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CalTOX 모델을 이용한 대산 석유화학단지의 활동단계에 따른 벤젠 흡입 노출평가

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Prediction of Inhalation Exposure to Benzene by Activity Stage Using a Caltox Model at the Daesan Petrochemical Complex in South Korea

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ABSTRACT

Background: Chemical emissions in the environment have rapidly increased with the accelerated industrialization taking place in recent decades. Residents of industrial complexes are concerned about the health risks posed by chemical exposure.

Objectives: This study was performed to suggest modeling methods that take into account multimedia and multi-pathways in human exposure and risk assessment.

Methods: The concentration of benzene emitted at industrial complexes in Daesan, South Korea and the exposure of local residents was estimated using the Caltox model. The amount of human exposure based on inhalation rate was stochastically predicted for various activity stages such as resting, normal walking, and fast walking.

Results: The coefficient of determination (R^2) for the CalTOX model efficiency was 0.9676 and the root-mean-square error (RMSE) was 0.0035, indicating good agreement between predictions and measurements. However, the efficiency index (EI) appeared to be a negative value at –1094.4997. This can be explained as the atmospheric concentration being calculated only from the emissions from industrial facilities in the study area. In the human exposure assessment, the higher the inhalation rate percentile value, the higher the inhalation rate and lifetime average daily dose (LADD) at each activity step.

Conclusions: Prediction using the Caltox model might be appropriate for comparing with actual measurements. The LADD of females was higher ratio with an increase in inhalation rate than those of males. This finding would imply that females may be more susceptible to benzene as their inhalation rate increases.

Key words: Inhalation exposure, CalTOX, activity stages, multimedia, multiple-pathway

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Highlights:

- Multiple-pathway and multimedia modeling are increasingly important in exposure assessment of chemical emissions.
- The predictions using the CalTOX model were shown in good agreement with the actual measurements.
- The higher the inhalation rate percentile value, the higher the inhalation rate and lifetime average daily dose at each activity step.
- Females may be more susceptible to benzene as their inhalation rate increases.

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I. Introduction

Recent epidemiological studies on human exposure assessment for chemicals released into the environment are to estimate the total dose received by an individual through various contact media and exposure pathways. In general, a complete exposure assessment must identify the chemical of concern and all potential exposure pathways. The concentrations of the pollutants in various contact media determine the frequency and duration of contact between the visible external boundary of the receptor and each exposure medium, and quantify the level of certainty in the exposure assessment.[1,2] Therefore, it would be necessary to predict the effects of exposure and to determine how much it can be reduced.[3]

A major technology in the exposure assessment is the multimedia and multiple-pathway exposure assessment approach.[4] This assessed all possible multiple-pathway by which humans may be exposed to pollutants at multimedia. Multimedia has media of air, water, soil, and sediment, and the multiple-pathway is divided into inhalation, skin absorption, and ingestion.

Though target chemicals can be measured directly in multiple-pathway human exposure, the research data collection could be impossible or insufficient due to economic and time constraints.[5] As a result, studies using models to simulate exposure scenarios and to assess human exposure have been considered.[6] Since 1985, the advancement of computer has resulted in the development of an entire discipline of multimedia modeling of pollutants, as well as many useful techniques and modeling tools have been developed.[7] Therefore, exposure can be effectively managed if modeling could be performed while considering multimedia and multiple-pathway in human exposure assessment.[8]

Modeling, on the other hand, provides a simplification of reality that is formed to gain insights into specific attributes of a physical, biological, economic, or social system.[9,10] However, there is uncertainty in the evaluation results because of a lack of accurate input variables, which reduces the reliability of the evaluation results.[11] For example, basic research on contact strength is needed to reduce the uncertainty caused across different activity stages such as resting, normal walking, and fast walking. The speed of contact with polluted media could be the primary focus of information on contact strength.[12] Using a model for assessing pollutants exposure, it might be critical to define the intensity of exposure from pollutants to identify important epidemiological and clinical applications either qualitatively or quantitatively.[13]

The U.S. Environmental Protection Agency (US EPA) classifies benzene, a volatile organic compound, as a Group A human carcinogen and the International Agency for Research on Cancer (IARC) classifies it as a Group 1 carcinogen.[14,15] Benzene is ubiquitous in the environment. Although large amount are released naturally through forest fires and volcanic activity, the main environmental sources of benzene are anthropogenic. Such sources include industrial emissions, burning of coal and oil, motor vehicle exhaust, and fuel evaporation.[16] Benzene is generally present in urban or industrialized areas.[17] Since the 1980s, along with the rapid development of industrial complexes in Korea, the types and amounts of circulating chemicals, including benzene, began to increase significantly.[18] As a result of tracking the emissions of five industrial complexes around Ulsan, there was a research result that chemical substances accounted for about 30% of the whole emissions.[19]

Researchers in Korea have been using multimedia and multiple-pathway models to assess human exposure to chemicals since the late 1990s.[20] Therefore, it could be critical to conduct a multimedia, multiple-pathway human exposure assessment using big data and to explain the model's validity.[21] In addition, research to reduce the uncertainty of the human exposure assessment results due to various activities should be conducted concurrently.[22] In this study, we predicted the atmospheric concentrations of benzene emitted from Daesan-eup, Seosan-si, Chungcheongnam-do, where industrial and petrochemical complexes were concentrated using a multimedia and multiple-pathway model, and analyzed the model efficiency. Furthermore, by probabilistically predicting human exposure based on the inhalation rates of men and women at various stages of activity, it was attempted to secure a basis for decision-making when determining environmental health policies for benzene by applying the Korean general exposure factor.

II. Materials and Methods

1. Description of the site

Daesan city is located in the Taean peninsula, an area that projects from the northwestern part of Chungcheongnam-do in Korea, where the Daesan petrochemical complex is situated, has an area of 105.71 km^2, and consists of different land categories including forests (41.3%), rice fields (18.4%), and the petrochemical complex (10.1%). Daesan petrochemical complex is divided into Industrial Complex 1, which houses oil refineries, industrial complex 2, which houses coke, briquette manufacturers, and refineries, and industrial complex 3, which is a chemical and petrochemical industrial complex.

2. Pollutant emission source tracking information

The location of the Daesan industrial complex, the area of the research and monitoring stations, and the major emission sources are shown in Fig. 1. The population was 13,926 as of the end of December in 2019. This study was conducted on men and women over the age of 19 using data from the 'Korean Exposure Factor Handbook' revised 2019 by the Ministry of Environment. The amount of benzene emitted into the atmosphere at the Daesan Industrial Complex's five emission sites was calculated using data from the 2017 pollutant release and transfer register (PRTR).[23] The input data for landscape properties and meteorological data were obtained from the Korea Meteorological Administration.[24] The area information was obtained from the National Statistical Office.[25]

3. Human exposure modeling

The human exposure models used in CalTOX were based on those described by the US EPA and consisted of relating pollutant concentrations in the multimedia model compartments to those in a media in contact with the human population (air, water, food, household dust, soil, etc.). The Potential dose, expressed as an average daily dose, was the amount of a chemical per body weight per day (mg/kg–day) that entered into the mouth of an exposed individual by inhalation or ingestion or the outer layer of the skin through dermal contact.[26]

The general exposure factors were obtained from the Korean Exposure Factors Handbook.[27] The different values of inhalation rate (m^3/kg–h), body weight (kg), and body surface area (m^2/kg) for males and females, and the inhalation rate values of 10th, 25th, 50th, 75th, and 95th percentiles corresponding to different activity stage such as resting, normal walking, and fast walking were used. The resting stage was 5 to 10 minutes, normal walking was 5 to 10 minutes (3.5 to 4 km/h), and fast walking was 5 to 10 minutes (5.5 to 6 km/h). The Lifetime Average Daily Dose (LADD) of the inhalation rate was analyzed by setting the exposure time to 20 years with an average of 30,185.5 days. The data on chemical properties of benzene were obtained from the California Environmental Protection Agency's Department of Toxic Substances Control (Cal EPA DTSC).[28]

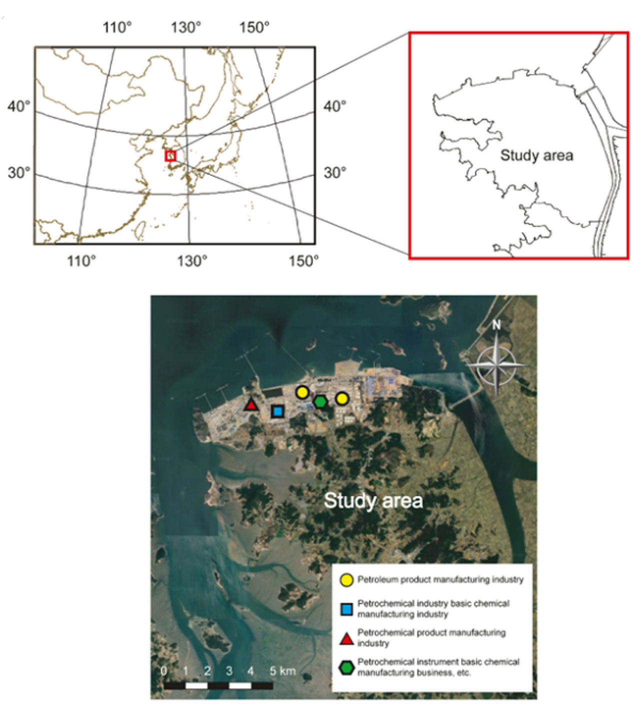

Fig. 1. Position of the industrial complex (upper left), study area and the locations of the monitoring stations (upper right), and locations of the major sources of emissions (below) in Daesan. The coordinates are expressed in UTM32 (m)

4. Uncertainty analysis in exposure assessment

Chemical properties in the form of a uniform probability distribution were used to predict probabilistic human inhalation exposure. The exponential probability distribution was used for precipitation, the Kumaraswamy probability distribution for annual average temperature, the Weibull probability distribution for annual average wind speed, and the exponential probability distribution for evaporation of water from surface water as input variables for regional characteristics. The body weight and inhalation rate were applied in the form of a triangular probability distribution to human exposure characteristic variables. The exposure duration were applied in the form of a uniform probability distribution. For other input variables of normal and lognormal probability distribution was applied. Monte-Carlo simulations were performed using @Risk 7.0 (US Palisade) for analysis of uncertainty in exposure assessments.

5. Efficiency assessment for models

The CalTOX model was used to estimate the atmospheric concentration of benzene emitted by the Daesan industrial complex and compare it with the measured concentration. Efficiency criteria are mathematical measures of how well a model simulation matches the available observations.[29] Data for measured concentrations were obtained from the Atmospheric Environment Yearbook of National Institute of Environmental Research. The efficiency criteria for the CalTOX model were evaluated by using a combination of the coefficient of determination (R^2), root-mean-square error (RMSE), and the Nash–Sutcliffe efficiency index (NSE).[30] The RMSE is an absolute criterion, and R^2 and NSE are relative criteria. They are calculated by equations (1), (2), and (3), respectively.

$$R^2 = \left\{ \frac{\sum_{i=1}^{n} [Q_o(i) - Q_m][Q_s(i) - Q_{ms}]}{\left[\sum_{i=1}^{n} [Q_o(i) - Q_m]^{0.5}\right]\left[\sum_{i=1}^{n} [Q_s(i) - Q_{ms}]^{0.5}\right]} \right\}^2 \quad (1)$$

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n} [Q_o(i) - Q_s(i)]^2} \quad (2)$$

$$NSE = 1 - \frac{\sum_{i=1}^{n} [Q_o(i) - Q_s(i)]^2}{\sum_{i=1}^{n} [Q_o(i) - Q_m]^2} \quad (3)$$

Where,

Q_m: $\frac{\sum_{i=1}^{n} [Q_o(i)]^{0.5}}{n}$ is the arithmetic mean of measured values of Q_o (i)

Q_{ms}: $\frac{\sum_{i=1}^{n} [Q_s(i)]^{0.5}}{n}$ is the arithmetic mean of measured values of Q_s (i)

Q_o (i): measured data at i

Q_s (i): simulated data at i

III. Results

1. Efficiency analysis of the model

The CalTOX model divided benzene emitted from industrial areas into gaseous and particulate phases to predict atmospheric concentrations. Since the predicted value of particulate phase concentration was negligible, the gaseous phase concentration was used in the analysis. benzene concentration using the CalTOX model was 1.08×10^{-4} mg/m^3, The predicted value of 5 percentile is 9.88×10^{-5} and the value of 95 percentile is 1.30 $\times10^{-4}$. Meanwhile, The measured concentration of benzene was 1.95×10^{-4} mg/m^3, and the distribution was analyzed in percentiles as shown in Table 1.

The results of the efficiency analysis of the model using the actual measured concentrations are the R^2 and RMSE were 0.9676 and 0.0035, respectively. However, the NSE was negative (–1,094.50). These findings indicated that the variance of predicted values was the good agreement as the variance of observed values, but the model was inefficient. The slope and the intercept of the regression were derived with the y=15.485x+0.002.

2. Inhalation exposure by behavior

The higher the percentile, the higher the inhalation rate at each activity stage for all males and females. Except for the 25 percentile of the fast walking stage, females inhaled at a higher rate than males. At the 25 percentile in fast walking, the inhalation rates of the males and females were 2.50×10^{-2} m^3/kg–h

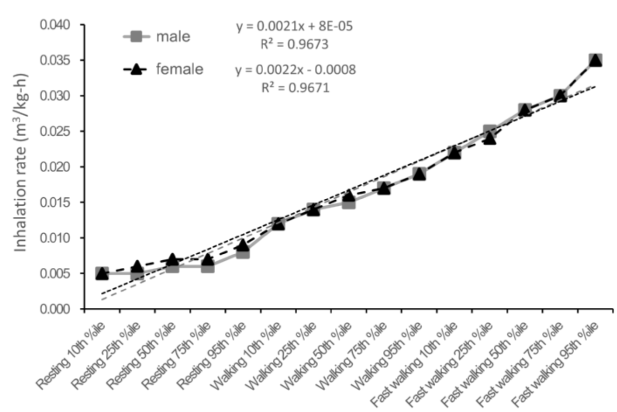

Fig. 2. Correlation between inhalation rates and different activity stages in males and females

Table 1. Monte-Carlo simulation results of the predicted and measured concentrations (mg/m^3) of benzene in the air

Concentration		Simulation				
		5th	25th	50th	75th	95th
Predicted	1.08×10^{-4}	9.88×10^{-5}	1.05×10^{-4}	1.09×10^{-4}	1.15×10^{-4}	1.30×10^{-4}
Measured	1.95×10^{-4}	1.69×10^{-4}	1.95×10^{-4}	2.16×10^{-4}	2.39×10^{-4}	2.83×10^{-4}

and 2.40×10^{-2} m^3/kg–h, respectively. The correlation between the percentiles of each activity stage and inhalation rate was $y=2.2\times10^{-3}x+8\times10^{-4}$ (R^2=0.96) for male and $y=2.1\times10^{-3}x+8\times10^{-5}$ (R^2=0.97) for female as shown in Fig. 2.

3. Human exposure assessment of benzene

The results of the Monte-Carlo simulation are shown in Table 2. All male and female exposures using CalTOX model (LADD_inhalation) were 1.20×10^{-4} (90% CI; 1.03×10^{-4} to 1.60×10^{-4}) mg/kg–day. The probabilistic prediction of human exposure based on the inhalation rate at various activity stages for males and females are shown in Table 2. Human exposures between males and females for each activity stage were statistically significant difference. This may explain the different levels of exposure to benzene in men and women at different stages of activity. Regression analysis between LADD and the percentiles of each activity stage was $y=2.05x\times10^{-5}+2.27\times10^{-5}$ (R^2=0.96) for male and $y=1.98x\times10^{-5}+3.47\times10^{-5}$ (R^2=0.96) for female.

Table 2. Monte-Carlo simulation of human exposure to benzene predicted by using the CalTOX model

			LADD (mg/kg-day)	Simulation (mg/kg–day)					p-value*
				5th	25th	50th	75th	95th	
LADD_inhalation			1.20×10^{-4}	1.03×10^{-4}	1.19×10^{-4}	1.29×10^{-4}	1.41×10^{-4}	1.60×10^{-4}	NA.
Resting	10th	Male	7.70×10^{-5}	6.10×10^{-5}	7.00×10^{-5}	7.70×10^{-5}	8.30×10^{-5}	9.60×10^{-5}	0.000
		Female	8.20×10^{-5}	6.90×10^{-5}	7.50×10^{-5}	8.20×10^{-5}	8.90×10^{-5}	1.02×10^{-4}	
	25th	Male	7.70×10^{-5}	6.20×10^{-5}	7.00×10^{-5}	7.60×10^{-5}	8.40×10^{-5}	9.70×10^{-5}	0.000
		Female	9.10×10^{-5}	7.30×10^{-5}	8.40×10^{-5}	9.10×10^{-5}	1.00×10^{-4}	1.13×10^{-4}	
	50th	Male	8.60×10^{-5}	6.90×10^{-5}	7.90×10^{-5}	8.60×10^{-5}	9.50×10^{-5}	1.07×10^{-4}	0.000
		Female	1.01×10^{-4}	7.90×10^{-5}	9.20×10^{-5}	1.00×10^{-4}	1.10×10^{-4}	1.27×10^{-4}	
	75th	Male	8.60×10^{-5}	6.80×10^{-5}	8.00×10^{-5}	8.70×10^{-5}	9.40×10^{-5}	1.07×10^{-4}	0.000
		Female	1.01×10^{-4}	7.60×10^{-5}	9.20×10^{-5}	1.02×10^{-4}	1.10×10^{-4}	1.27×10^{-4}	
	95th	Male	1.15×10^{-4}	9.10×10^{-5}	1.05×10^{-4}	1.10×10^{-4}	1.26×10^{-4}	1.48×10^{-4}	0.000
		Female	1.20×10^{-4}	9.00×10^{-5}	1.09×10^{-4}	1.20×10^{-4}	1.31×10^{-4}	1.51×10^{-4}	
Normal walking	10th	Male	1.43×10^{-4}	1.14×10^{-4}	1.31×10^{-4}	1.44×10^{-4}	1.57×10^{-4}	1.81×10^{-4}	0.000
		Female	1.48×10^{-4}	1.17×10^{-4}	1.36×10^{-4}	1.48×10^{-4}	1.61×10^{-4}	1.88×10^{-4}	
	25th	Male	1.62×10^{-4}	1.30×10^{-4}	1.49×10^{-4}	1.62×10^{-4}	1.78×10^{-4}	2.07×10^{-4}	0.000
		Female	1.67×10^{-4}	1.32×10^{-4}	1.53×10^{-4}	1.68×10^{-4}	1.82×10^{-4}	2.08×10^{-4}	
	50th	Male	1.72×10^{-4}	1.34×10^{-4}	1.57×10^{-4}	1.73×10^{-4}	1.89×10^{-4}	2.15×10^{-4}	0.000
		Female	1.86×10^{-4}	1.47×10^{-4}	1.71×10^{-4}	1.86×10^{-4}	2.04×10^{-4}	2.32×10^{-4}	
	75th	Male	1.91×10^{-4}	1.51×10^{-4}	1.77×10^{-4}	1.91×10^{-4}	2.08×10^{-4}	2.40×10^{-4}	0.000
		Female	1.96×10^{-4}	1.54×10^{-4}	1.79×10^{-4}	1.97×10^{-4}	2.14×10^{-4}	2.44×10^{-4}	
	95th	Male	2.10×10^{-4}	1.64×10^{-4}	1.90×10^{-4}	2.10×10^{-4}	2.31×10^{-4}	2.69×10^{-4}	0.001
		Female	2.15×10^{-4}	1.71×10^{-4}	1.98×10^{-4}	2.17×10^{-4}	2.35×10^{-4}	3.64×10^{-4}	
Fast walking	10th	Male	2.39×10^{-4}	1.91×10^{-4}	2.17×10^{-4}	2.38×10^{-4}	2.63×10^{-4}	3.03×10^{-4}	0.003
		Female	2.43×10^{-4}	1.90×10^{-4}	2.23×10^{-4}	2.44×10^{-4}	2.68×10^{-4}	3.10×10^{-4}	
	25th	Male	2.67×10^{-4}	2.08×10^{-4}	2.44×10^{-4}	2.67×10^{-4}	2.91×10^{-4}	3.40×10^{-4}	0.009
		Female	2.63×10^{-4}	2.04×10^{-4}	2.40×10^{-4}	2.63×10^{-4}	2.87×10^{-4}	3.32×10^{-4}	
	50th	Male	2.96×10^{-4}	2.34×10^{-4}	2.69×10^{-4}	2.95×10^{-4}	3.24×10^{-4}	3.78×10^{-4}	0.022
		Female	3.01×10^{-4}	2.32×10^{-4}	2.74×10^{-4}	3.00×10^{-4}	3.30×10^{-4}	3.88×10^{-4}	
	75th	Male	3.15×10^{-4}	2.51×10^{-4}	2.87×10^{-4}	3.15×10^{-4}	3.46×10^{-4}	3.99×10^{-4}	0.020
		Female	3.20×10^{-4}	2.54×10^{-4}	2.92×10^{-4}	3.19×10^{-4}	3.51×10^{-4}	4.07×10^{-4}	
	95th	Male	3.62×10^{-4}	2.87×10^{-4}	3.32×10^{-4}	3.64×10^{-4}	3.97×10^{-4}	4.55×10^{-4}	0.029
		Female	3.67×10^{-4}	2.88×10^{-4}	3.34×10^{-4}	3.66×10^{-4}	4.01×10^{-4}	4.76×10^{-4}	

*p-value: Differences between male and female.

IV. Discussion

As a result of Korea's economic growth-oriented policies, the environmental emission of chemicals used in industrial complex has increased dramatically.[31] However, as it became clear that these chemicals had harmful effects on humans, there was an increasing demand for quality of life and concerns about the health of residents around industrial complexes. In this regard, the chemicals might be managed by performing additional monitoring or detailed evaluation after the primary modeling of the screening level in human exposure assessment.

In this study, the concentration of atmospheric benzene predicted using the CalTOX model was 1.08×10^{-4} (95% CI; 1.30×10^{-4} to 9.88×10^{-5}) mg/m^3, and the measured benzene concentration was 1.95×10^{-4} (95% CI; 1.69×10^{-4} to 2.93×10^{-4}) mg/m^3. As the result of the efficiency analysis of the model, the R^2 was 0.9676, it would be explained that benzene (x) emitted from the Daesan industrial complex contributed to the atmospheric concentration (y) by $y=0.002+15.485x$. In addition, the RMSE was 0.0035. These findings suggested that the variance of predicted values would be the good agreements as the variance of observed values.

The data used in this study for source term to air (mol/d) were from National data from the Korean Pollutant Release and Transfer Register. These data set were only collected and discloses the information on the first, second, and third types of industrial facilities in Daesan industrial complex. Emission data collection for industrial facilities of types 4th and 5th with emissions of fewer than 10 tons might be important because the emission is an important input factor and may be underestimated during exposure assessment. Therefore it should be secured for more accurate emission data calculation for future model applications.[19]

The difficulty of quantifying the effectiveness of a model can increase with the complexity and size of the model. Therefore, the possibility of significant uncertainty attached to the many constituent parameters of the model will increase. Such uncertainty may lead to ambiguity in the predictions by the model about the nature of a system.[32] The concern for model validation lies with the use of models in the generic screening process and in other 'data-poor' situations. The results of this study shown that the inhalation exposure to benzene in the atmosphere was the primary risk pathway in humans. Thus, inhalation rates could be considered important in assessing benzene exposure.[33] In this study, it was found that most inhalation rates at percentiles for each activity stage were directly proportional to human exposure. Therefore, it is important to accurately and reliably determine the activity patterns of the population related to the inhalation rate. Risk studies show that preventive efforts are more effective when multiple sources provide consistent and accurate information.[34]

The inhalation rate increased as the percentile value increased according to the activity stage during resting, normal walking, and fast walking, and the inhalation route LADD increased in both men and women. However, inhalation routes LADD in females were found to increase at a higher rate with each increase in inhalation rate than in males. These findings suggest that females may be at a significantly higher risk for certain benzene-related effects as the inhalation rate increases.

Using the CalTOX model to predict human inhalation exposure to benzene, it is very important to choose the probability density function (PDF) format of the input variables.[35] Most of the PDFs of input variables have been assumed to be in the form of a normal or a logarithmic probability distribution in Korea.[36,37] Therefore, in order to perform reliable evaluations, standards or guidelines for PDFs of measurement data and statistical data input variables should be published in Korea.

Previous studies on human exposure assessment to benzene in Korea focused on high-concentration exposure or emission and concentration characteristics of harmful air pollutants around industrial complexes.[17] The evaluation of human exposure to benzene in males and females using a model has been studied in the association between exposure and blood cancer. Currently, researches are needed to predict the residents who will be affected by low-concentration chronic exposure through modeling.[38] In addition, the quantitative data used to validate the model's validity have been insufficient. The general exposure factors in Korean have aided in addressing this, and relevant big data has been collected and disclosed for the application of exposure assessment at the National level.[39] As a result, studies on human exposure prediction using models based on these data should be conducted more actively to provide the necessary information for decision makers to make rational decisions on a scientific basis in evaluating the chronic low-dose exposure of residents in industrial areas.

V. Conclusions

The CalTOX model used in this study predicted the atmospheric concentration of benzene emitted from industrial complex assessed the model's efficiency, and the human exposures based on inhalation rates at various activity stages were prediected. The inhalation rate increased as percentile values increased in each activity stage during resting and normal walking, and the inhalation route LADD increased in both males and females. Compared to the female inhalation rate, the male inhalation route LADD increased at a faster rate with each increase in inhalation rate. As a result of analyzing the increase rate of the LADD_model by regression analysis, $y=2.05x\times10^{-5}+2.27\times10^{-5}$ ($R^2=0.96$) for male and $y=1.98x\times10^{-5}+3.47\times10^{-5}$ ($R^2=0.96$) for female. The LADD of females were higher ratio with an increase in inhalation rate than those of males. This finding would suggest that females may be more susceptible to benzene as their inhalation rate increases comparing to males.

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Conflict of Interest

No potential conflict of interest relevant to this article was reported.

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