Diesel Particulate Matter Modeling and Inhalation Cancer Risk in Nashville, Tennessee, Using Models-3/CMAQ

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ABSTRACT

The fine and ultra fine sizes of diesel particulate matter (DPM) are of greatest health concern, which significantly contributes to the overall cancer risk from air toxics. The composition of these fine and ultra fine particles is composed principally by elemental carbon (EC) with adsorbed compounds such as VOCs, sulfate, nitrate, ammonia, metals, and other trace elements. The purpose of this research was to use EPA's Models3/CMAQ version 4.3 to predict diesel aerosol concentrations and inhalation cancer risk that come from diesel fueled sources (DFS) in the Nashville urban area, Tennessee, by linking the MM5v3 meteorological model, the Sparse Matrix Operator Kernel Emissions (SMOKE 2.0) model. The national emissions inventory version 3 for the year 1999 (NEI99) was used in this analysis for point, area, and non-road, whereas NMIM was used to create the on-road emissions. The year 2003 was used for meteorological data and as reference to compare the monitored concentrations and model performance. The modeling domain consisted of a 36 km domain and the area of analysis was Nashville, TN. Five emissions scenarios were selected in the domain to compare the main results; base case without DFS, base case without on-road DFS, base case without light-duty diesel vehicles (LDDVs), base case without heavy duty diesel vehicles (HDDVs), and the scenario considering the in effect regulations on mobile sources for the year 2020. The DPM inhalation cancer risk was 163.6 in one million of population, which generated the higher lifetime cancer risk excess among the monitored air toxics acetaldehyde, benzene, 1,3-butadiene, and formaldehyde in Nashville, posing a cancer risk that was 4 times higher than the combined total cancer risk from all those air toxics. The main DPM reductions on concentrations and cancer risk were due non-road DFS (52.7 % in cancer risk). For on-road sources, the main DPM contribution was due HDDVs sources, which accounted for 41.6% in cancer risk reduction. The DPM reduction in 2020 due to on-road sources was 38.1%, which is not enough of a health point of view, since the cancer risk reductions were as low as 32.7 % for Nashville.

Acknowledgment: Acknowledgments of NCAR's assistance in this project.

INTRODUCTION

Diesel particulate matter (DPM) is currently a topic of great concern from both pollution and public health standpoints. The U.S. EPA finalized strict new regulations on diesel particle emissions. Although specific output depends on operating conditions, the largest single component of DPM emissions is carbonaceous soot produced by the incomplete combustion of diesel fuel. A great research effort is currently being devoted to reducing the amount of DPM emissions. These efforts include reducing the diesel fuel sulfur content, making the diesel engine combustion process more efficient, as well as removing particles from the exhaust stream (Burtscher, 2005; U.S. EPA, 2001 and 2004). DPM is part of a complex mixture. The sizes of diesel particulates, which are of greatest health concern, are in the categories of fine, ultra fine, and nano particles (Biswas and Wu, 2005; Lloyd and Cackette 2001; Lighty et al., 2000; Kittelson, 1998), as shown in Figure 1. The mixture of these particles is composed mainly of elemental carbon (EC) with adsorbed compounds, such as organic carbon (OC), sulfate, nitrate, metals, and other trace elements (Kleeman et al., 2000; Kittelson, 1998). The elemental fraction stems from fuel droplet pyrolysis, while the organic fraction originates from unburned fuel, lubricating oil, and combustion byproducts (Shah et al., 2004).

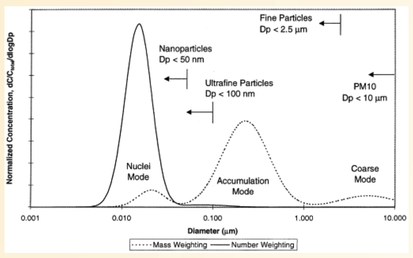

Figure 1. Typical engine exhaust size distribution: both in mass and in number. (Reprinted from Kittelson et al., 1998)

Many carcinogenic and mutagenic compounds have been measured in the DPM, such as polycyclic aromatic hydrocarbons (PAHs) and nitroarenes, as well as irritants or inflammatory agents such as acrolein, among others (Jasco Inc, 2004; Kittelson, 1998; Rosenkranz, 1996). A diesel particle initially consists of an agglomeration of EC spheres coated with organic and inorganic compounds that are adsorbed or absorbed at the surface of this agglomerate (Vioutsis et al., 2005; Kim et al., 2002a, 2002b; Kittelson, 1998). It may be noted that for some time, the diesel exhaust nano-particles are formed in part due to the continuation of in-stack coagulation and adsorption, along with the condensation of significant quantities of organic and inorganic compounds present in diesel exhaust (Biswas and Wu, 2005).

The composition and emissions quantity of DPM are strongly dependent of the diesel fuel sulfur content, since the lowest sulfur concentration is in the fuel, the lowest will be the particulate matter emissions from the diesel engines (Saiyasitpanich et al., 2005; Liang et al., 2005; U.S. EPA, 2001 and 2004). For this reason, in January 2001 and in June 2004, the U.S. EPA finalized the Clean Diesel Trucks and Buses Rule and the Clean Non-road Diesel Rule, respectively, with more stringent standards for new diesel engines and fuels. The rules require the use of sulfur content as low as 15 ppm beginning in 2006 for highway diesel fuel, and 2007 for non-road diesel fuel. These fuels will enable the use of after treatment technologies for new diesel engines, which can reduce harmful emissions by 90 percent or more. After treatment technologies will start phasing into the diesel sector beginning in 2007 for highway and 2011 for non-road (U.S. EPA 2001 and 2004).

The smallest DPM can deeply penetrate in the lungs and enter to the blood stream, carrying in the toxins to the rest of the body (Nemmar et al., 2002; Donaldson et al., 2001) where they can affect the respiratory and cardiovascular systems, and other organs, causing a high number of premature deaths (Delfino et al., 2005; Zanobetti and Schwartz, 2005; Sioutas et al., 2005; Schultz et al., 2005; Riedl et al., 2005; Pope et al., 2004) The fine particles aggravate the cardiovascular diseases increasing heart and brain attacks, since they invade the blood stream and start an inflammatory response, interrupting the heart beats and increasing the sanguineous coagulation (Riedl et al., 2005; Delfino et al., 2005; Peters, 2001 and 2002) On the other hand, a consistent and evident relationship between the exposition to the DPM and lung cancer has been published in more than thirty two epidemiological studies in humans (Krewski et al., 2005; CalEPA, 1998; Lipsett and Campleman, 1999) In 1989, the International Agency for Research on Cancer (IARC) concluded that DPM is a probable human carcinogen, classifying it in the group 2A. In 1990, California through its Environmental Protection Agency (CalEPA) identified the DPM as a chemical that caused cancer (CalEPA, 2005) and defined an inhalation cancer risk factor of $3x10^{-4}$ [m^3/ug]. High morbidity and mortality levels of respiratory diseases have been reported in communities with high DPM concentrations as well (Krewski et al., 2005; Conrad et al., 2005; Fruin et al., 2004; Adonis et al., 2003a).

In order to better manage air quality, it is important to know the sources or source categories that contribute to the concentrations of DPM at a particular area or receptor. The receptor models have been used widely to characterize the sources that contribute to specific pollutants, such as PM2.5 and PM10 (Zheng et al., 2002; Kavouras et al., 2001; Koutrakis et al., 2005) However, they do not fully take into account the chemical reactions involved in the formation of secondary fine particles nor distinguish among a heavy diesel engine vehicle, a light diesel engine vehicle, or a diesel non-road engine (U.S. National Research Council, 1999).

METHODOLOGY

The methodology explained in this section has the objective of giving a structure to the development of an analytical protocol to assess the health risk of emission scenarios and of regulatory actions over future on-road sources scenarios to reduce DPM concentrations. The overall approach included running CMAQ version 4.3 model over the base case (BC) with and without the following sources categories: diesel fueled sources (DFS), on-road DFS, heavy-duty diesel vehicles (HDDVs), as well as a future 2020 year with and without DFS containing the in effect mobile sources regulations (Table 1). The year 2020 was selected as a future scenario to compare the projections estimated by the U.S. EPA in its study Analysis of the Impacts of Control Programs on Motor Vehicle Toxic Emissions and Exposure Nationwide (U.S. EPA, 1999a and 1999b).

Table 1. Emissions Scenarios run in SMOKE2.0 and CMAQ 4.3

Emission Scenario	Name	Objective
All Sources Present (Base Case)	BC	Base Case
Base Case Without DFS	NO DFS	Contribution of DFS to DPM and health risk
Base Case Without On-Road DFS	NO ONROAD_DFS	Contribution of on-road DFS to DPM exposure and health risk
Base Case Without HDDVs	NO HDDVS	Contribution of HDDVs sources to DPM exposure and health risk
In effect MSATs regulation for 2020	YEAR 2020	Contribution of in effect MSATs regulations to the 2020 BC
In effect MSATs regulation for 2020 without DFS	YEAR 2020_NODFS	Contribution of in effect MSATs regulations to DPM exposure and health risk

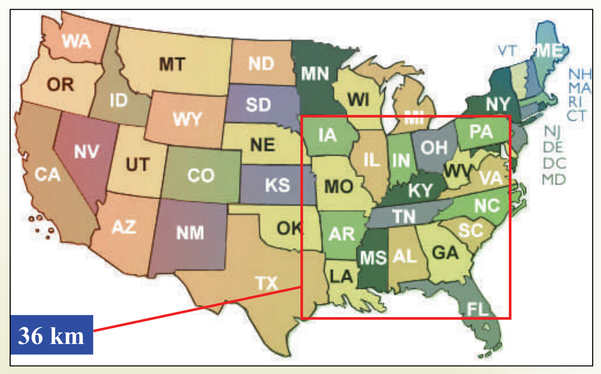

Figure 2. 36-km Modeling Domain

The protocol developed here allows the estimation of the cancer risk, by taking into account the modeled annual DPM concentrations obtained as output of the Models-3/CMAQ and linked with toxicological cancer risk equations. With those components, the cancer risk expected in a particular area of the modeling domain can be estimated and compared for different emission scenarios. It may be noted that Models-3/CMAQ was assumed as the inhalation exposure model, i.e., the population is exposed to the outdoor ambient DPM concentrations without taking into account for the time that people spend indoors and outdoors. In addition, the risk assessment was designed to be a picture for measuring progress in reducing risks from exposure to DPM. For this reason, this study is based on a 1999 inventory of criteria pollutants emissions. It then assumes individuals spend their entire lifetimes (70 years) exposed to that DPM. Therefore, it does not account for the reductions in emissions that have occurred since 1999 or those that will happen in the near future due to new regulations for mobile and industrial sources.

These assumptions could be considered satisfactory to do emission scenarios analysis for pollutants, since the analysis approach intended by this proposed modeling involves considering the difference in mass concentrations and health risk values among the proposed emission scenarios as compared to the base case scenario rather than the absolute mass concentration or health risk values. This assumes that the factors that contributed to the under and over prediction of those DPM concentrations would contribute similarly in all the scenarios considered in the analysis, causing minimal effects on the differences among the scenarios.

The 2003 daily and annual ground level diesel aerosol concentrations were predicted using CMAQ version 4.3 through a 36-km modeling domain (Figure 2) and compared with available 2003 monitored data from Nashville, TN, base on monthly concentrations of March, June, September, and December of 2003, starting 5 days before the 1st of each month to avoid the initial conditions effects on the CMAQ runs. Therefore, it was assumed that each month represented a season; March represented spring, June represented summer, September represented fall, and December represented winter season. Those area, point, on-road, non-road, and biogenic emissions were temporal and spatially allocated using SMOKE version 2.0. On-road sources were predicted using National Mobile Inventory Model (NMIM) for the whole modeling domain, whereas the 1999 UTK emission inventory and the NEI99 were used for point, area, and non-road sources for Tennessee and for the rest of the 23 states in the modeling domain, respectively. For the year 2020, all the in effect on-road sources regulations were applied on NMIM, including the reformulated gasoline (RFG) program, national low emission vehicle (NLEV) program, emissions standards for passenger vehicles and gasoline sulfur control requirements (Tier 2), and the 2007 heavy-duty vehicle standards and highway diesel fuel sulfur control requirements, which are expected to yield significant reductions of mobile source air toxics (U.S. EPA, 2001). A future 2020 scenario for non-road, area, and point sources were not simulated since no activity growth data were available for the 24 states by the time when this study was conducted, as well as the in effect non-road regulations on diesel engines were not available in the model NMIM or NONROAD by the time when this analysis was conducted.

The meteorological variables were generated for those 4 months of 2003 through the mesoscale model (MM5) version 3.7 (PSU/NCAR, 2005) and processed by the meteorology-chemistry interface processor (MCIP) version 2.2. This general conceptual model is shown in Figure 3. Since the EC is the main component of the DPM, the modeling performance was done over monitored EC data measured in Nashville, for 2003.

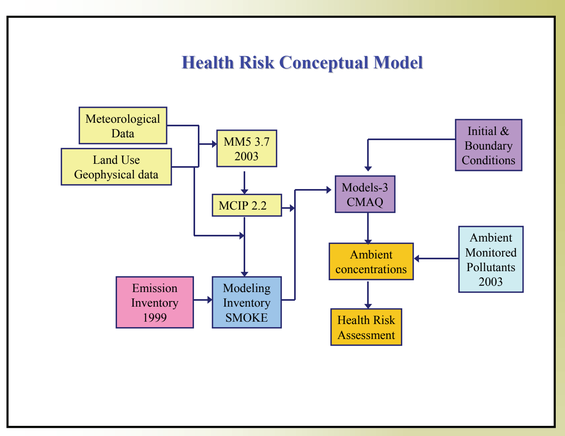

Figure 3. Health Risk Conceptual Model

Because those DPM concentrations were assumed primary PM2.5, they were estimated considering the Equation 2-5 for the following primary PM2.5 Aitken and accumulation mode species defined in CMAQ 4..3: AECI, ACEJ, A25I, A25J, AORGPAI, and AORGPAJ. The equation used to estimate primary DPM was:

DPM= 1.167*(AORGPAI + AORGPAJ) + AECI + AECJ + A25I + A25J Equation 1

Thus, the difference between the BC scenario and the NO DFS scenario were the DPM concentrations for the base case run, whereas the difference between the scenario YEAR 2020 and the YEAR 2020_DFS scenario were the DPM concentrations for the year 2020. The difference between the NO ONROAD_DFS scenario and the NO DFS scenario were the DPM concentrations contributed by the on-road sources. Whereas, the difference between the NO HDDVs scenario and the NO DFS scenario were the DPM concentrations contributed by the HDVs. Finally, the Equation 2 allows estimating the DPM concentrations contribution by the LDVs.

DPM (LDDVs) = [BC - (NO HDDVs-NO ONROAD_DFS)] - NO DFS Equation 2

The toxicity values for cancer risk were calculated using the unit risk values of $3x10^{-4}$ [m^3/ug]. The annual mean DPM concentration from a location for each analyzed scenario was multiplied by its unit risk to produce the individual cancer risk (Equation 3). DPM with cancer risks greater than 1×10^{-6} were considered a potential human health concern (U.S. EPA, 2005a).

$$\text{Cancer Risk} = C_i \left[\frac{ug}{m^3}\right] x\ IUR_i \left[\frac{m^3}{ug}\right] \qquad \text{Equation 3}$$

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RESULTS

Elemental Carbon Emissions

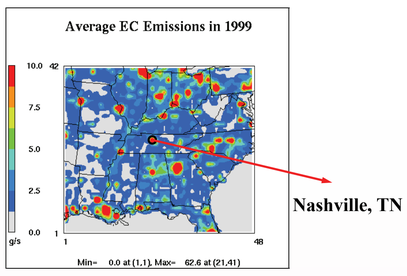


Figure 4. Average EC Emissions in 1999

MM5 and CMAQ Modeling Performance

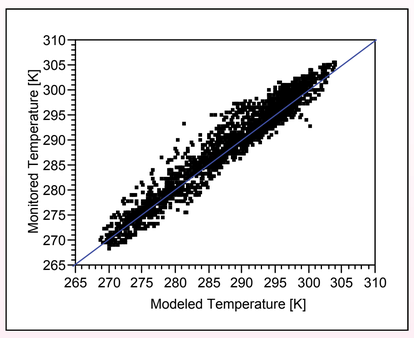


Figure 5. MM5 3.7 Modeling Performance on Temperature

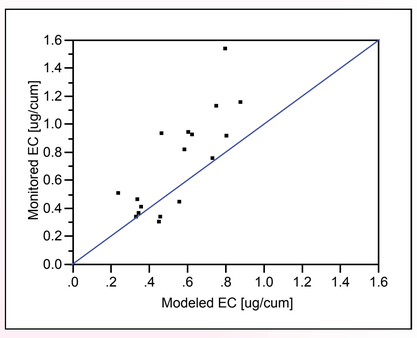


Figure 6. CMAQ 4.3 Modeling Performance on EC

Table 2. Comparison of hourly temperature measured at the Nashville International Airport, TN, to MM5 (layer 1).

# Obs	T [K]		SDEV [K]		Normalized Bias [%]	Normalized Gross Error [%]	R^2
	MM5	Obs	MM5	Obs			
2497	288.3	288.8	8.6	8.9	-0.19	0.55	0.945

Table 3. Comparison of daily EC concentrations measured in Nashville, TN, to CMAQ (layer 1).

# Obs	EC [ug/cum]		SDEV [ug/cum]		Normalized Bias [%]	Normalized Gross Error [%]	R^2
	CMAQ	Obs	CMAQ	Obs			
17	0.55	0.72	0.2	0.36	-13	28	0.65

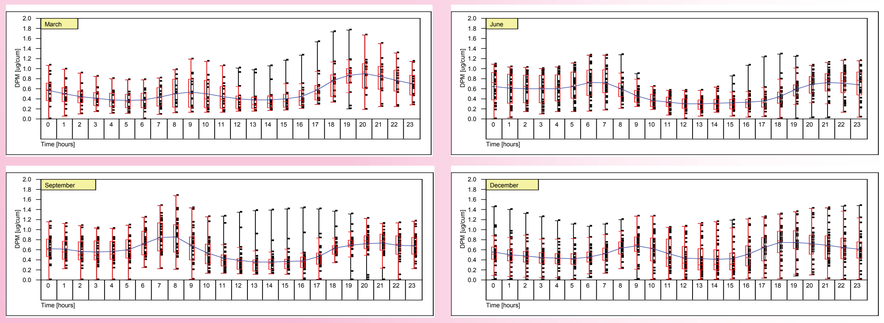


Figure 7. Box-Plots of the Modeled Hourly Average DPM Concentration at Nashville, TN, (March, June, September, and December 2003)

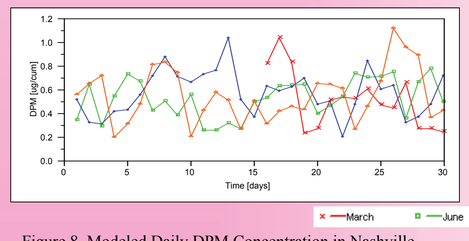


Figure 8. Modeled Daily DPM Concentration in Nashville

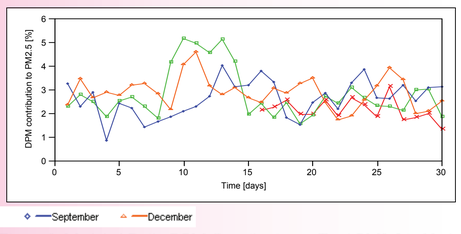


Figure 9. Modeled Daily DPM Contribution to Total PM2.5 in Nashville

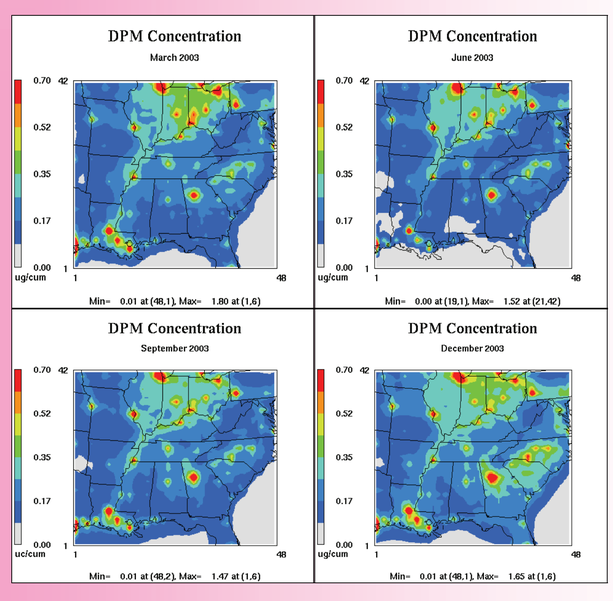
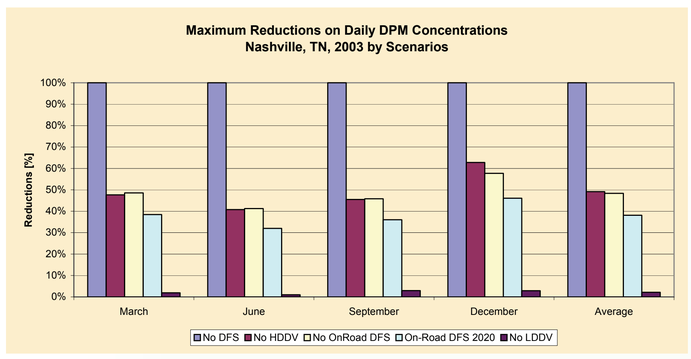


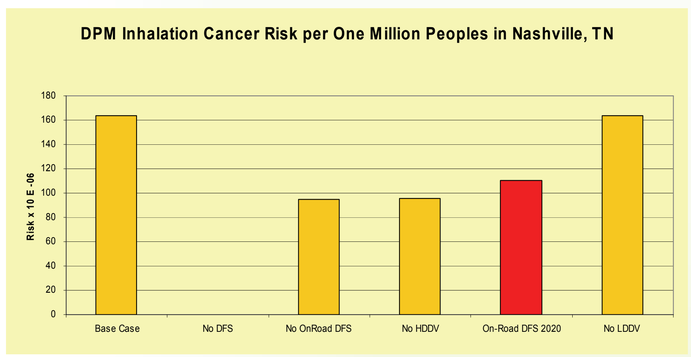
Figure 10. Modeled Monthly DPM Concentrations on the 36-km Modeling Domain

DPM Concentrations by Emissions Scenarios



DPM Scenarios	AV. Reductions
No DFS	100.0%
No HDDV	49.2%
No Onroad DFS	48.3%
On-Road DFS 2020	38.1%
No LDDV	2.2%

Figure 11. Maximum Reductions on Daily DPM Concentrations Nashville, TN, 2003 by Scenarios



Scenario	DPM x 1E-06	Reductions [%]
Base Case	163.6	
No DFS	0.0	100.0%
No Onroad DFS	94.7	42.1%
No HDDV	95.6	41.6%
Onroad DFS 2020	**110.0**	**32.7%**
No LDDV	163.5	0.01%

Figure 12. DPM Inhalation Cancer Risk per One Million Peoples in Nashville, TN, by DFS Scenarios

Spatial Distribution on the Domain

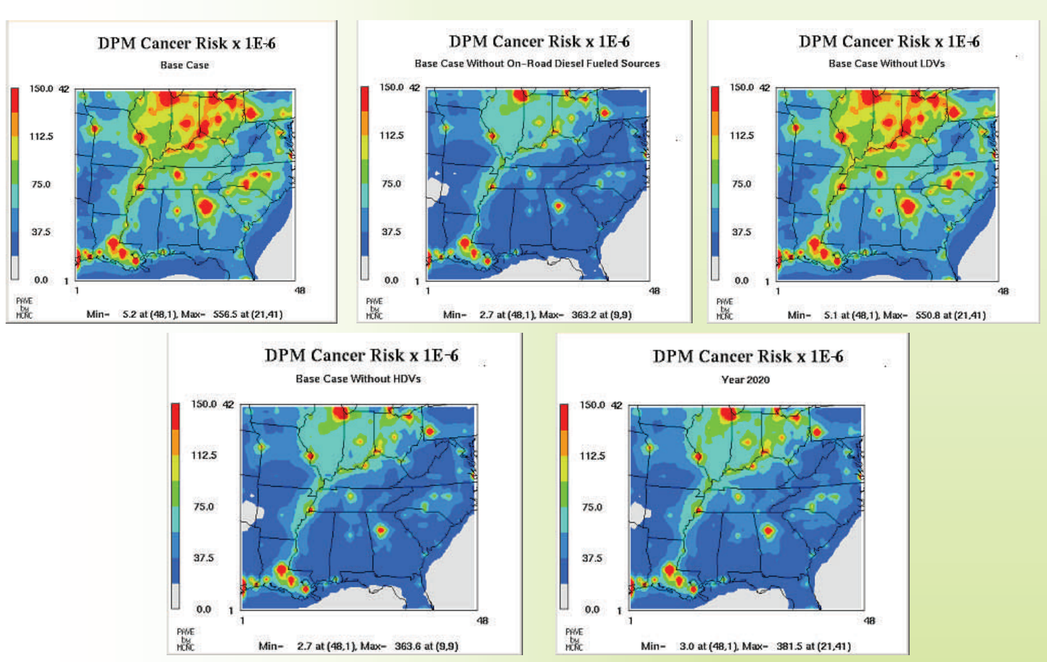


Figure 13. Spatial Distribution of the DPM Inhalation Cancer Risk on the Modeling Domain by DFS Scenarios

Conclusions

- The model under predicted the EC concentrations in Nashville for 2003.
- The CMAQ base case performance was good enough to analyze the proposed emission scenarios on ground level concentrations and health risk, since the analysis approach considered the difference in mass concentrations and health risk values among the proposed emission scenarios and the base case scenario rather than the absolute mass concentration or health risk value.
- The urban DPM concentrations decreased rapidly outside the source or urban areas to relatively low background concentrations.
- The maximum hourly EC and DPM concentrations occurred between 6 and 9 PM and the minimum concentrations occurred between 1 and 3 PM. A second maximum occurred between 6 and 10 AM
- The main sinks for the emitted primary EC and DPM were the wind dispersion at afternoon hours, as well as, nucleation, accumulation, and particles deposition. An important amount of EC and DPM emitted during morning rush-hour traffic were dispersed at around 2 PM as a result of the higher wind speeds during afternoon hours. At the end of the day and at night, the EC and DPM emitted during afternoon rush-hour traffic dropped and dispersed slowly until the new EC and DPM were emitted during morning traffic congestion again.
- The highest DPM concentrations were produced in Atlanta, GA, which were typically a factor of 11 times higher than rural sites.
- The Nashville population was more exposed to DPM at morning and afternoon rush-hour traffic. The monthly DPM concentrations were almost similar for all months at Nashville. The annual modeled DPM concentration was 0.55 ug/m^3 in Nashville, TN, 2003. The daily DPM contribution to the total PM2.5 was similar for each month and was around 2.6%.
- The DPM inhalation cancer risk was 163.6 in one million of population, which generated the higher lifetime cancer risk excess among the monitored air toxics acetaldehyde, benzene, 1,3-butadiene, and formaldehyde in Nashville, posing a cancer risk that was 4 times higher than the combined total cancer risk from all those air toxics.
- The DPM reduction in 2020 due to on-road sources was 38.1%, which is not enough of a health point of view, since the cancer risk reductions were as low as 32.7 % for Nashville.
- On-road DFS contributed by almost 48.3% meanly due to HDDVs, the rest was contributed by Non-road DFS (52.7 %) in Nashville.
- The highest cancer risk occurred in Atlanta, GA, on the Southeastern U.S. area.