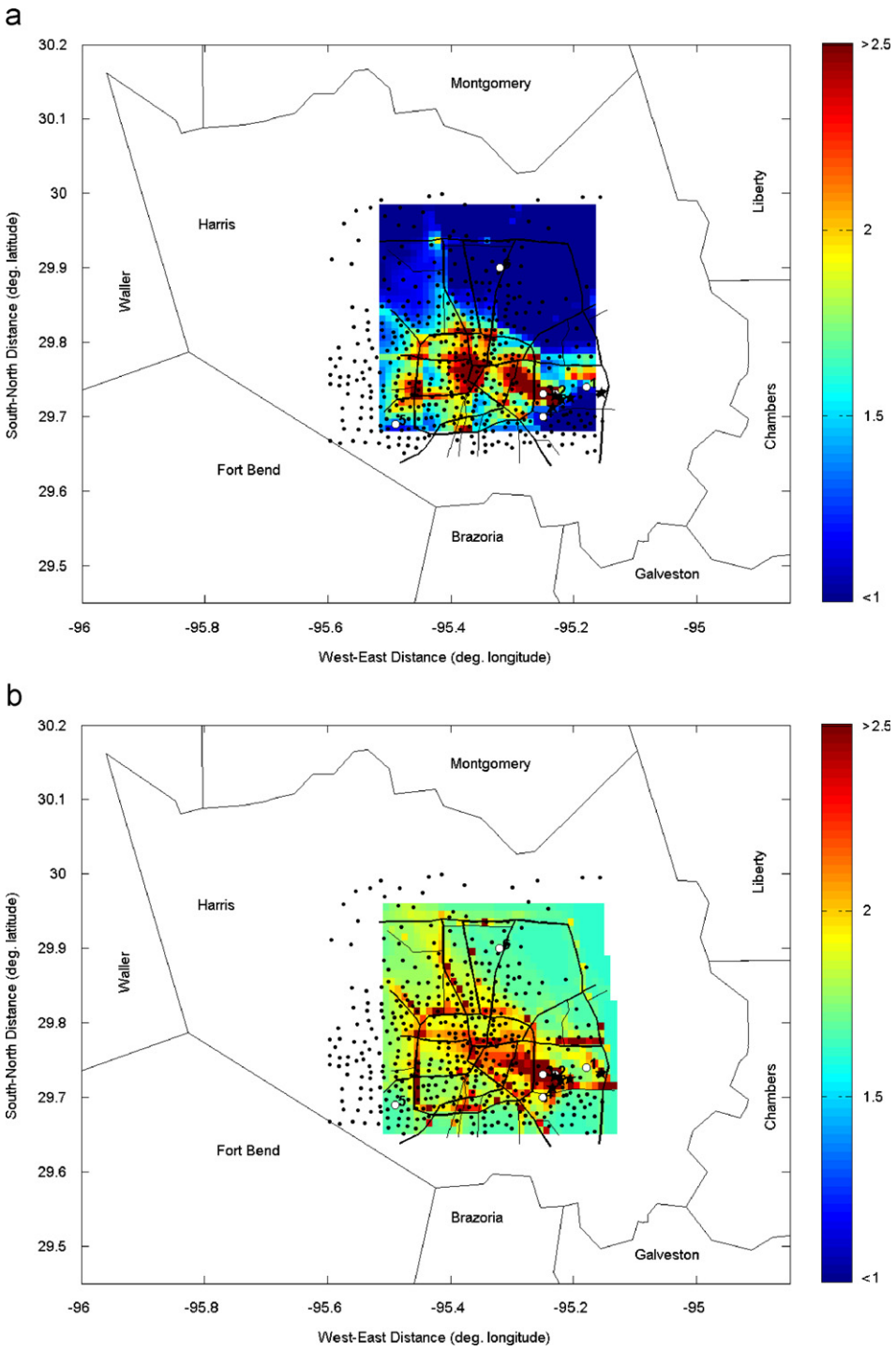

Fig. 5. Spatial distributions of 24-h average benzene concentrations on 22 August 2000 from (a) CMAQ at $1 \times 1\,km^2$ resolution, and (b) from HYSPLIT + AERMOD + CMAQ.

approach shows a slightly better agreement with the measurements than the nested $1 \times 1\,km^2$ CMAQ model output.

Combining the results from local- and regional-scale models is not straightforward. Including the same emission sources in both the regional-scale

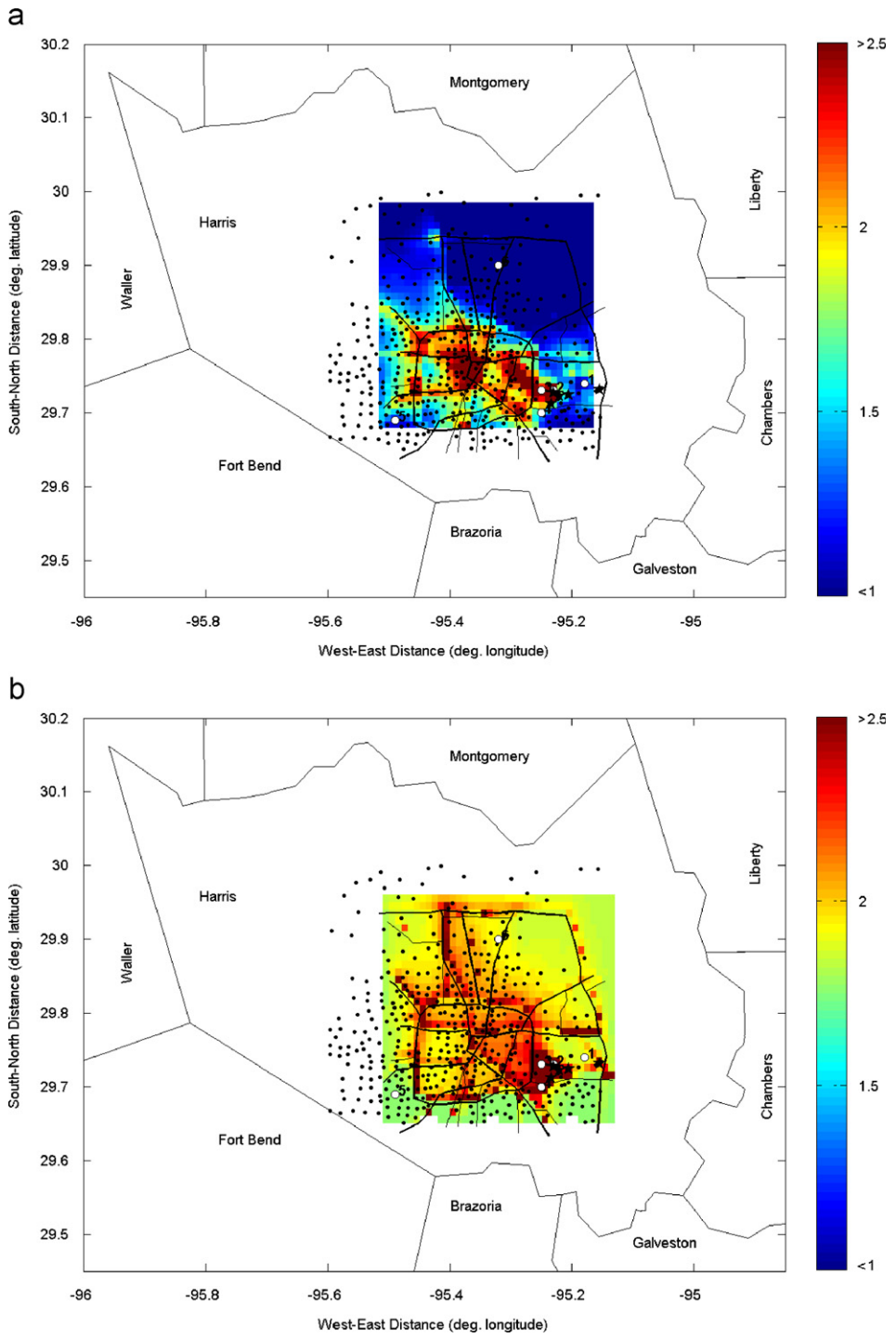

Fig. 6. Spatial distributions of 24-h average benzene concentrations on 28 August 2000 from (a) CMAQ at $1 \times 1\,km^2$ resolution, and (b) from HYSPLIT + AERMOD + CMAQ.

model and micro-scale model and adding the modeling results will 'double count' the impact of these sources. In order to avoid 'double counting', a "zero out" approach has to be used. In this study, two regional model simulations have been conducted: one for the base emission case, and another

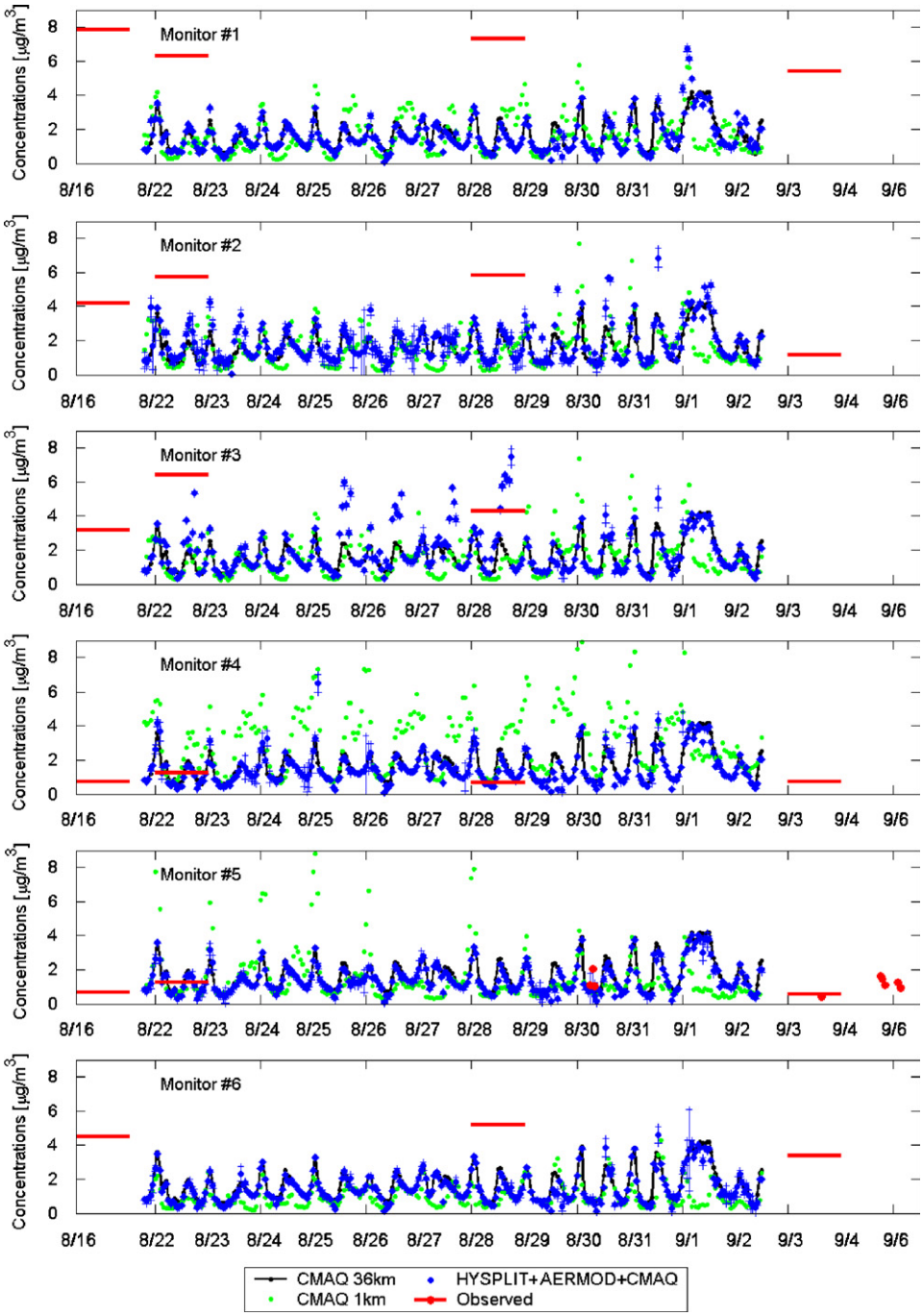

Fig. 7. Time series of benzene concentrations as calculated by the HYSPLIT + AERMOD + CMAQ($36 \times 36\,km^2$) system (blue dots), the CMAQ $1 \times 1\,km^2$ (green dots), CMAQ $36 \times 36\,km^2$ (black line), and 24-h average measurement (red line).

one excluding 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. For this case study, this difference is $<10\%$ (not shown). Therefore, the double-counting effect has no noticeable impact in this example.

Table 2
Statistical performance of the CMAQ $1 \times 1\,km^2$ and the hybrid modeling approach

	Monitor									
	1	2	3	4	5	6	Raw bias	Normalized raw bias	Raw gross error	Normalized gross error
22 August 2000										
Observed	6.3276	5.7503	6.4297	1.2778	1.2459	N/A				
CMAQ 1 km	1.2095	1.3846	1.2025	2.3702	1.8229	0.7966	2.61	21.26	3.28	73.98
Co−Cm	5.12	4.37	5.23	−1.09	−0.58					
(Co−Cm) × 100/Co	80.89	75.92	81.30	−85.49	−46.31					
|Co−Cm|	5.12	4.37	5.23	1.09	0.58					
|Co−Cm| × 100/Co	80.89	75.92	81.30	85.49	46.31					
HYSPLIT + AERMOD + CMAQ	1.5976	2.1485	2.3332	1.561	1.3527	1.3528	2.41	34.07	2.56	46.37
Co−Cm	4.73	3.60	4.10	−0.28	−0.11					
(Co−Cm) × 100/Co	74.75	62.64	63.71	−22.16	−8.57					
|Co−Cm|	4.73	3.60	4.10	0.28	0.11					
|Co−Cm| × 100/Co	74.75	62.64	63.71	22.16	8.57					
28 August 2000										
Observed	7.3482	5.8461	4.2864	0.6709	N/A	5.205				
CMAQ 1 km	1.9494	1.2204	1.1696	2.7873	1.4592	0.8902	3.07	−1.45	3.91	124.73
Co−Cm	5.40	4.63	3.12	−2.12		4.31				
(Co−Cm) × 100/Co	73.47	79.12	72.71	−315.46		82.90				
|Co−Cm|	5.40	4.63	3.12	2.12		4.31				
|Co−Cm| × 100/Co	73.47	79.12	72.71	315.46		82.90				
HYSPLIT + AERMOD + CMAQ	1.5923	2.2308	3.7788	1.8566	1.5191	1.5289	2.47	9.18	2.95	79.87
Co−Cm	5.76	3.62	0.51	−1.19		3.68				
(Co−Cm) × 100/Co	78.33	61.84	11.84	−176.73		70.63				
|Co−Cm|	5.76	3.62	0.51	1.19		3.68				
|Co−Cm| × 100/Co	78.33	61.84	11.84	176.73		70.63				

Co is the observed and Cm is the modeled benzene concentration. Raw bias, normalized raw bias, raw gross error, and normalized gross error are defined as the average of Co−Cm, (Co−Cm) × 100/Co, |Co−Cm|, and |Co−Cm| × 100/Co, respectively, over the six monitoring stations.

On the other hand, in order to gain insight on the variability of the benzene concentrations originating from point sources we take advantage of the capability of the HYSPLIT model to encompass a range of simulation outcomes. Fig. 8 shows the benzene concentration standard deviation as a function of the concentration arising from variations in the meteorological input field and in the random component of the diffusion calculation. It is interesting to notice that the standard deviation and the concentrations of benzene are of the same order of magnitude for all the ensembles but there are substantial differences in the relative contribution of each type of variation in each of the measurement sites. The ensemble created by varying the random seed number for the modeled diffusion shows the highest concentrations and the lowest relative variability (standard deviation/concentration). On the other hand, shifting the meteorological field produces lower concentrations but higher relative variability. Furthermore, the monitors with the most variability (2 and 3) were also where the model showed the best performance with respect to the monitoring data (Fig. 7). The large variability at those sites supports the difficulty in evaluating a model's performance when model predictions and measured concentrations are paired in space and time. This result contrasts the small model prediction variability shown for monitors 1 and 6 and the consistent biases shown in Fig. 7 at those sites.

5. Summary and conclusions

Local-scale modeling is necessary to resolve fine-scale variations of pollutants in investigations when local contributions are believed to be significant. Air

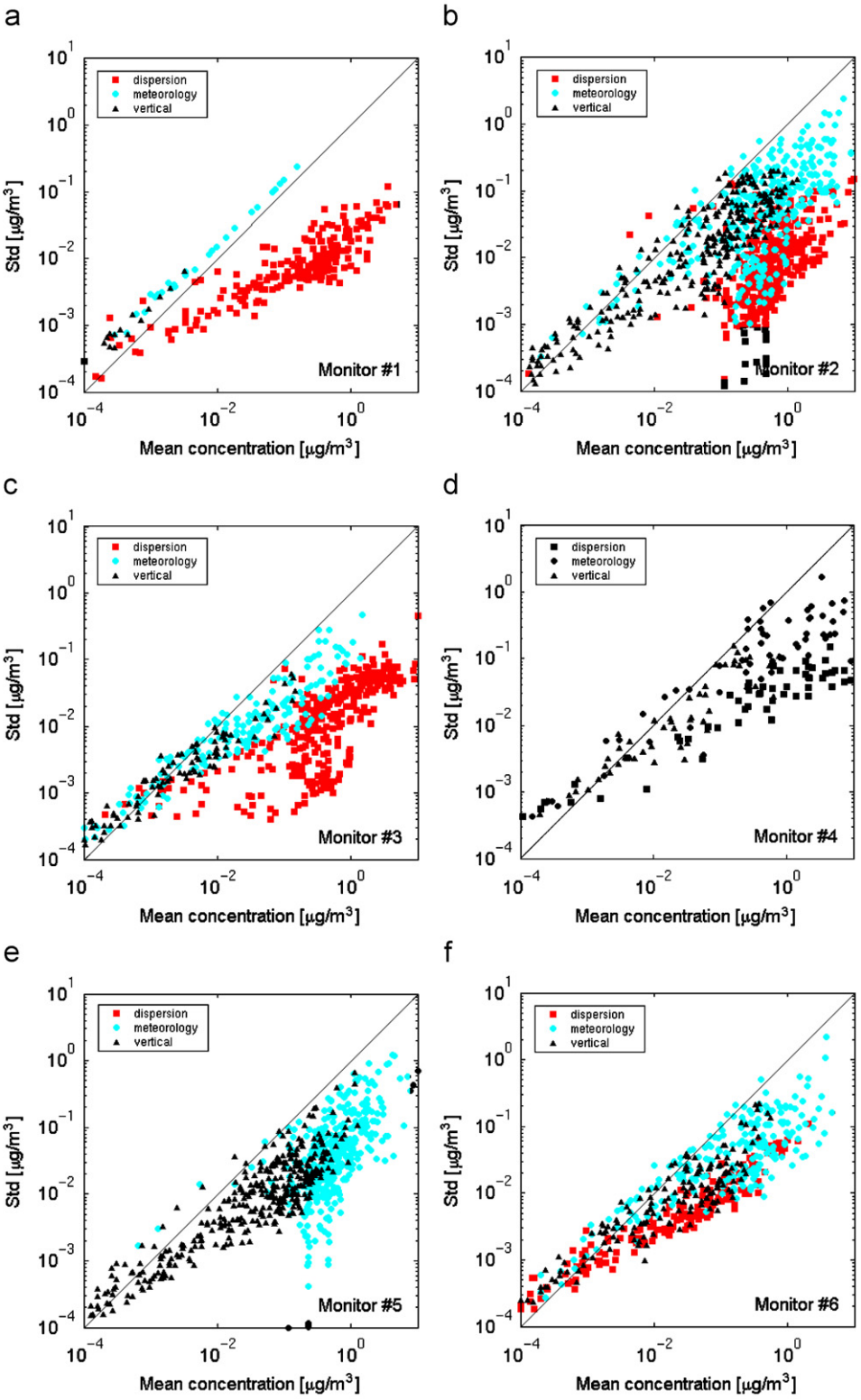

Fig. 8. Benzene standard deviation as a function of concentration from HYSPLIT ensemble runs (light-blue circles: variability due to meteorology (nine members, 1-grid horizontal shifting); red squares: variability due to dispersion (27 members, different seed random number for dispersion); black triangles: variability due to vertical structure (nine member, 1-grid horizontal shifting, increase stack release height by 150 m, approximately).

pollutants are often chemically reactive and may have background concentrations originated from distant sources. Grid models, such as CMAQ, are the best-suited tools to handle the regional features of these chemicals. However, this kind of models cannot resolve pollutant concentrations on local scales due to technical and computing time limitations. Furthermore, many species of interest have reaction time scales that are longer than the travel time across an urban area. For this kind of pollutants, chemical reactions can be ignored in describing local dispersion from strong individual sources, making Lagrangian and plume-dispersion models practical.

A hybrid modeling method to resolve fine-scale pollutant variability involving multiple air quality models has been applied for a case study to calculate the spatial and temporal distribution of benzene concentrations in the greater Houston metropolitan area. For this prototype demonstration of the hybrid approach for a relatively slow reacting pollutant, the CMAQ photochemical grid model provides the regional background concentrations and urban-scale photochemistry, the HYSPLIT model simulates the spatially resolved concentrations due to selected notable point source emissions, and the AERMOD dispersion model treats the near-surface mobile emissions from selected major roads.

Results from this model combination have been compared with CMAQ $1 \times 1\,km^2$ nested grid results and with available field measurements. The hybrid approach shows a somewhat better statistical performance than the high resolution nested Eulerian model, however, differences are evident between both modeling methods and available monitoring site data. When monitors are relatively close to emission sources as in this case, any difference between modeled and actual wind flows can cause significant spatial displacements between observed and modeled concentrations, especially for point source plume emissions. In fact, data assimilation of available observed winds was not performed in the MM5 meteorological simulations for the $1 \times 1\,km^2$ gridded domain application since this technique is not standard practice at this grid resolution. Consequently, some of the error between the modeled and observed concentration paired values is attributed to differences between modeled and observed winds at this fine scale.

Nevertheless, the modeling combination presented here explicitly treats individual source emissions and exhibits the capability to estimate concentration variability without rerunning the entire simulation system. The HYSPLIT model has been applied with different sets of initial conditions and internal physical parameters to create a concentration ensemble for selected point sources. These simulation results have showed that the standard deviation in the benzene concentration contributed by point sources is substantial and of the order of magnitude of the concentrations.

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