J1.7 IMPACT OF THE ON-ROAD AND MOBILE SOURCES ON THE BENZENE AND TOLUENE EMISSIONS AND CONCENTRATIONS IN THE HOUSTON-GALVESTON AREA

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1. INTRODUCTION

Aromatic hydrocarbons in the Houston-Galveston area are largely emitted from on-road and non-road sources as well as from some local point sources. Benzene and toluene are two aromatic species with large emissions coming from such sources, as shown in the Environmental Protection Agency (EPA)'s National Emissions Inventory from 1999 (NEI99). According to NEI99 more than 75% of the total emissions for benzene and toluene respectively are produced by mobile sources for the Houston-Galveston area (HGA). Benzene and toluene are recognized as hazardous air pollutants with serious effects on the human health, benzene being known for a long time as a human carcinogen; according to EPA, they belong to the group of Mobile Sources Air Toxics (MSAT), which contains a total of 21 toxic species.

In this paper we study the importance of the on-road and mobile (on-road and non-road) sources on the total emissions and concentrations of benzene and toluene in the HGA. Emission data was prepared with the Sparse Matrix Operator Kernel Emissions (SMOKE) system for further processing with the Community Multiscale Air Quality (CMAQ) system. For this study we used an extended State-wide Air Pollution Research Center (SAPRC-99) chemical mechanism, which allows us to simulate many aromatic species, including benzene and toluene. Carter (2005) has developed software that gave us the possibility to incorporate our detailed aromatic hydrocarbon mechanism into the SMOKE system. Using this software, we prepared the model species assignments for profiles (SMOKE gspro file) and mechanism-depended files for the CMAQ system.

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2. BACKGROUND

One of the contributors to air quality problems in the HGA is the heavy rush-hour traffic in the morning and late afternoon, when high levels of pollutants are emitted from the vehicles operating on the highways and main roads. Non-road sources, such as airplanes, boats, trains, construction equipment, are also important sources of pollutants; Diesel-powered non-road vehicles are in particular responsible for high emissions of aromatics such as benzene.

Benzene and toluene exhibit serious health effects and therefore strongly needs a better understanding of their sources and fates in HGA. It is well known that short-term inhalation of these aromatic compounds causes eye, throat and nose irritations, as well as dizziness, drowsiness and headaches, while long-term exposure of humans to such pollutants leads to problems of internal organs (heart, liver, kidney); at high levels, these compounds lead to unconsciousness, even death. EPA has classified benzene as a Group A, human carcinogen, based on the increased incidence of leukemia observed in human exposed to benzene; on the other hand, toluene was classified a Group D, with no carcinogenic effects.

For the Harris County of HGA, NEI99 shows an amount of 2249.9 tons/year emissions coming from mobile sources for benzene; for toluene the emissions from mobile sources are 5202.5 tons/year, twice as high as benzene annual emissions. (Table 1)

source	benzene	toluene
point	462.2571	565.8788
nonpoint	192.6354	1180.389
onroad	1520.865	3977.761
nonroad	729.0383	1225.78

Table 1. Emissions for benzene and toluene from NEI99 (Hazardous Air Pollutants inventory) for different source categories for the Harris County of the HGA. Emissions are in tons/year.

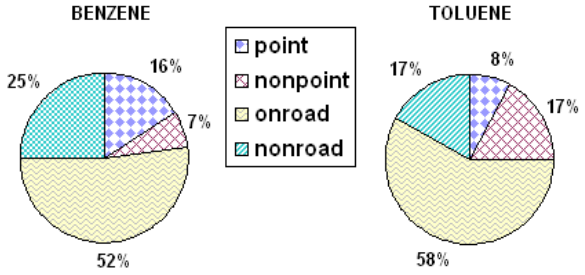


Figure 1. Emission distribution of NEI99 (HAPs) based on the source categories for benzene and toluene in the Harris County of the Houston-Galveston area.

3. METHODOLOGY

We performed simulations with an extended SAPRC-99 aromatic chemical mechanism for the 22–31 August 2000 modeling episode; the modeling domain was set up for a 4km-resolution, which covered the southeastern part of Texas, a part of Louisiana, and the Gulf of Mexico (the Houston-Galveston area).

The National Emissions Inventory from 1999 (NEI99) was processed using the SMOKE2.1 system, which is required to prepare the emission data inputs for the CMAQ4.4 system.

We performed simulations for three cases: a) all emissions sources were considered (case1); b) all sources excluding the on-road sources (case2); c) all sources excluding mobile sources (case3).

The study was done in five areas with different emissions and environmental conditions within the HGA: center of urban, downwind of urban, center of industry, downwind of industry and rural. (Figure 2) For each area of interest, we averaged the emissions and concentrations for each case (case1, case2 and case3) over the number of cells within each small domain.

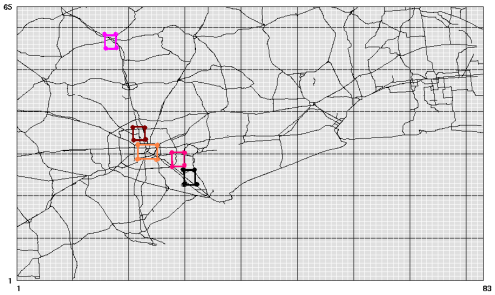


Figure 2. Location of the study areas within the Houston-Galveston 4km domain (83x65 cells): center on urban (orange), downwind of urban (brown), center of industry (black), downwind of industry (pink), rural (violet). The roads and highways are drawn with gray lines.

4. RESULTS AND DISCUSSION

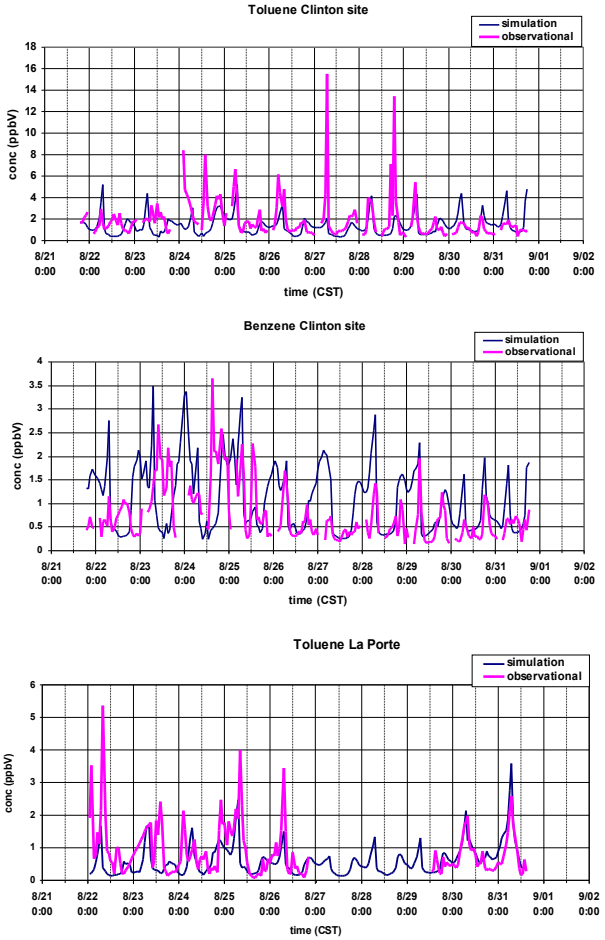
To better understand the impact of the on-road and mobile sources we analyzed the absolute and relative differences among the three simulated cases.

The absolute and relative differences were calculated according to the following equations:

AbsDiff = caseA - caseB (moles/sec for SMOKE; ppmV for CMAQ);
RelDiff = (caseA - caseB)/caseA,

where A is 1 (all sources considered in the simulation) and B is either 2 (all sources considered except for the on-road sources), or 3 (all sources considered except for the mobile sources).

In Figure 3 we present the comparison of the simulated concentrations for benzene and toluene, when all the sources were considered, with the observational data from two surface sites of the Houston-Galveston area (Clinton and La Porte)



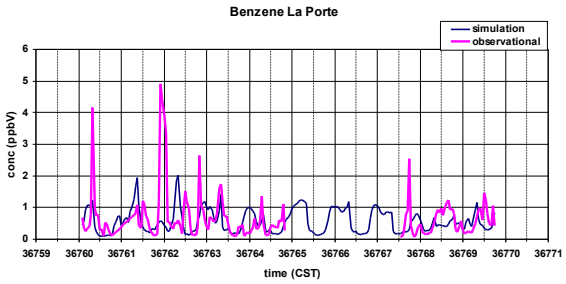


Figure 3. Comparison of the simulated benzene and toluene concentrations with the observational data of the Clinton and La Porte sites.

4.1. SMOKE2.1 results

The analysis done using the SMOKE results showed that the highest onroad/mobile emissions were located in the urban areas, with peaks in the late afternoon corresponding to the rush hour time (4pm CST). The rural areas were characterized by the smallest emission values compared to those observed in the industrial and urban areas. As expected, the toluene SMOKE results showed higher emissions than for benzene species, the peak value for the urban area being twice as high as the benzene peak observed at the same moment of the day. (Figure 4)

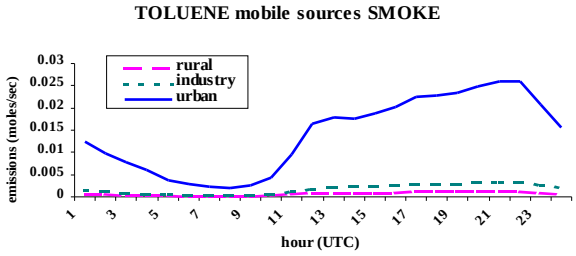


Figure 4. Toluene emissions from mobile sources for different areas: urban, rural and industry. (Time: UTC = CST+6 hours)

The contributions of the onroad/mobile sources to the total benzene emissions (Figure 5) were almost the same for the urban and rural areas, with smaller values during the night (minimum: ~50% for onroad sources and ~65% for mobile sources) and higher values during the day (70-90%). In the industrial area, the onroad/mobile contribution was high during the day (~40-60%) and very small during the night, when emissions from other sources such as point sources (refineries) were more important.

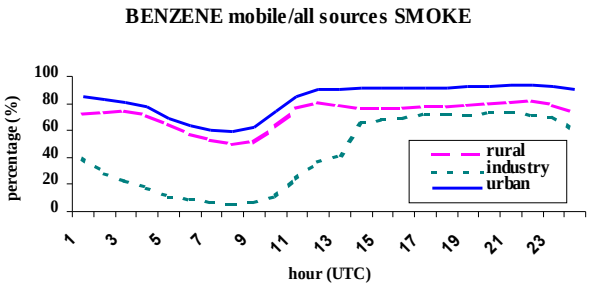


Figure 5. Mobile sources emissions contribution for the benzene species for the urban, rural and industrial areas. (Time: UTC = CST+6 hours)

For toluene, the contribution of onroad/mobile sources is almost the same for the three different areas. (Figure 6)

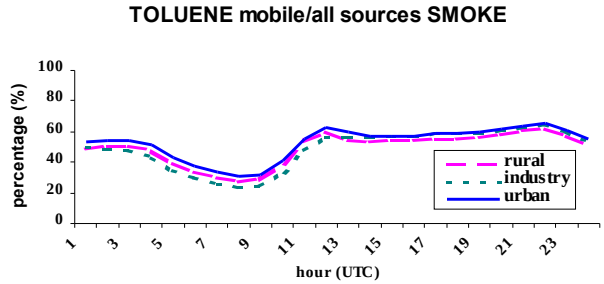


Figure 6. Mobile sources emissions contribution for the toluene species for the urban, rural and industrial areas. (Time: UTC = CST+6 hours)

4.2. CMAQ4.4 results

According to the CMAQ results, the simulated benzene and toluene concentrations were high in the morning, with a peak at 7am (CST), due to the low boundary layer and absence of the OH radical chemistry at this time of the day. The concentration decreased during the day when the boundary layer heights were higher and both benzene and toluene are consumed by the hydroxyl radicals, with a minimum concentration value observed in the late afternoon. The highest peak value was observed for the urban area, corresponding to the highest onroad/mobile emissions area. The rural area analysis showed higher concentrations during the night decreasing in the morning and afternoon, winds being responsible for the transport of the emissions/concentrations from the urban and industrial areas to the rural areas, which were

accumulated on top of the local nighttime emissions/concentrations, leading to the higher values than those obtained in the afternoon due to local emissions.

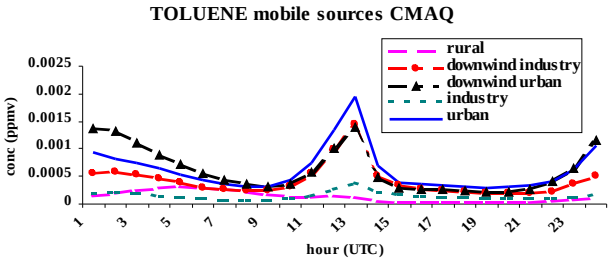


Figure 7. Toluene concentrations coming from the mobile sources emissions for five areas in the Houston-Galveston area: urban, downwind of urban, industry, downwind of industry and rural. (Time: UTC = CST+6 hours)

The five areas (urban, downwind of urban, industry, downwind of industry and rural) with distinctive emissions and environmental conditions were characterized by different contributions of the onroad/mobile concentrations to the total benzene concentrations. Industrial and downwind of industry areas showed lower onroad/mobile contributions than for the other areas, with large variations during the day: from 10%, minimum observed early in the morning, to almost 50%, in the late afternoon. Rural areas were characterized by smaller variations of the onroad/mobile contribution over the day, while urban and downwind of urban areas presented higher contributions of the onroad/mobile sources, with smaller contribution during the night-early morning hours and almost constant contribution during the afternoon (Figure 8).

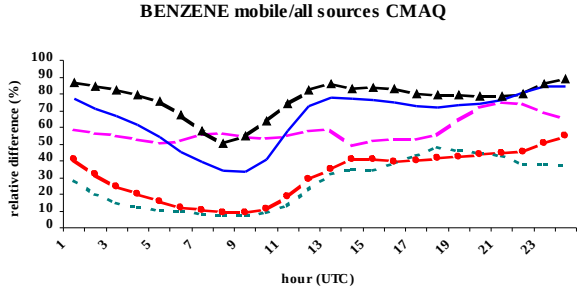


Figure 8. The mobile source contribution to the benzene concentrations for five areas: urban, downwind of urban industry, downwind of industry and rural. (Time: UTC = CST+6 hours)

For the toluene case it was observed that onroad and mobile sources had similar contribution profiles for all areas analyzed, except for the rural area as described by a constant on-road/mobile sources contribution over the day.

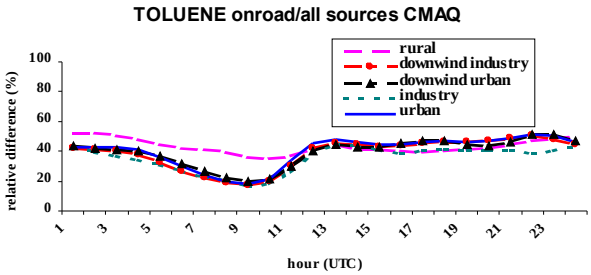


Figure 9. The on-road sources contribution to the benzene concentrations for five areas: urban, downwind of urban, industry, downwind of industry and rural. (Time: UTC = CST+6 hours)

Although the toluene emissions coming from on-road and mobile sources were higher than the benzene emissions, the higher contributions of such sources to the total emissions and concentrations were observed for benzene (Table 2, Table 3, Table 4)

	benzene			toluene		
source type	urban	rural	industry	urban	rural	industry
onroad sources	50-80%	50-70%	5-60%	20-50%	30-50%	20-50%
mobile sources	70-90%	60-80%	5-65%	40-65%	40-60%	35-60%

Table 2. The on-road and mobile sources contributions to the total benzene and toluene emissions for the urban, industry and rural areas, over a 24-hour period.

	benzene				
source type	urban	downwind of urban	rural	industry	downwind of industry
onroad sources	35-75%	40-80%	40-70%	10-30%	10-40%
mobile sources	40-80%	60-90%	55-80%	15-50%	15-50%

Table 3. The on-road and mobile sources contributions to the total benzene concentrations for five areas within the modeling domain, over a 24-hour period.

	toluene				
source type	urban	downwind of urban	rural	industry	downwind of industry
onroad sources	25-50%	25-50%	40-50%	25-45%	25-50%
mobile sources	40-60%	40-60%	50-60%	35-60%	35-60%

Table 4. The on-road and mobile sources contributions to the total toluene concentrations for five areas within the modeling domain, over a 24-hour period.

5. CONCLUSIONS

Benzene and toluene are two aromatic species with high annual emissions coming from on-road (almost 50%) and mobile sources (almost 75%), as presented in the National Emissions Inventory of the 1999-year. The largest emissions were observed in the late afternoon, corresponding to the afternoon rush hour time (4pm CST); the highest benzene and toluene concentrations occurred early in the morning, around 7am (CST), when large emissions were emitted due to the morning rush hour. In addition the low boundary layer and a lack of chemistry processes due to the absence of the OH radicals favor the accumulation of both aromatics.

The study done in the five areas with different emissions and environmental conditions showed that for benzene and toluene, on-road/mobile sources were more important in the urban and rural areas than in the industrial area; for benzene, the industrial area is defined by very small contribution from such sources during the night and early morning, and higher contribution in the afternoon, but still smaller than the contributions observed in the urban and rural areas. Toluene exhibited similar contributions in the analyzed areas.

Urban areas were characterized by the largest benzene and toluene emissions and concentrations and by the highest on-road/mobile sources contributions to the total benzene and toluene emissions and concentrations.

The contribution profiles for benzene varied significantly among the analyzed areas, while for toluene, they were in the same range, except for the rural area, which is characterized by almost a constant contribution profile.

6. ACKNOWLEDGEMENT

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