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A GIS-based national emission inventory of major VOCs and risk assessment modeling: Part II - quantitative verification and risk assessment using an air dispersion model

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Abstract−We compiled contour maps of the concentrations of major volatile organic compounds (VOCs) and corresponding chronic inhalation and carcinogenic risks in South Korea for the year 2004. We used the CALPUFF modeling system, an atmospheric dispersion model, to calculate the concentrations of five species of VOCs (benzene, ethylbenzene, styrene, toluene, and xylene). This modeling approach was used to validate the emission rates estimated in Part I of this study. The predicted concentrations show good overall agreement with the observed concentrations. In terms of risk assessment, we studied the chronic and carcinogenic effects on human health based on toxicity data and predicted concentrations of VOCs in ambient air. Risk levels were influenced by regional characteristics and spatial emission patterns. The results of such risk assessments could be used in support of air quality management in South Korea.

Key words: CALPUFF, Contour Map, Regional Distribution, Risk Assessment, VOCs

INTRODUCTION

Volatile organic compounds (VOCs) are of wide concern because of their adverse effects on human health, including acute toxic effects (primarily neurological), carcinogenic effects (e.g., leukemia), neurobehavioral effects, and risk of kidney damage [1]. In addition, ambient VOCs in the air contribute to the formation of ozone and photochemical oxidants implicated in the formation and fate of airborne toxic chemicals and fine particles [2]. Therefore, it is important to assess human exposure levels based on VOC concentrations in the ambient air.

Exposure levels are generally evaluated based on air quality monitoring, emission inventories, and air quality modeling and mapping [3]. Monitoring is typically the most accurate of these methods; however, this approach requires much time and money in estimating national-scale exposure levels. Emission inventories and air quality modeling are used because it has the drawback of requiring huge amounts of monitoring data. Air dispersion models can be used to calculate VOC concentrations at large spatial scales; consequently, such models represent an important tool in assessing national-scale human exposure levels as a component of risk assessment.

A number of air dispersion models have been developed and assessed in terms of their ability to estimate pollutant transport from emission sources using mathematical equations: (e.g., AURORA, CPB, and PBM), Gaussian models (e.g., CALINE4, and AERMOD), Lagrangian/Eulerian models (e.g., GRAL, CALPUFF, and TAPM), and CFD models (e.g., ARIA Local and MICRO-CALGRID) [4,5]. Of these, we chose the CALPUFF modeling system, Lagrangian puff atmospheric dispersal model as recommended by the U.S. Environmental Protection Agency (USEPA) for simulating long-range transport, for modeling the influence of VOCs on air quality. CALPUFF has the ability to generate and handle complex three-dimensional wind fields, and includes a complex terrain algorithm that is necessary when the target domain is expanded to include all of South Korea, as in the present study [6].

We used CALPUFF to model the VOC concentrations (i.e., benzene, ethylbenzene, styrene, toluene, and xylene) emitted from all anthropogenic sources. The detailed spatial trends and emission rates profiles from VOC emission sources were calculated and qualitatively validated in Part I of this study. In the present study, these emissions were validated via air dispersion modeling, with the predicted concentrations being compared with air-quality monitoring data. The spatial trends of atmospheric concentrations of the target VOCs were plotted on contour maps and analyzed in terms of site characteristics, including emission inventory categories.

Furthermore, the individual health risks of VOCs were evaluated from predicted concentrations based on inhalation toxicity data. The chronic risk of target VOCs was analyzed via a hazard quotient (HQ), and the carcinogenic risk of benzene was calculated using a slope factor. We also calculated the contribution of each VOC to the total HQ to compare the risk factors of the different target VOCs in each region of South Korea. We examined the major factors that influence the risk obtained for each region, based on the spatial patterns and profiles of VOC concentrations apparent in the risk contour maps.

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METHODS

1. CALPUFF Modeling System

The CALPUFF modeling system, combined with a three-dimensional meteorological and land-use field, was developed for modeling the movement of the contaminants that make up air pollution [6]. The CALPUFF model is a multi-layer non-steady-state puff dispersion model designed to model the dispersion of gases and particles. The model contains spatially and temporally varying meteorology based on similarity theory, turbulence, emission strength, transformation, and removal. CALPUFF was approved and recommended by the U. S. Environmental Protection Agency (USEPA) for regulatory applications involving long-range transport [7].

The system includes three main components (CALMET, CALPUFF, and CALPOST) and a large set of preprocessing programs designed to interface the model to standard, routinely available meteorological and geophysical datasets. CALMET is a meteorological model that develops hourly wind and temperature fields on a three-dimensional modeling domain. CALPUFF is a transport and dispersion model that advects "puffs" of material emitted from modeled sources, as well as simulating dispersion and transformation processes. Meteorological data in the CALPUFF model are run with a 1 hour time step for an average duration of 24 hours for each sampling period, using the meteorological data generated in the CALMET modeling system. CALPOST is used to produce gridded concentrations that summarize the simulation results, identifying the highest and second-highest 3-hour average concentrations at each receptor. The CALPUFF model can be used to study in detail the contribution to the model domain area of substances from various pollutant sources [6].

2. Domain and Area Emission-source Approach Using GIS

In using the CALPUFF modeling system, we created a 400×600 km modeling domain that covered all of the populated areas in South Korea. To demonstrate the nationwide distribution of pollutant concentrations, the modeling employed 20×30 grid cells with receptor points in both the X and Y directions, with a grid spacing of 20 km. Physical data employed in the model included landuse at a grid size of 30 m (as derived from an environmental geographic information system, EGIS) and terrain height at a grid size of 100 m (as generated from a GIS program). Meteorological data, derived from 70 surface stations distributed throughout South Korea and 1 upper-air station for the period from January to December of 2004 (1 year, long-term monitoring), are input to the CALMET modeling system to generate the meteorological field (Fig. 1). The five vertical layers in the meteorological field were set at heights of 20, 100, 500, 1,000, and 2,000 m.

VOC emission data are required for CALPUFF modeling. The spatially resolved emission quantities of various pollutants (1×1 km)

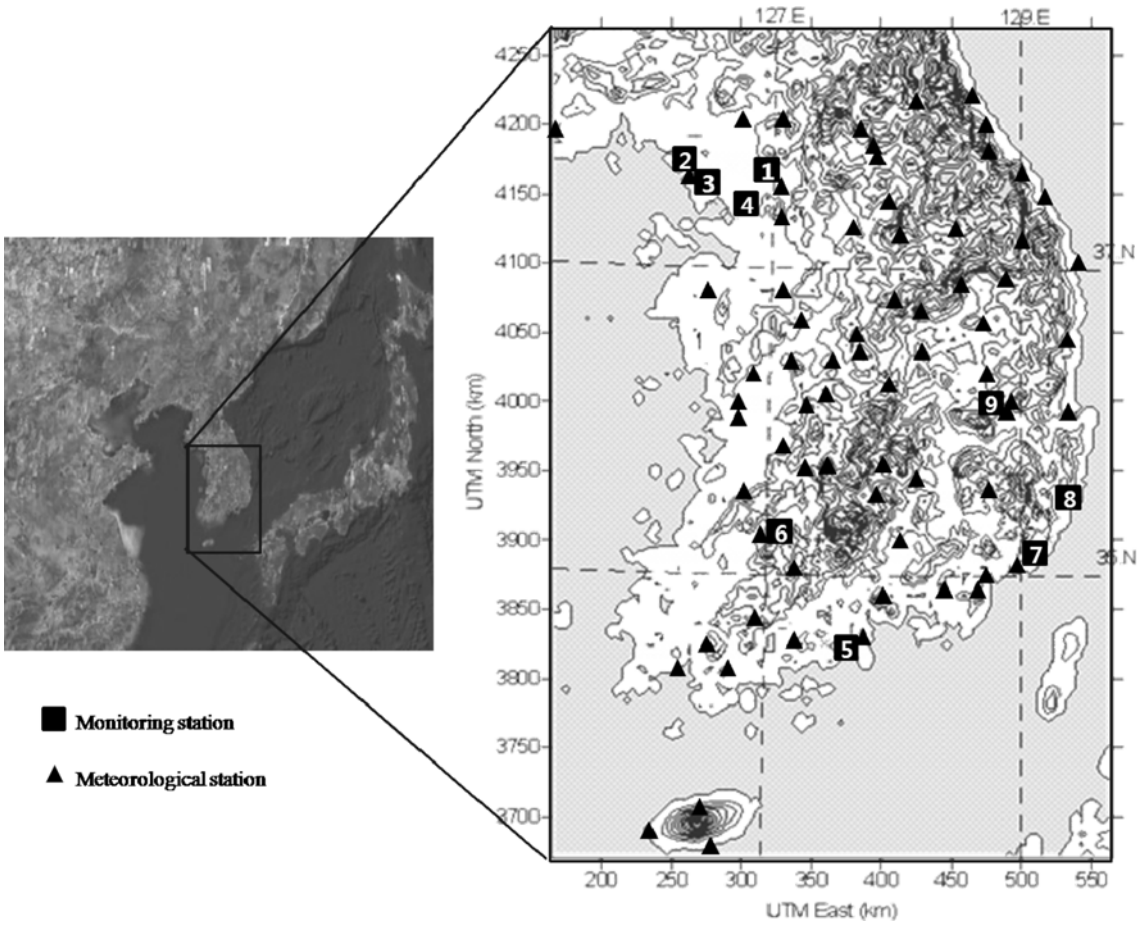

Fig. 1. Locations of monitoring and meteorological stations plotted on a topographic map of the study area (key to station names: 1. SU, 2. IC, 3. GH, 4. SH, 5. YS, 6. GJ, 7. BS, 8. US, 9. DG).

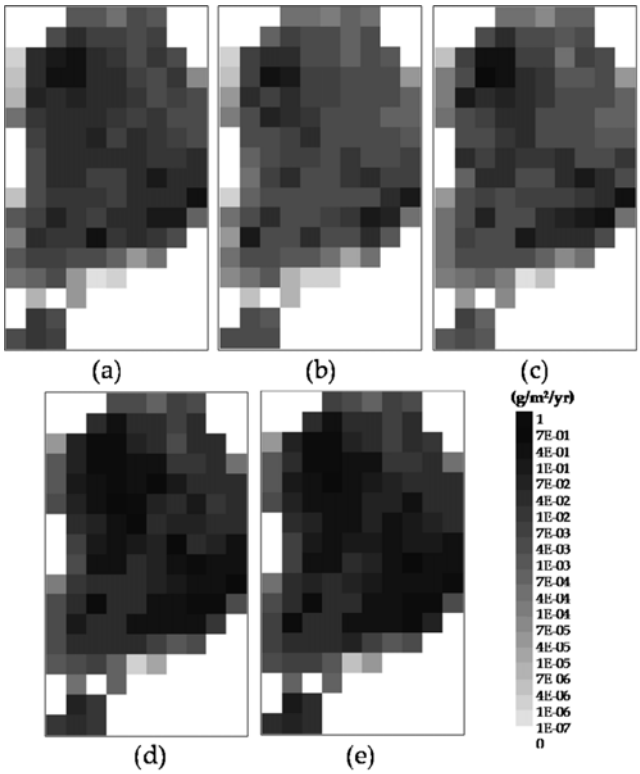

Fig. 2. South Korea-wide emission quantities of VOCs mapped using GIS, taken from Part I of this study. (a) benzene, (b) ethylbenzene, (c) styrene, (d) toluene, and (e) xylene. The area of each map was contained the study area shown in Fig. 1.

were sourced from Part I of this study. To input the emission sources into CALPUFF modeling, 1×1 km emission data were rearranged into 10×17 gridded sub-sections. The resulting spatial distribution of the emission quantities of target VOCs in South Korea is shown in Fig. 2. The sub-sections are numbered from upper left to lower right over the modeling area (see Fig. 5 in Part I), with some being representative of urban areas (sub-sections 23, 24, 34, 65, 88, and 103), industrial areas (100, 115, and 125), and mixed urban-industrial areas (33, 44, 54, 55, 108, and 109). Additional information regarding the emission inventory and emission factors used in this study can be found in Part I.

3. Model Evaluation

The accuracy of model predictions was assessed by comparing the predictions with observed concentrations sourced from monthly air quality reports for 2004 published by the Ministry of Environment, South Korea [8]. The observed data were measured quarterly. So, the mean predicted and observed concentrations in each quarter of the year were obtained and compared at the sites of nine monitoring stations (SU, IC, GH, SH, YS, GJ, BS, US, and DG), as shown in Fig. 1.

4. Risk Characterization

To investigate the effects on human health from the obtained concentrations of target pollutants, we assessed the risks associated with each VOC. The risk to human health in a given region depends on the possibility of exposure and the properties of the relevant pollutants. The overall risk involves exposure via several possible pathways (e.g., oral, dermal, or inhalation routes). The high volatility of the target VOC species considered in the present study means that the dominant exposure risk for humans is by inhalation [9]; consequently, only inhalation exposure is considered herein.

Data regarding VOC toxicity effects on human health were sourced from an integrated risk information system (IRIS). To estimate chronic human-health risk, annual average concentration was compared with the reference concentration (RfC) of chronic toxicity data. In general, the RfC value is an estimate of the inhalation exposure that is unlikely to result in an appreciable risk of deleterious effects during the person's lifetime [10].

The quantitative carcinogenic risk arising from inhalation exposure was estimated by the slope factor (SF), the value derived from the slope of a dose-response curve assumed to be linear at low concentrations [11]. This is the only applicable information regarding carcinogenic risk arising from inhalation exposure to benzene, because there exists a lack of clear data regarding the carcinogenic toxicity effects of other VOCs. Toxicity data for the five VOCs are listed at Table 1.

The chronic risks of VOCs can be expressed as HQ, defined as the ratio of a predicted environmental concentration (PEC) to a given RfC [12]:

$$HQ=PEC/RfC \quad (1)$$

When HQ<<1, the chronic risk effects are negligible. The sum of the HQs values for all the VOCs of interest was obtained to assess the chronic impacts of air pollution; this summed figure indicates the overall effect of VOCs in each region [13].

The predicted benzene concentration was also compared with the specified cancer-risk levels for evaluation of the carcinogenic risk. The risk associated with carcinogenic pollutants is expressed as the probability of contracting cancer via chemical exposure. With linear extrapolation, the risk associated with relatively low concentrations of pollutants is generally approximated by multiplying the slope factor by the estimated exposure:

$$Risk=Slope\ factor\times Exposure \quad (2)$$

Table 1. Chronic and carcinogenic toxicity data obtained for the five main VOCs [10]

Chemicals	Chronic toxicity (RfC, mg/m^3)	Carcinogenic toxicity (unit risk, (μg/m^3)$^{-1}$)
Benzene	0.03	8.1E-6
Ethylbenzene	1	Not classifiable for human carcinogenicity
Styrene	1	Not assessed under the IRIS program
Toluene	5	Inadequate information to assess carcinogenic potential
Xylene	0.1	Inadequate information to assess carcinogenic potential

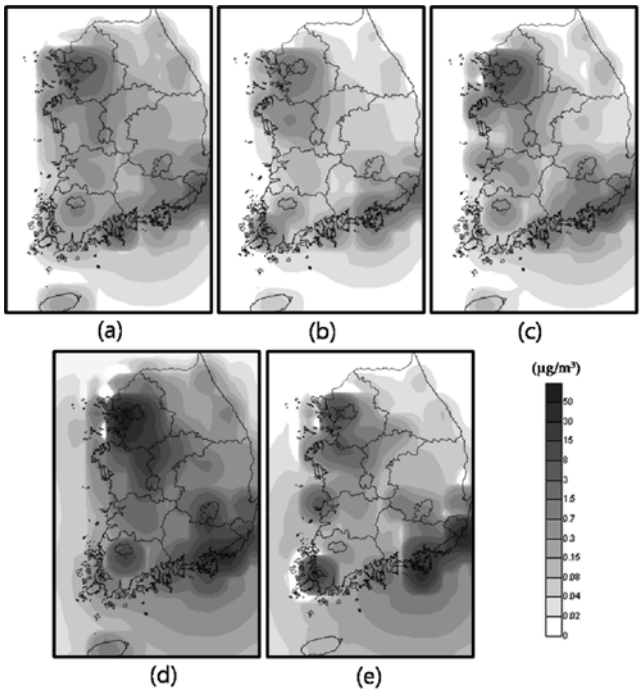

Fig. 3. Contour maps of the simulated annual average concentrations of (a) benzene, (b) ethylbenzene, (c) styrene, (d) toluene, and (e) xylene for the year 2004.

The exposure parameters used in our calculations were obtained from a previous study [14]. Areas with elevated cancer risks were identified based on the calculated carcinogenic risk values.

RESULTS AND DISCUSSION

1. Spatial Distribution of VOC Concentrations

Fig. 3 shows two-dimensional pollutant contour maps showing the concentrations of selected VOCs (benzene, ethylbenzene, styrene, toluene, and xylene) for 2004. The maps show the spatial ranges in concentrations of benzene (0.001-8.27 μg/m^3), ethylbenzene (0.001-3.19 μg/m^3), styrene (0.001-7.83 μg/m^3), toluene (0.01-83.8 μg/m^3), and xylene (0.005-66.8 μg/m^3). In almost all regions, toluene shows the highest concentration of the target VOCs, followed by xylene, benzene, styrene, and finally ethylbenzene. The concentration distributions reflect the emission patterns and the local distribution of air pollutants, thereby revealing the relevant emission sources.

We now consider the concentration distributions of each sector type, such as urban areas and industrial areas, according to the estimates based on grid-based emissions. In almost all cases, VOC concentrations are higher in urban regions with high population densities than in rural regions; in particular, the largest cities in South Korea (metropolitan areas and surrounding regions) have the highest average VOC concentrations. This finding could reflect the high emission levels in and around metropolitan areas because of the significant amount of pollution produced by large numbers of vehicles, a high population density, and VOC-emitting industries.

Areas with relatively high VOC concentrations were also observed in the southeast of South Korea, possibly reflecting the influence of large industrial areas with high emission rates of VOCs.

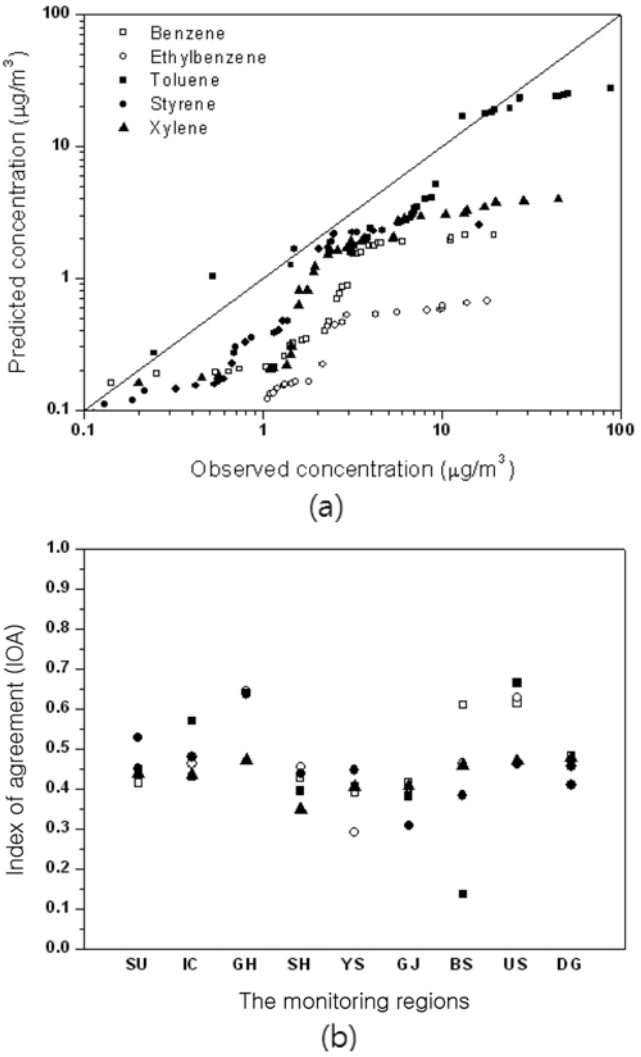

Fig. 4. Scatterplot (a) and IOA (b) for the comparison of observed and predicted mean concentrations of five target VOCs at nine sites in South Korea for 2004.

Consequently, the above results show that high VOC concentrations indicate the presence of sources in the area that emit large quantities of VOCs. In terms of air dispersion models, it is important that these distribution trends could reflect spatial variations in emission source characteristics and wind speed.

2. Comparison with Monitoring Data

To evaluate the accuracy of our modeling results, we compared the modeling data with monitoring data at nine sites (Fig. 4(a)). The predicted concentrations show good overall agreement with the observed concentrations, although the modeling data are generally slightly underestimated, with the exception of benzene and ethylbenzene, which are more strongly underestimated. The estimated benzene and ethylbenzene concentrations are lower than the observed concentrations by a factor of about 10; however, the estimated xylene, toluene, and styrene concentrations are generally within a factor of two to three of the observed concentrations, with the exception of several data points for toluene and styrene.

The results presented in Part I of this study revealed that the major emission sources of benzene and ethylbenzene are road trans-

port, and the major sources of styrene, toluene, and xylene are industrial installations, coating/painting in the construction industry and other industries, and domestic solvent use. The concentrations of benzene and ethylbenzene were much lower than measured values, probably because emissions from motorcycles and mopeds were not considered in assessing the total emissions from road transport, resulting in a likely underestimation.

Statistical measures, such as root mean square error (RMSE) and index of agreement (IOA) are frequently used to evaluate model performance in previous studies [15]. However, comparing RMSE between different pollutants and sites does not provide useful information because the magnitude of the measurements affects the RMSE, and then the concentrations of VOCs and the sites have different magnitudes. Thus, we use only IOA to evaluate the model performance across pollutants and sites. IOA is determined from each pair of predicted (P) and observed (O) values by using the following equation:

$$IOA = 1 - \left[\frac{\sum (P - O)^2}{\sum (|P - \overline{O}| + |O - \overline{O}|)^2} \right] \quad (3)$$

where $\overline{O}$ is the average values of observed data.

Fig. 4(b) shows IOA values of five VOCs in the nine regions. An IOA value above 0.5 is generally considered to be good in accuracy [16]. In our results, the IOA values range from 0.14 to 0.66 for all of the variables. 23% of IOA values are in the range from 0.5 to 0.66 at GH and US, which is the representative rural and industrial areas, respectively. 60% of IOA values range from 0.4 to 0.5, which is slightly smaller than the values considered to be good. This is caused by slight underestimation for overall emission rates. 17% of IOA values are lower than 0.3 at SH, YS, GJ and BS sites. Although there is a slight discrepancy between the predicted concentration and observed concentration based on the values of IOA, the model outputs show acceptable performance to understand the condition of environmental pollution.

The fact that the overall predicted VOC concentrations were lower than the observed concentrations suggests additional emission sources of VOCs not considered in our calculations. Therefore, further studies would require more precise emission quantities, including taking into account those items missing from the emission inventory employed in the present study (i.e., motorcycles, mopeds, and other emission sources).

The comparison between modeled and monitored concentrations revealed a normal error in the model predictions: the ambient background levels of past emissions. It should also be noted that uncertainties in the predictions might have arisen from two different sources: emission calculations and dispersion modeling. The insufficient data fully including emission quantity all the nationwide were used in CALPUFF. The VOCs are ones of the very reactive atmospheric compounds presented in the atmosphere. For the more accurate estimation of the VOCs concentrations in the air, complicated photochemical reactions should be considered. However, the CALPUFF model provides an estimation result of the VOCs concentrations after considering simple dispersion process, based on the three-dimensional atmospheric turbulence and deposition. Errors and uncertainties associated with the measurement techniques employed at the monitoring stations might also have contributed to the differences obtained between the modeled and monitored concentrations.

3. Risk Characterization and Spatial Distribution of Risk throughout South Korea

Using VOC concentrations predicted by the CALPUFF modeling system, we conducted a regional risk assessment for VOCs in South Korea for 2004. Chronic risk values for individual VOCs are low compared with RfC (HQ<1). Although toluene showed the highest atmospheric concentrations, the maximum risk value was found for xylene (0.67) because of its high level of toxicity, followed by benzene (0.28), toluene (0.017), styrene (0.008), and ethylbenzene (0.003).

To investigate the relative contribution of each species to chronic risk in different regions, we summed the HQs of the five VOC species, and plotted the chronic risk impact of each VOC for 170 spatial sub-sections representing 10×17 gridded sectors over the entire area of South Korea (Fig. 5). The highest HQ values were found in sectors that included large cities and industrial areas, reflecting emissions from road transport and the use of solvents in the domestic and commercial sectors. The highest risks values are associated with benzene and xylene, which account for over 97% of the chronic risk.

The contribution of each VOC to HQ varies with the emission characteristics in each sub-sector (Fig. 6). For example, the HQ value

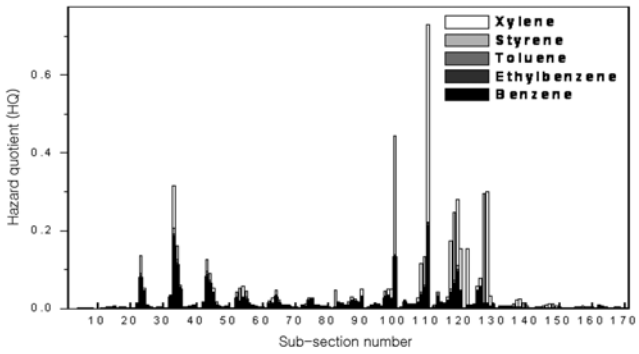

Fig. 5. Relative contributions to the overall hazard quotient of chronic risk of the five target species of VOCs for 170 sub-sections (see Fig. 5. in part I) in South Korea.

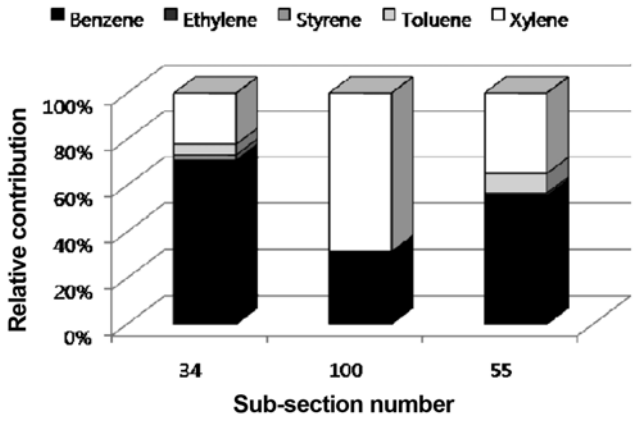

Fig. 6. Relative contributions of the five target VOCs to the hazard quotient calculated for three representative sub-sections (34: urban region, 100: industrial region, 55: mixed urban-industrial region) for the year 2004.

Table 2. Statistics for the five species of VOCs concentration in the nine regions by CALPUFF modeling

Pollutant	Region	$AVG_{\_OBS}$ (μg/m^3)	$STD_{\_OBS}$ (μg/m^3)	$AVG_{\_MOD}$ (μg/m^3)	$STD_{\_MOD}$ (μg/m^3)	IOA
Benzene	SU	3.70	5.13	1.8	0.25	0.41
	IC	4.46	6.02	1.76	0.26	0.43
	GH	2.37	1.42	0.05	0.01	0.64
	SH	4.38	4.55	1.93	0.13	0.43
	YS	3.91	1.55	0.79	0.08	0.39
	GJ	1.74	0.96	0.21	0.01	0.41
	BS	1.38	1.88	0.36	0.07	0.61
	US	8.02	10.16	0.3	0.08	0.61
	DG	2.27	2.06	0.17	0.02	0.48
Ethylbenzene	SU	3.13	4.62	0.54	0.08	0.49
	IC	4.93	5.99	0.49	0.07	0.46
	GH	0.89	0.49	0.01	0.00	0.64
	SH	3.59	3.17	0.61	0.04	0.45
	YS	1.09	0.32	0.15	0.02	0.29
	GJ	1.69	0.95	0.08	0.00	0.40
	BS	1.92	2.48	0.17	0.03	0.46
	US	9.64	8.38	0.11	0.03	0.63
	DG	3.92	4.47	0.05	0.01	0.46
Styrene	SU	0.52	0.66	1.94	0.27	0.45
	IC	1.30	0.86	1.93	0.29	0.57
	GH	0.32	0.28	0.02	0.01	0.64
	SH	0.99	0.99	2.29	0.16	0.40
	YS	2.96	1.39	0.43	0.05	0.40
	GJ	2.39	1.41	0.15	0.01	0.38
	BS	0.27	0.20	0.36	0.08	0.14
	US	1.84	1.14	0.20	0.06	0.66
	DG	5.05	7.38	0.10	0.01	0.46
Toluene	SU	8.81	7.44	19.62	2.64	0.53
	IC	19.02	16.65	20.68	3.12	0.48
	GH	6.42	5.82	0.21	0.06	0.66
	SH	15.21	12.02	25.24	1.74	0.44
	YS	4.97	2.09	1.76	0.20	0.45
	GJ	7.55	1.48	2.87	0.21	0.31
	BS	7.93	12.93	4.15	0.69	0.38
	US	16.38	21.03	1.34	0.25	0.46
	DG	46.83	36.08	2.93	0.46	0.41
Xylene	SU	5.58	7.79	1.82	0.23	0.44
	IC	9.65	12.8	3.45	0.53	0.43
	GH	0.93	0.78	0.03	0.01	0.47
	SH	5.25	5.96	3.4	0.29	0.35
	YS	2.19	1.00	0.84	0.20	0.40
	GJ	3.14	1.84	0.21	0.04	0.41
	BS	3.74	4.59	2.09	0.50	0.45
	US	17.49	19.93	1.86	0.65	0.47
	DG	5.86	6.14	0.22	0.06	0.48

*Key: AVG=Arithmetic Average, STD=Standard Deviation, OBS=Observations, MOD=Model Predictions, IOA=Index of Agreement (0=no agreement, 1=perfect agreement)

obtained for sub-section 34 (an urban area) is strongly influenced by benzene (which contributes 70% of the HQ), as ambient benzene is known to be related to vehicle emissions. In a representative industrial area (sub-section 100), HQ is strongly influenced by xylene (69%), related to emission sources of industrial solvents. A representative mixed urban-industrial area (sub-section 55) shows risk values intermediate between those of sub-sections 34 and 100.

We also created contour maps showing the geographical distri-

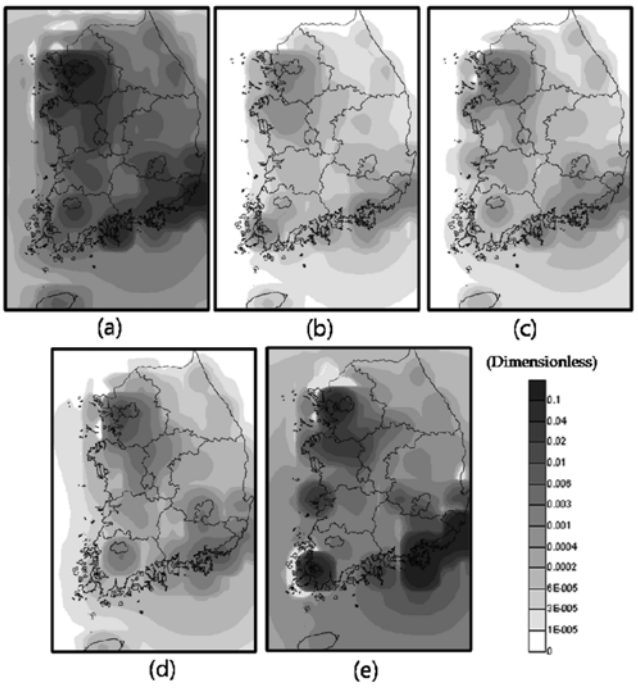

Fig. 7. Maps of the chronic risk levels of target VOCs for the year 2004. (a) benzene, (b) ethylbenzene, (c) styrene, (d) toluene, and (e) xylene.

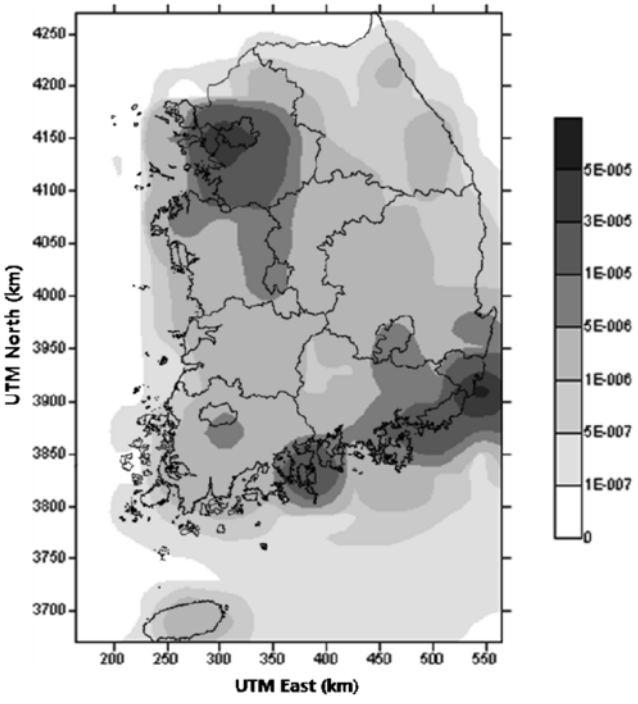

Fig. 8. Contour map of the distribution of carcinogenic risk level associated with benzene for the year 2004.

bution of chronic risk (Fig. 7) and carcinogenic risk (Fig. 8) in South Korea for the year 2004. Metropolitan areas show the maximum risk levels in all cases because of their high rate of VOC emissions. Large amounts of VOCs are emitted in areas with large numbers of vehicles, high population densities, and VOC-producing industries. In the case of xylene, the chronic risk in southern of South Korea is higher than that in other regions (except for metropolitan areas), reflecting the high concentration of xylene-emitting industrial installations in the south.

We estimated the quantitative carcinogenic risk from inhalation exposure based on the slope factor for the carcinogenic toxicity of benzene. In most regions of South Korea, the calculated carcinogenic risk values were significantly higher than 1.0×10^{-6} (i.e., less than one occurrence in 1 million of the population), representing a standard risk-protection factor, as stated in USEPA regulations [17].

The carcinogenic risk values for benzene are relatively high in some urbanized areas in South Korea, as shown in Fig. 8, indicating the high possibility of an above-average cancer rate. Our results indicate that emission-related risks to human health are the highest in metropolitan areas in South Korea. The risk of cancer mortality arising from the carcinogenic effects of benzene may have been underestimated in the present study; hence, benzene emissions should be restricted to reduce this risk.

Among the five major VOCs considered in this study, we propose that benzene and xylene are the principal sources of chronic risk to the environment and human health. In addition, the carcinogenic risk associated with benzene exceeds standard risk-protection factors provided by the USEPA. The risk values obtained for each sub-section were predominantly influenced by emission quantity and the characteristics of the emission inventory.

We emphasize that the results presented in this paper are the preliminary findings of an ongoing investigation. The proposed emission rates and CALPUFF modeling system will be further developed, taking into account more specific information related to emission sources and inventories, along with site characteristics.

CONCLUSIONS

We developed a decision-support system, using air quality modeling and risk assessment, for air quality management throughout South Korea. The air quality model CALPUFF is the main component of the system, which is mainly used in estimating air quality. The CALPUFF modeling system was successfully employed in estimating the ambient VOC concentrations throughout South Korea. To evaluate the accuracy of the estimated emissions, the predicted concentrations were compared with observed concentrations at the sites of nine monitoring stations. The obtained spatial distribution of VOC concentrations reveals some important trends, including the locations of the main industrial regions and institutions that emit VOCs.

Based on predicted VOC concentrations and toxicity data, we assessed the inhalation risk throughout South Korea in terms of chronic (benzene, ethylbenzene, styrene, toluene, and xylene) and carcinogenic (benzene) effects on human health. For all regions, the assessment of chronic risk revealed values below the maximum acceptable levels reported by USEPA. To investigate the contribution of each species to the chronic risk in each region, we summed the HQ values for the five VOC species. A carcinogenic risk assessment was also performed to evaluate the inhalation-related cancer risk associated with ambient benzene concentrations.

Using the above modeling data, we compiled contour maps of VOC concentration and risk level in South Korea for the year 2004.

The concentration of VOCs was highest in urban and industrial areas. Likewise, risk values are relatively high in and around urban areas, especially metropolitan areas. These findings probably reflect the quantities of emission produced in metropolitan and industrial areas because of large numbers of vehicles and VOC-emitting industries.

Our risk assessment revealed that the chronic risks of VOCs were mainly related to benzene in urban areas and xylene in industrial areas. The carcinogenic risk of benzene exceeded standard risk-protection factors; therefore, a policy is required to reduce emissions of benzene from road transport in urban areas, and reduce emissions of xylene from industrial installations located in southern of South Korea. Future studies of air quality management in South Korea should perform quantitative risk assessments of other chemicals (i.e., in addition to those considered in the present study) in the ambient air.

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