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Authors: Tunlathorntham, Supitchaya, and Thepanondh, Sarawut

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Supitchaya Tunlathorntham[1,2] and Sarawut Thepanondh[1,2]

[1]Department of Sanitary Engineering, Faculty of Public Health, Mahidol University, Bangkok, Thailand. [2]Center of Excellence on Environmental Health and Toxicology (EHT), Bangkok, Thailand.

ABSTRACT: The AERMOD dispersion model was evaluated for its performance in predicting 1-hour average nitrogen dioxide (NO_2) concentrations in the vicinity of the largest petrochemical industrial complex in Thailand during the period between January 2012 and December 2013. Measured data from 10 ambient air monitoring stations were intensively used to compare with modeled results. Model results indicated that the tier 1 approach (full conversion of NOx to NO_2) provided the most accurate results compared with other tiers. It also performed very well in predicting the extreme end of NO_2 concentrations. With an absence of emission data from mobile sources, tier 1 was concluded as the most appropriate scheme for prediction of ambient NO_2 ground-level concentrations in this study.

KEYWORDS: AERMOD, nitrogen dioxide, Map Ta Phut, OLM, PVMRM

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CORRESPONDING AUTHOR: Sarawut Thepanondh, Department of Sanitary Engineering, Faculty of Public Health, Mahidol University, Bangkok 10400, Thailand. Email: sarawut.the@mahidol.ac.th

Introduction

A steady change in the atmospheric composition since the commencement of the industrial revolution is mainly due to the combustion of fossil fuels used for the generation of energy and transportation.[1] In Thailand, industrial activities have been promoted by the government with the aim to move the Thai economy forward from developing to developed country. Map Ta Phut industrial area (MA) located in Rayong Province in the eastern region is the largest industrial complex in Thailand. It serves as one of the most important complexes for heavy industries in terms of the production capacities in the Southeast Asia region. Various industrial manufacturing and production processes in this complex include petrochemical industry (48%), metal processing (10%), gas separation (10%), oil refining (2%), electricity generation (5%), chemical product (17%), and other industries (9%).[2] The development of MA has brought out local environmental concerns, particularly air pollution problems. Major air pollutants emitted from this industrial area include sulfur dioxide (SO_2), particulate matter, volatile organic compounds, and nitrogen dioxide (NO_2).[3] To support air quality management in this area, the Thai government declared the MA as a pollution control zone in 2009. This designation requires all the relevant organizations to seek proper measures to limit and control emissions to the environment. NO_2 and SO_2 are air pollutants required by the government for consideration when assessing the impacts of an industrial facility to acquire a permit for operation in this pollution-controlled zone. Furthermore, they are also the parameters that are required to be assessed when planning for future expansion of industrial activities in the MA.[4,5]

Nitrogen oxides (NOx) occur naturally and are also produced by man's activities. In nature, they are a result of bacterial processes, biological growth and decay, lightning, and forest and grassland fires. The primary source of artificial nitrogen oxides is from the burning of fossil fuels.[6] Most of the NOx emissions are in the form of nitric oxide (NO). The amount of nitrogen oxides emitted varies with the temperature of combustion; as temperature increases, so does the level of nitrogen dioxide.[7]

The chemical mechanism of NOx (NO and NO_2) formation during combustion results from hundreds of elementary chemical reactions. Depending on the temperature range, stoichiometric ratio, and type of nitrous species present in the combustion zone, it is possible to distinguish predominant groups of chemical reactions, which are called the mechanisms of nitrogen oxide formation. Usually, the type of flame determines the conditions of the predominant mechanism of NOx formation.[8] Major sources of NOx formation during combustion have 3 recognized mechanisms on NOx formation—thermal, fuel, and prompt.[9] Thermal NOx is produced by the reaction of atmospheric oxygen and nitrogen at elevated temperatures and is reputed to contribute about 20% of the total NOx emission in pulverized coal firing, but is the dominant mechanism when the fuel contains little or no inherent nitrogen (ie, gas firing). Where high air preheat temperatures are used, for example, in cement kilns, thermal NOx can also contribute considerably to the overall NOx emission. Prompt NOx is formed by the reaction of hydrocarbon radicals with atmospheric nitrogen to produce HCN and hence NOx via a complex series of gas phase reactions. The contribution of the prompt NOx to the total emission in pulverized coal combustion is small (about 5%). Measures, which are effective in minimizing thermal and fuel NOx, are also effective in minimizing

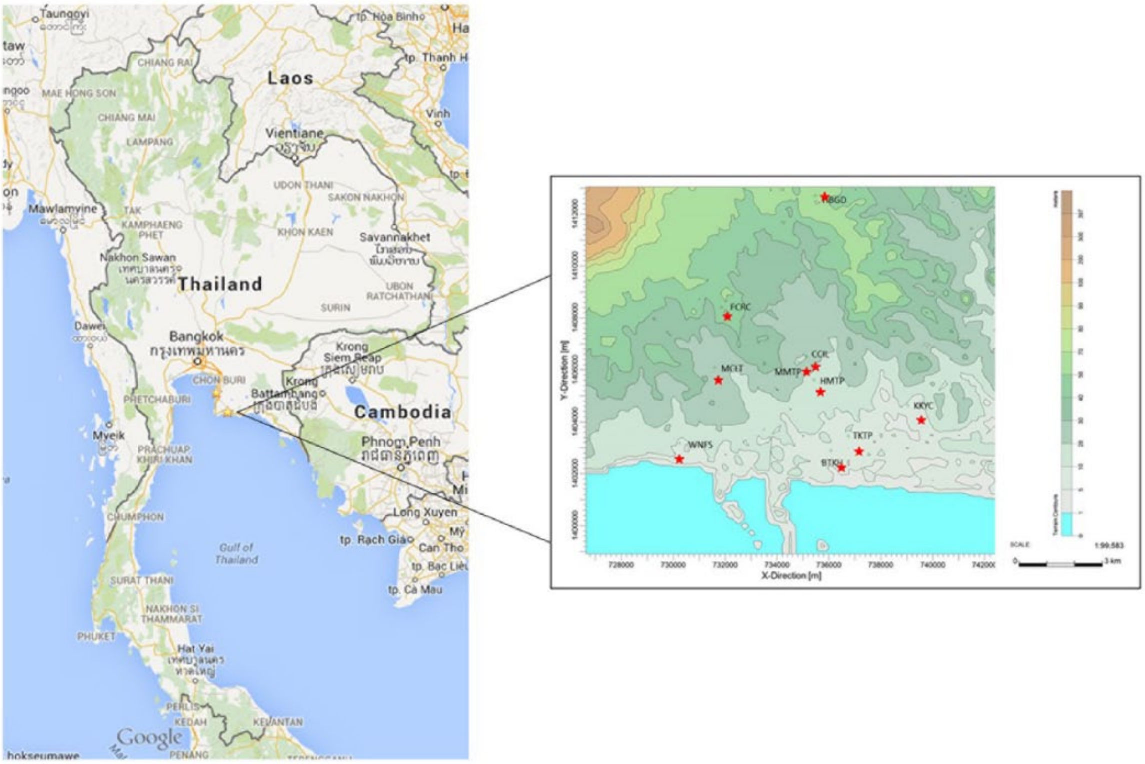

Figure 1. Study area and locations of air quality monitoring stations.
HMTP, health promotion hospital maptaphut; FCRC, field crops research center; BTKH, ban ta kuan public health center; WNFS, wat nong fap school; MMTP, muang mai maptaphut; KKYC, krok yai cha; MCLT, map chalut temple; TKTP, ta kuan temple; HBGD, herbal garden; CCIL, chum chon islam.

prompt NOx. Fuel NOx arises from the reaction of the organically bound nitrogen in the fuel with oxygen. The process is complex (reaction schemes typically consider the order of 50 intermediate species and several hundred separate reversible reactions, there is still considerable uncertainty as to the true value of the various rate constants, etc).[10]

This study presents the method to determine the choices to predict ambient NO_2 concentration emitted as NOx from combustion sources. At ambient temperature and excess oxygen, NO primarily released from thermal process is subsequently oxidized to NO_2. In the presence of tropospheric O_3, NOx can also react with this oxidant and change to NO_2. In this study, 3 different tiers, recommended by US Environmental Protection Agency (US EPA), are tested and evaluated for their ability in predicting NO_2 ambient concentration. Performance of each calculated tiers was assessed by comparing predicted data with intensively measured data from continuous ambient air quality monitoring stations located in the surrounding area of the industrial complex.

Material and Methods

Evaluation of AERMOD performance in this study was conducted by comparing predicted NO_2 concentrations with those measured data in the vicinity of the industrial complex. Hourly NO_2 concentrations measured from 10 ambient air quality monitoring stations were compared with hourly predicted data during the period from January 1, 2012, to December 31, 2013 (2 years).

Study area

The Map Ta Phut industrial complex is located in Rayong Province about 185 km from Bangkok on the Gulf of Thailand's coast (Figure 1). This area consists of 117 industrial plants which include 45 petrochemical factories, 8 coal-fired power plants, 12 chemical fertilizer factories, and 2 oil refineries.[11] The National Economic and Social Development Board of Thailand reported that Rayong is a province with a relatively high gross domestic product (GDP) per capita among all 76 provinces in the country. This high GDP is mainly due to the presence and growth of a large industrial sector.[3]

Emission data

Emission data used in this study were obtained from the Office of Natural Resources and Environmental Policy and Planning (ONEP) database. These data consisted of emission source coordinates, stack heights, exit temperatures, exit velocities, and NOx emission rates. Totally, there were 292 point sources used as emission inputs in this model simulation. It should be noted that NOx emissions in this study were solely derived from stack combustion sources. Emissions from mobile sources in the study area were not included.

Meteorological data

Due to unavailability of upper air data, both of the surface and upper air data were simulated using the MM5 meteorological model for the years 2012 and 2013. Meteorological data contained included hourly wind speed, temperature, cloud cover, ceiling height, surface pressure, and relative humidity. The upper air data included vertical profile of wind speed, wind direction, elevation, temperature, and pressure, which were also simulated using the MM5 model. Data periods read from meteorological data files were started from the first hour in January 1, 2012, to the 24th hour in December 31, 2013.

Tiering options

Three-tier approaches were evaluated for their ability to predict NO_2 ground-level concentrations. Assumptions of each tier can be described as follows:

Tier 1. Total conversion or all NOx = NO_2 (the entire NO component of NOx emission is assumed to instantaneously react and convert to NO_2).

Tier 2. A default NO_2/NOx ratio of 0.60 is applied.[12] This approach assumed that 60% of the NOx emitted from a source are converted to NO_2.

Tier 3. In this approach, O_3 ground-level concentrations are used for calculation of NO_2 ambient concentrations. The calculations were based on the Ozone Limiting Method (OLM) and the Plume Volume Molar Ratio Method (PVMRM). The OLM involves an initial comparison of the estimated maximum NOx concentration and the ambient O_3 concentration to determine the limiting factor of NO_2 formation.[13] The PVMRM determines the conversion rate of NOx to NO_2 based on a calculation of the NOx moles emitted into the plume and the amount of O_3 moles contained within the volume of the plume between the source and receptor.[14] Ambient O_3 concentrations intensively measured in the study areas were used as input data for this approach.

Model configurations

AERMOD dispersion model was used in this study. This model was developed by the American Meteorology Society and the US EPA. AERMOD is designated as a regulatory model by the US EPA. It is a steady-state plume model which assumes that concentrations at all distances during a modeled hour are governed by the temporally averaged meteorology of the hour.[15] The horizontal and vertical distributions in the convective boundary layer are assumed to be Gaussian and bi-Gaussian probability density function, respectively. Using a relatively simple approach, AERMOD incorporates current concepts about flow and dispersion in complex terrain. Where appropriate, the plume is modeled as either affecting or following the terrain. All terrain is handled in a consistent and continuous manner while considering the dividing streamline concept in stably stratified conditions. One of the major improvements that AERMOD brings to the applied dispersion modeling is its ability to characterize the planetary boundary layer through both surface and mixed layer scaling. AERMOD constructs vertical profiles of required meteorological variables based on measurements and extrapolations of those measurements using similarity (scaling) relationships.[16]

The modeling system includes of a key program (AERMOD) and 3 preprocessors: AERMET, AERMAP, and AERSURFACE.[17] The modeling domain in this study was designed for a radius of 10 km (outer grid spacing of $20 \times 20\ km^2$) with the finest grid spacing of 200 m. Urban dispersion coefficient was selected together with the regulatory modeling options for model simulation in this research. Hourly average of NOx emissions and NO_2 concentrations was calculated on elevated terrain height options.

The MM5 model was used to simulate surface and upper meteorological parameters. The gridded data required by AERMET were selected from the Digital Elevation Model (DEM) data. Terrain characteristics were derived from the Shuttle Radar Topography Mission (SRTM3) database.

Evaluation of the model performance

Model performance was evaluated to ensure that the modeling results were accurate.[18] The US EPA proposed a tool to determine the ability of model to ensure that the best model is properly used for each regulatory application. It is also used to confirm that the model is not arbitrarily imposed. Model performance in this study was evaluated through the measures of difference and correlation. Measures of difference quantitatively estimate the size of the differences between observed and modeled data. The association between modeled and observed data is quantitatively determined using the measures of correlation.[19]

To serve this purpose, evaluation of model was conducted for each case using the following statistical parameters: observed mean (O_{mean}), predicted mean (P_{mean}), observed SD/sigma (O_{std}), predicted SD/sigma (P_{std}), fractional bias (Fb), fractional variance (Fs), root mean square error (RMSE), Pearson correlation coefficient (r^2), index of agreement (IOA), and the robust highest concentration (RHC).

Results and Discussion

In this study, the performance of the AERMOD dispersion model in predicting 1-hour average concentration of NO_2 in the vicinity of the largest petrochemical industrial complex in Thailand was conducted for the years 2012-2013. Hourly average ambient ground-level concentrations of NO_2 at each of the monitoring sites were computed. Results were compared with those measured data at the same sites. Statistical analysis of model performance evaluation for each tier is presented in Table 1.

Results from statistical evaluation indicated that there were differences between the predicted and observed values. However, these differences were much lower than

Table 1. Performance evaluation using statistical measures.

MONITORING SITE	NO. OF DATA	MEAN	SD	R^2	RMSE	IOA	FB	FS	RHC
1. HMTP									
Observed	12047	48.83	72.04	—	—	—	—	—	65.31
Tier 1	12047	51.54	33.50	0.98	7.79	0.99	−0.05	0.73	80.41
Tier 2	12047	45.93	101.04	0.87	83.61	0.80	0.06	−0.34	75.56
Tier 3_OL[a]	12047	39.62	25.77	0.99	10.69	0.99	0.21	0.95	62.96
Tier 3_PV[b]	12047	2.41	9.47	0.99	8.79	0.99	1.81	1.54	13.23
2. FCRC									
Observed	10089	34.14	85.45	—	—	—	—	—	48.14
Tier 1	10089	23.98	25.83	0.89	16.12	0.99	0.34	1.07	32.76
Tier 2	10089	19.23	20.33	0.88	16.96	0.99	0.56	1.23	24.86
Tier 3_OL	10089	22.67	23.14	0.91	14.94	0.99	0.40	1.15	30.53
Tier 3_PV	10089	20.35	18.92	0.93	15.16	0.99	0.51	1.28	26.77
3. BTKH									
Observed	10265	41.05	79.49	—	—	—	—	—	55.98
Tier 1	10265	37.81	23.16	0.99	4.89	0.99	0.08	1.10	57.65
Tier 2	10265	27.69	14.22	0.98	15.02	0.99	0.39	1.39	40.62
Tier 3_OL	10265	35.45	20.28	0.99	8.46	0.99	0.15	1.19	53.59
Tier 3_PV	10265	24.15	13.25	0.99	19.17	0.99	0.52	1.43	34.69
4. WNFS									
Observed	10768	25.36	93.83	—	—	—	—	—	37.06
Tier 1	10768	19.25	24.12	0.87	12.31	0.99	0.27	1.18	25.93
Tier 2	10768	15.97	19.88	0.89	11.66	0.99	0.46	1.30	20.58
Tier 3_OL	10768	18.53	21.48	0.91	10.02	0.99	0.31	1.25	24.75
Tier 3_PV	10768	15.89	19.23	0.92	11.36	0.99	0.46	1.32	20.26
5. MMTP									
Observed	8187	49.68	72.01	—	—	—	—	—	68.18
Tier 1	8187	60.60	40.47	0.97	13.33	0.99	−0.20	0.56	84.02
Tier 2	8187	36.74	18.04	0.98	14.88	0.99	0.30	1.19	52.18
Tier 3_OL	8187	51.65	31.93	0.97	5.88	0.99	−0.04	0.77	72.44
Tier 3_PV	8187	25.07	17.68	0.99	25.51	0.98	0.66	1.21	36.92
6. KKYC									
Observed	10513	30.08	90.05	—	—	—	—	—	42.48
Tier 1	10513	24.64	16.57	0.99	7.63	0.99	0.20	1.37	37.26
Tier 2	10513	17.73	16.37	0.99	16.15	0.99	0.52	1.39	25.64

Table 1. (Continued)

MONITORING SITE	NO. OF DATA	MEAN	SD	R^2	RMSE	IOA	FB	FS	RHC
Tier 3_OL	10 513	22.30	16.10	0.98	10.44	0.99	0.29	1.39	33.39
Tier 3_PV	10 513	18.62	16.64	0.99	14.52	0.99	0.47	1.38	27.16
7. MCLT									
Observed	9774	26.17	92.93	—	—	—	—	—	37.35
Tier 1	9774	18.63	23.09	0.88	12.29	0.99	0.34	1.20	24.96
Tier 2	9774	16.07	20.35	0.91	11.78	0.99	0.48	1.28	21.08
Tier 3_OL	9774	18.77	21.54	0.92	10.27	0.99	0.33	1.25	25.31
Tier 3_PV	9774	16.48	18.97	0.94	10.94	0.97	0.45	1.32	21.72
8. TKTP									
Observed	1778	34.88	83.59	—	—	—	—	—	42.39
Tier 1	1778	63.96	38.58	0.98	31.02	0.95	−0.58	0.74	78.28
Tier 2	1778	37.38	11.94	0.99	2.88	0.99	−0.07	1.50	45.69
Tier 3_OL	1778	54.74	27.48	0.99	20.34	0.98	−0.44	1.01	66.65
Tier 3_PV	1778	33.33	12.88	0.99	3.71	0.99	0.05	1.47	42.87
9. HBGD									
Observed	10 661	58.97	67.36	—	—	—	—	—	84.41
Tier 1	10 661	45.89	39.32	0.99	14.59	0.99	0.25	0.53	72.71
Tier 2	10 661	30.67	22.67	0.99	30.30	0.96	0.63	0.99	46.78
Tier 3_OL	10 661	39.81	32.59	0.99	18.85	0.98	0.39	0.69	62.37
Tier 3_PV	10 661	24.66	22.23	0.98	36.48	0.95	0.82	1.01	36.43
10. CCIL									
Observed	2428	22.52	95.84	—	—	—	—	—	28.69
Tier 1	2428	36.76	30.01	0.99	7.26	0.99	0.05	0.93	89.66
Tier 2	2428	28.44	13.08	0.95	16.29	0.99	−0.52	1.52	48.16
Tier 3_OL	2428	61.95	34.06	0.94	39.96	0.93	−0.93	0.95	76.64
Tier 3_PV	2428	28.68	18.69	0.98	11.62	0.99	−0.24	1.35	42.83
All stations									
Observed	86 510	38.74	82.65	—	—	—	—	—	60.24
Tier 1	86 510	36.76	30.01	0.99	7.26	0.99	0.05	0.93	58.26
Tier 2	86 510	24.84	18.54	0.99	15.42	0.99	0.44	1.27	37.97
Tier 3_OL	86 510	32.16	24.91	0.99	7.67	0.99	0.19	1.07	50.48
Tier 3_PV	86 510	21.78	17.61	0.99	18.59	0.98	0.56	1.29	32.98

Abbreviations: Fb, fractional bias; Fs, fractional variance; IOA, index of agreement; RHC, robust highest concentration; RMSE, root mean square error; r^2, Pearson correlation coefficient; HMTP, health promotion hospital maptaphut; FCRC, field crops research center; BTKH, ban ta kuan public health center; WNFS, wat nong fap school; MMTP, muang mai maptaphut; KKYC, krok yai cha; MCLT, map chalut temple; TKTP, ta kuan temple; HBGD, herbal garden; CCIL, chum chon islam.
No. of data were based on availability of measured data. Unit of concentration is μg/m^3.
[a]Tier 3_OL = tier 3_OLM
[b]Tier 3_PV = tier 3_PVMRM.

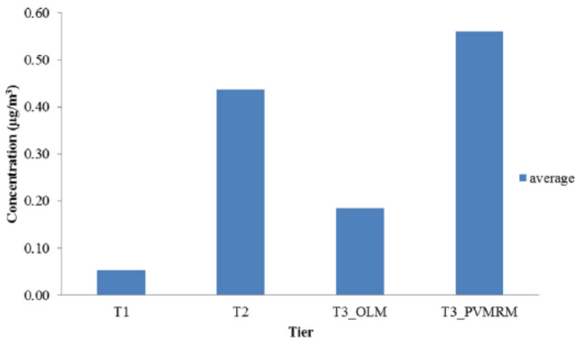

Figure 2. Performance evaluation of fraction bias (tiers 1-3).

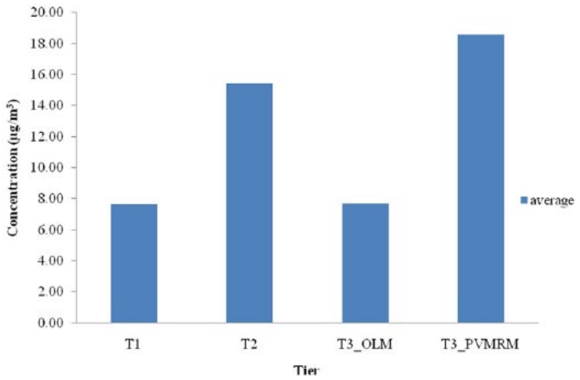

Figure 3. Performance evaluation of root mean square error (tiers 1-3).

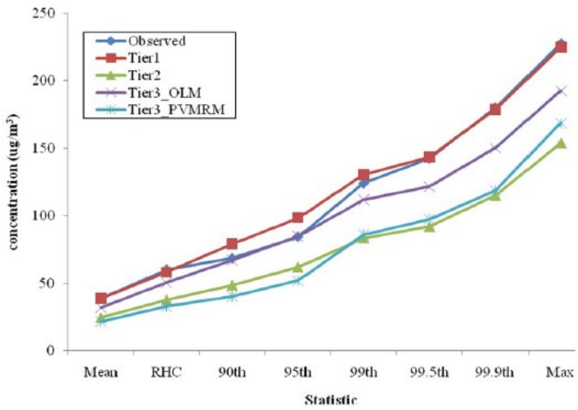

Figure 4. Annual mean, maximum, robust highest concentration (RHC), and percentile statistics for modeled and observed NO_2 for all sites.

their respective standard deviations (sigma) (RMSE < standard deviation), indicating that accurate results were being shown by the model. Generally, AERMOD performed well for the prediction of average concentration at every monitoring site, at least within the accuracy of the observations (standard deviation) for every tiering options.

The Fb and Fs values varied between −2 (extreme overprediction) and +2 (extreme underprediction). It was found that Fb and Fs values for all tiers were negative, indicating overprediction. The good model performance can be interpreted when the values of these parameters are near 0. The highest Fb and Fs values were calculated for modeled data at every stations in tier 3_PVMRM (Fb = 0.56), whereas the best model performances were found in tier 1 (Fb = 0.05), as shown in Figure 2. The lowest value of RMSE (7.26 µg/m³) was also obtained from simulation results under tier 1 (Figure 3). These findings supported the ability of tier 1 in predicting overall concentrations of NO_2 in this study.

The RHC is preferred to the actual peak value and represents a rounded estimate of the highest concentrations, based on a tail exponential fit to the upper end of the distribution. With this procedure, the effect of extreme values on model comparison is reduced.[20] Results from the RHC revealed that tier 1 gave the best result in predicting the extreme end of NO_2 ambient concentration (Figure 4). The robust highest concentration of measured data (combining all receptors) was 60.24 µg/m³, whereas predicted results from tier 1, tier 2, tier 3_OLM, and tier 3_PVMRM are 58.26, 37.97, 50.48, and 32.98, respectively. This finding indicated that tier 1 provided the best result in an attempt to predict episodes of air pollution in this study.

The maximum ground-level concentrations of NO_2 within the modeling domain were also predicted for each tier. It should be noted that the values at each receptor (10 monitoring sites) did not exceed the Thai's ambient air quality standards (NO_2 < 320 µg/m³ for 1-hour average). However, the maximum NO_2 concentrations predicted within modeling domain were greater than the standard values for all simulated tiers. This finding depicts the importance of siting an appropriate location of ambient air monitoring station by considering not just only individual factory but also as an area-based emission for better management of air pollution in this industrial area. Spatial distributions of NO_2 simulated map are as presented in Figures 5 to 8.

Conclusions

The AERMOD dispersion model was used to predict the concentrations of ambient NO_2 emitted from stack combustion sources in the MA, Thailand. Evaluation of the model's performances was conducted by comparing predicted data with those measured concentrations over the period of 2 years (from January 1, 2012, to December 31, 2013). AERMOD was simulated using local emission sources, terrains, and meteorological characteristics within the study domain. The model predicted 1-hour average concentrations of ambient NO_2 at 10 receptors where there were intensive NO_2 monitoring stations installed to serve the model's validation purpose. A total of 292 stack combustion sources were accounted as emission inputs for the simulation of the model. The models were tested for its performance under 3 different scenarios to evaluate the most appropriate approach for further application in environmental impact assessment both in this industrial zone and in other areas. The performance of the model

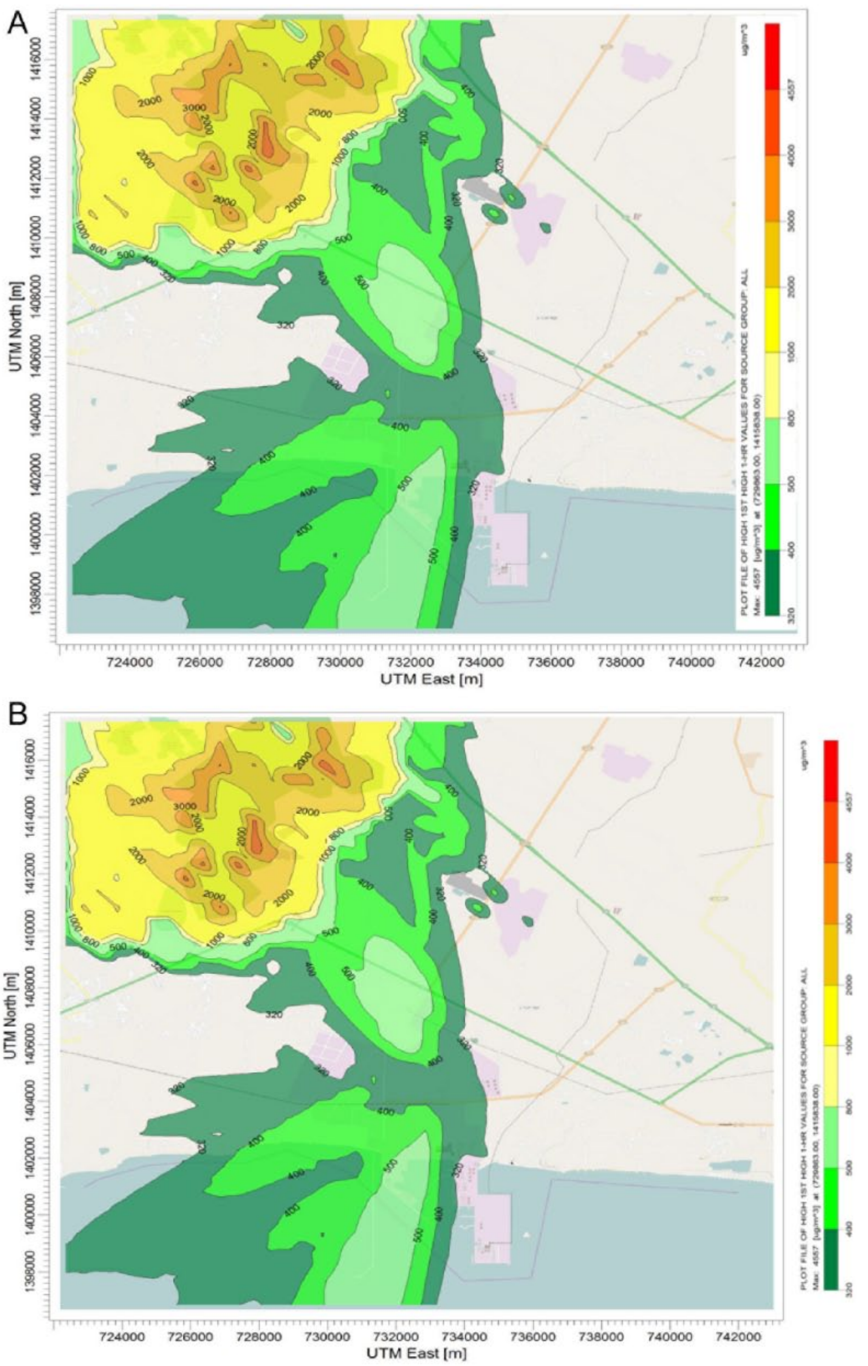

Figure 5. Plot file of the first highest 1-hour averages of tier 1 in the years (A) 2012 and (B) 2013.

was evaluated through the measures of difference and correlation between observed and modeled data using statistical analysis.

The tier 1 approach (100% conversion of NOx to NO_2) resulted in the highest predicted value of NO_2 concentrations. Overall predicted results obtained from tier 1 were shown to have less bias with those measured results compared with other tiers. It is also the best option to determine the maximum ground-level concentration as depicted by its ability to predict the extreme end concentration of NO_2 in this study. This finding indicated that tier 1 can be considered as the most appropriate simulation scheme in the prediction of annual concentration of NO_2 in this industrial area. Results from this study revealed the fact that emission inventory of oxide of nitrogen may be underestimated. NO_2 concentrations are contributed by emissions from both industrial and mobile source emissions. However, lack of emission data for mobile sources occurring in many areas constrains the application of air dispersion models in such areas. Efforts in using the background concentration of NO_2 to compensate mobile source contribution still do not overcome this problem. In the

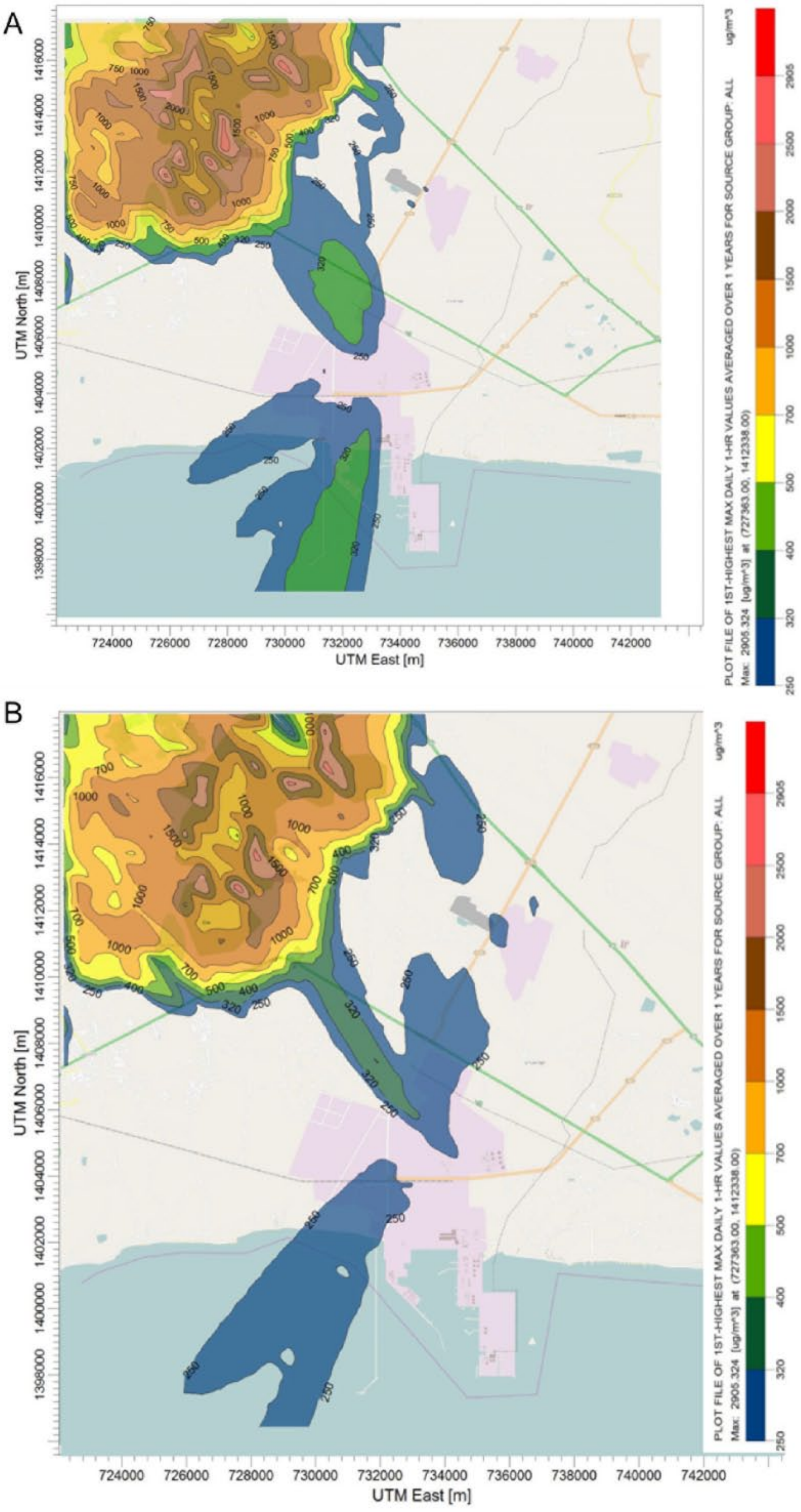

Figure 6. Plot file of the first highest 1-hour averages of tier 2 in the years (A) 2012 and (B) 2013.

presence of O_3 data, the behavior of NO_2 should be more refined. Therefore, tier 3 which involves chemistry of O_3 and NOx has been developed to explain the characteristics of atmospheric chemistry of those pollutants once emitted from emission sources. However, this latest tier cannot perform well when emissions of NOx are underestimated. Therefore,

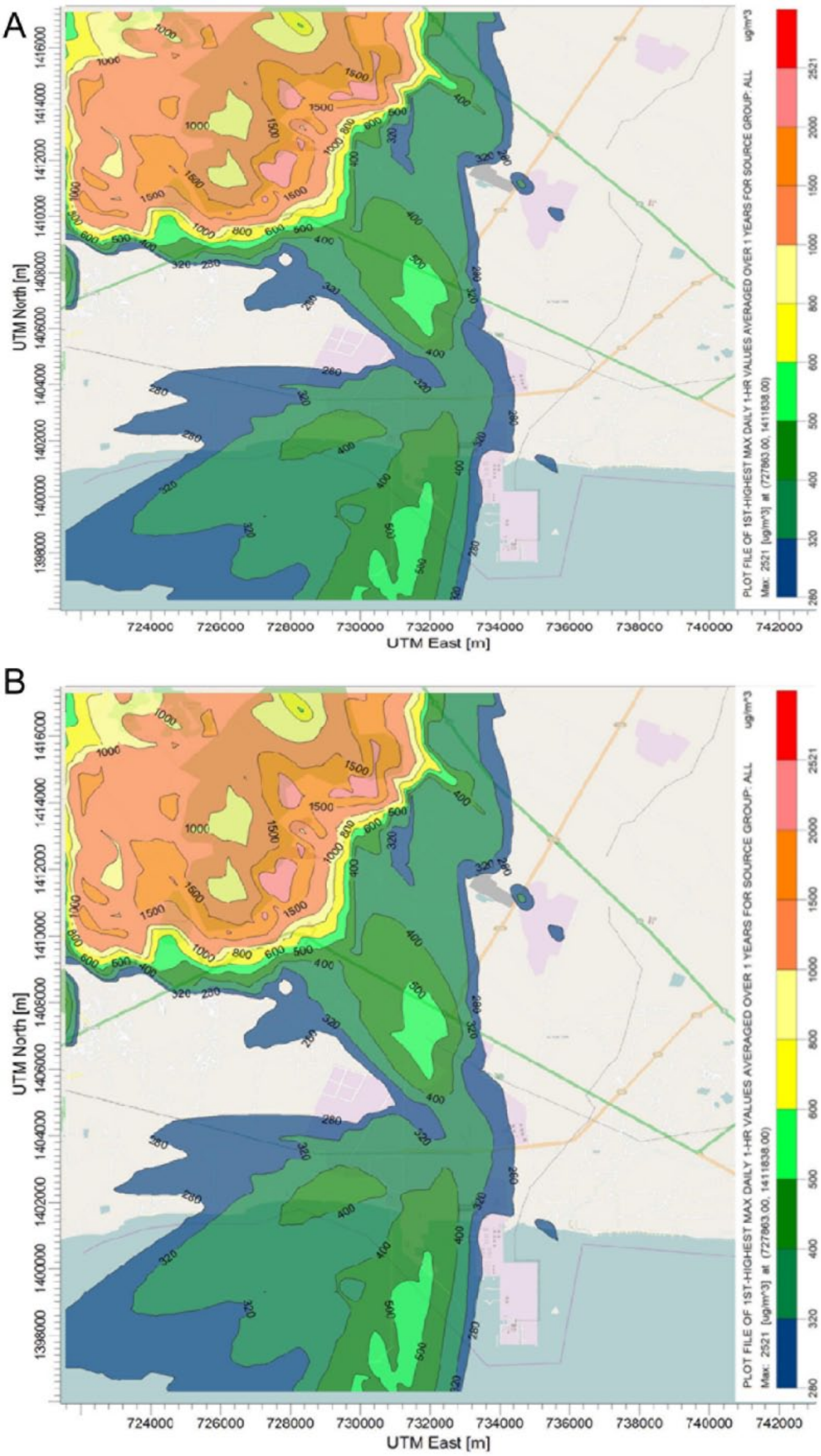

Figure 7. Plot file of the first highest 1-hour averages of tier 3_OLM in the years (A) 2012 and (B) 2013.

availability of input data is the most crucial factor when considering types and options of model simulated in each area. The other approach to support this limitation is the use of both NO and NO_2 ambient concentrations measured at the receptors to compare with predicted data rather than using only NO_2 ambient concentrations. This analysis could assist to determine the extent of NO_2/NOx concentration which is the specific value in each area.

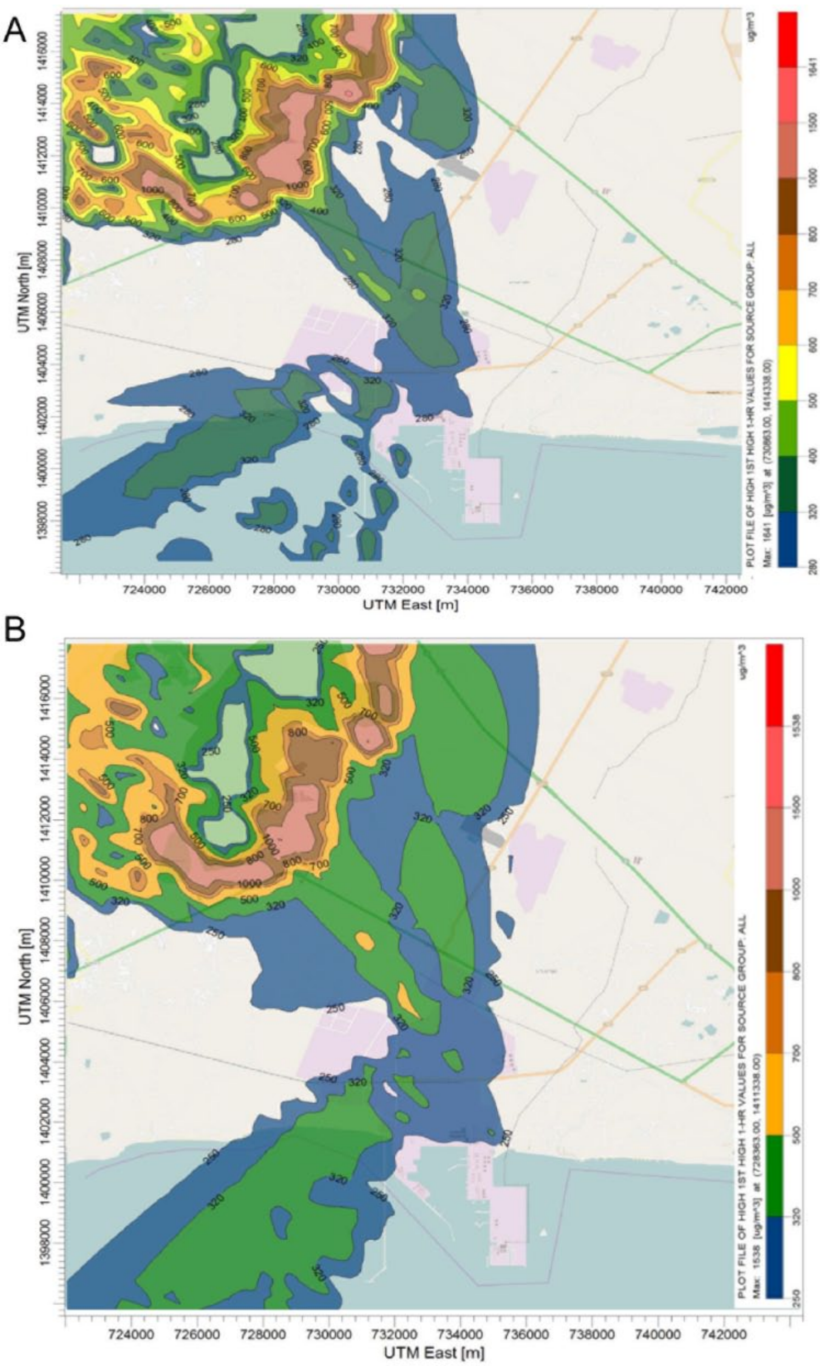

Figure 8. Plot file of the first highest 1-hour averages of tier 3_PVMRM in the years (A) 2012 and (B) 2013.

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Author Contributions

ThS conceived and designed the experiments, contributed to the writing of the manuscript, agree with manuscript results and conclusions, and jointly developed the structure and arguments for the paper. TuS analyzed the data and wrote the first draft of the manuscript. ThS made critical revisions and approved the final version. All authors reviewed and approved the final manuscript.

Disclosures and Ethics

As a requirement of publication, authors have provided to the publisher signed confirmation of compliance with legal and ethical obligations including, but not limited to the, following: authorship and contributor ship, conflicts of interest, privacy and confidentiality, and (where applicable) protection of human and animal research subjects. The authors have read and confirmed their agreement with the ICMJE authorship and conflict of interest criteria. The authors have also confirmed that this article is unique and not under consideration or published in any other publication, and that they have permission from rights holders to reproduce any copyrighted material. Any disclosures are made in this section. The external blind peer reviewers report no conflicts of interest.

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