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Change of Odor Distribution due to Improvement of Wastewater Treatment Plants in a Tapioca Starch Industry

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Abstract

Odor nuisance is considered as one of the major environmental problems which deteriorate the quality of life. In this study, the emissions and spatial distributions of acetaldehyde which is the major chemical released from the wastewater treatment of tapioca starch factory is evaluated. Emission rate of acetaldehyde was evaluated by direct measurement through the chamber experiment. Results were further used as input data to evaluate the extent and magnitude of spatial distribution of acetaldehyde in the vicinity of the factory. Short-term peak concentrations (over 3 min) derived from one-hour modeled concentrations were also used to assess the odor perception. Quantitative analysis of the existing status of the problem under the business as usual of the factory (opened-lagoon) was compared with the modified covered-lagoon. Results clearly indicated the success of this measure toward the reduction of the extent and magnitude of odor problem.

Keywords: Odor; Peak to mean; Spatial distribution; Tapioca starch industry; Wastewater treatment plants

1. Introduction

Tapioca starch production has played an important role among the agro-industries to the Thai economy (DOA, 2015). Tapioca is the third most important agricultural crop in Thailand, after rice and sugarcane. However, the growth of the tapioca starch industry has resulted in environmental pollution (Chavalparit and Ongwandee, 2009). The starch extraction process requires a huge volume of water which in turn produces large amount of wastewater with the high organic loading (Tanticharoen and Bhumiratanatries, 1995; DIW, 2006). Generally, wastewater that contains a high level of organic contents requires a very specific treatment processes. It is often unfortunate to use only one typically treatment processes but rather requires more than one procedure to efficiently remove organic contents and pollutants (Raj and Anjaneyulu, 2005).

Wastewater treatment plants (WWTPs) of

tapioca industry have been identified as one of the major VOCs emission sources (Hall, 1997; Cheng *et al*., 2008; Wimolrattanasil *et al*., 2018). There is a growing concern regarding the volatile organic compounds (VOCs) emissions from the WWTP released to the environment (Oyama and Hunter, 2000; Hamoda, 2006). Volatile organic compounds (VOCs) are regulated aerial pollutants that have environmental and health concerns. Many of them contribute to malodor from the WWTP (Ni, 2015).

Quantifying the impact of odor on public health and quality of life of citizens is gaining increasing importance (Wing *et al*., 2008). The concentration of malodor in the air is often monitored and controlled with the aim of complying with odor regulations while keeping a respectable public image of the emission sources. Odor can be defined as a "sensation associated with one or more compounds which, when present in sufficiently high concentrations, can trigger olfactory responses in exposed individuals" (Nicell, 2009). Since odor responses can be instantaneously induced, odor peaks may create more annoyance than longer lasting odor emissions, due to human olfactory adaptation (Both *et al*., 2004; Alfosin *et al*., 2015).

Air quality modeling is a tool that can apply for calculate the chemical dispersion. The AERMOD model has been widely used in numerous studies, including dispersion analyses for various air pollutants, such as particulate matter (PM), NOx, SO2 and other pollutants (Kakosimos *et al*., 2011; Seangkiatiyuth *et al*., 2011; Thawonkaew *et al*., 2015 and Truong *et al*., 2016). Dispersion models usually calculate ambient concentrations over an integration time of up to one hour. When applied to odor pollution, a modification is needed to somehow account for the ability of the human nose to perceive odor within a single breath; in other words, a peak-to-mean concept is needed (Piringer *et al*., 2016). This dispersion model calculates one hour mean values. Odor-hours are derived by multiplying the hourly mean value with a constant factor of concentration. The assessment of maximum values for shorter periods than one hour is not only relevant for environmental odor but also for toxic and inflammable pollutants. This estimation by one hour mean values can lead to an underestimation of the ambient air concentration. This error depends on the observed impact of the ambient concentration (Schauberger and Piringer, 2012).

Wimolrattansil *et al*. (2018) found that the major volatile organic compounds emitted from this tapioca wastewater treatment plant is acetaldehyde and its odor characteristic is very strong pungent also easily to volatile and detectable (Table S1). With respect to this circumstance, the main objective of this study was to compare the spatial dispersion of acetaldehyde under different scenarios by using predicted concentrations in term of the maximum 1 h and peak values. Results were discussed for their impacts on odor annoyance for the people who live around the vicinity. Finding from this study will clarify the magnitude of polluted areas and odor nuisance problems of the tapioca factory.

2. Materials and methods

2.1 Study site

The large tapioca industry located at Chanthaburi province in the Eastern region of Thailand was chosen as study site (Figure 1). The total capacity of the starch plant is 400 t/d. Approximately 3,000 m^3/d day of wastewater produced from starch production are treated in this wastewater treatment plant. The starch process aims to use 3,000 m^3 of the wastewater as well as additional 100 t of root cake/d as substrate for the biogas production for electricity generation.

2.2 Characteristics of the wastewater treatment process

Schematic diagram of WWTP processes are shown as Figure 2. Sump tank is the first process of treatment plant, use for storing wastewater release from starch production process. Modified covered lagoon treats wastewater and generates biogas. The technology involves covering an open lagoon with a High Density Poly Ethylene (HDPE) sheet. At the

bottom of the lagoon, stirrers are fixed as in a Continuous Stirred Tank Reactor (CSTR) in order to facilitate proper mixing of pulp and wastewater. This design is a combination of an anaerobic covered lagoon and a CSTR. Hence the type of wastewater treatment is called as Modified Covered Lagoon (MCL). Characteristics of wastewater and MCL are presented as scenario 2 in Table 1.

2.3 Direct measurement of VOC emissions

On-site chamber experiment was conducted adjacent the sump tank for direct measurement of acetaldehyde emissions from WWTPs. The homogeneous wastewater from the sump tank were transferred to the tray. Ten air samples are collected from experimental chamber; daytime (4 samples) and nighttime (6 samples). During sampling time, the tapioca factory has normally operates starch production. The chamber (0.064 m^3 volume) has a continuous ventilation rate of 41.09 m^3/h (Figure 3). The volumetric flow rate used for the calculation of emission rate was about 0.0114 m^3/s. In this study, wastewater from the sump tank were transferred to the tray (0.071 m2) located inside the chamber. Air samples were simultaneously collected for 3 h using 6 L evacuated canisters (0.05 mmHg) and analyzed in accordance with the U.S. EPA TO-15 method for the determination of toxic organic compounds. Acetaldehyde samples were introduced into the canisters then pressurized in order to prevent the contamination entering the sample canister. The quality control program followed during this study included field blanks, spiked samples, and duplicate measurements. Field blank samples were transported along with the sampling canisters to the site and analyzed to ensure that there were no contaminations from the sample collection, transportation, or storage. The cleaned canister was tested by pressurizing with humidified nitrogen and analyzed by a GC/MS. The criteria for the cleaned canister was proven reliable based on the criteria for individual target compounds to be <0.2 μg/L. Any canister that did not meet the criteria was re-cleaned. The performance of the entire analytical system was checked by means of analyzing blanks and canisters with known analyte concentrations. A new calibration curve was obtained each time. Standard canisters were prepared by spiking the standard mixtures into clean canisters and exposing them to a nitrogen flow for 300 s in order to evaporate the solvent. One quality control canister was also measured for every ten samples to check the recovery of the analyzed compounds. All the samples were analyzed with two replications in order to test the precision of the sampling and analyzing techniques. The relative standard deviation mean of duplicates was within 10% for the target compounds. Detection limits were defined as three times the standard deviation of the blank values. Quantification limits were defined as ten times the standard deviation of the blank values (Thepanondh *et al.*, 2011; Saew and Thepanondh, 2015). Air samples were transferred to the thermal desorption unit, working as a pre-concentrator prior to being sent to gas chromatography/mass spectrometer (GC/MS) (Agilent Technologies Model 7890B GC).

Concentrations of acetaldehyde were quantified by GC/MS. In order to estimate acetaldehyde emission rate, a fixed-box model was applied (O'Shaughnessy, 2011; Schauberger *et al.*, 2013). In the steady state condition, acetaldehyde evaporated from the container will be diffused throughout the box. Hence, concentrations of acetaldehyde within the box are constant and can be used to calculate for the emission rates. Emission rates of acetaldehyde were calculated by using its chemical concentrations multiply with chamber ventilation rate. Emission rates of acetaldehyde were expressed in the unit of μg/s.

2.4. Dispersion evaluation

The vendor-supplied modeling package (AERMOD ViewTM ver.9.3, Lakes Environmental, Waterloo, Ontario, Canada) was used for emissions concentrations estimation in this study. The AERMOD was simulated using an Intel(R) Core(TM) i5-2320 CPU at 3.00 GHz. AERMOD model was simulated to predict the maximum hourly concentrations of VOCs emitted from wastewater treatment plant.

Table 1. Descriptions of model scenario and characteristics of wastewater treatment system

Characteristic/Scenario	Scenario 1 (Before the WWTP improvement)	Scenario 2 (After the WWTP improvement)
Type of WWTP	Open anaerobic lagoon; OAL	Modified covered lagoon; MCL
Size of open-area	50,286 m^2	286 m^2
Volume of digester	150,715 m^3	150,000 m^3
COD of the wastewater	31,156 mg/L	31,156 mg/L
Average wastewater flow	3,000 m^3/d	3,000 m^3/d
Methane content in biogas	-	60%

Spatial distributions of the concentrations were compared with the odor threshold in order to identify the magnitude of the odor nuisance.

-AERMOD model configuration

The meteorological data were prepared over one year (1st January 2015 to 31th December 2015). Data used in this study were generated by the Mesoscale Meteorological Model (MM5). Data were then pre-processed using AERMET processor. The gridded data needed by AERMAP was selected from the Digital Elevation Model (DEM) data and collected during the Shuttle Radar Topography Mission (SRTM3). In this study, the model was simulated over the area of 6×6 km2 centered at the wastewater treatment plant to predict ambient concentrations. The Cartesian receptor grid has a uniform spacing of 50 m.

AERMOD simulations were performed to predict the maximum concentration of acetaldehyde and its spatial distribution over the study domain during the year. Results from the model simulations were illustrated as the pollution map using the maximum 1-h concentration values (U.S. EPA, 2004).

- Scenario analysis

Estimated emission rate of VOCs in this study was used as input data of AERMOD dispersion model. The model was simulated under 2 scenarios, the first one was the business as usual where the type of wastewater treatment plant was open anaerobic lagoons system (OAL) and the second one was after some improvement where the type of wastewater treatment plant was modified covered lagoons system (MCL). Basically, the differences between these scenarios were the size of evaporation area. The study scenarios are presented in Table 1.

2.5 The peak-to-mean concept

For the assessment of peak values, describing the biologically relevant exposure, often so called peak to mean concept is used. The assumption of a constant peak to mean factor can only be used as a very rough estimate. Short-time peak concentrations derived from one-hour mean values, to mimic odor sensation of the human nose. The short-term peak concentrations required for the assessment of odor perception. Given a mean concentration over one hour, the mean value of a shorter period can be calculated using the well-known relationship (Smith, 1973). With the mean concentration, Cm, calculated for an integration time of Tm and the peak concentration Cp, for an integration time of Tp. The peak-to-mean factor is given by:

$$C_p/C_m = (T_m/T_p)^u \quad (1)$$

Where: *Cm* is the measure of mean concentration over an averaging time scale *Tm* for which meteorological conditions are persistent, while *Cp* is the expected peak concentration, averaged over time *Tp*, during the time *Tm*. *Cm* is typically a ten minute or one hour average and is routinely and reliably modelled. *U* is the constant; this study use 0.2 for the calculation (Schauberger and Piringer, 2012).

The peak concentration values of acetaldehyde were calculated from the equation

Figure 1. Location of tapioca factory (study site)

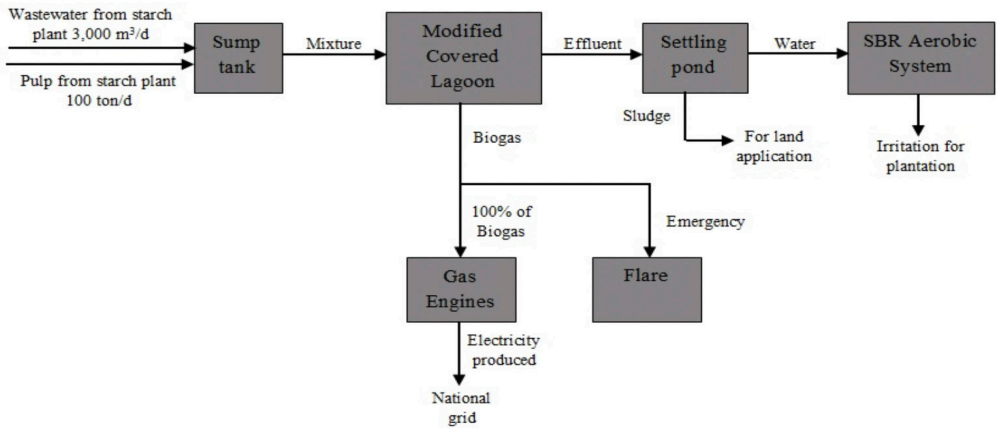

Figure 2. Schematic diagram of wastewater treatment processes (MCL)

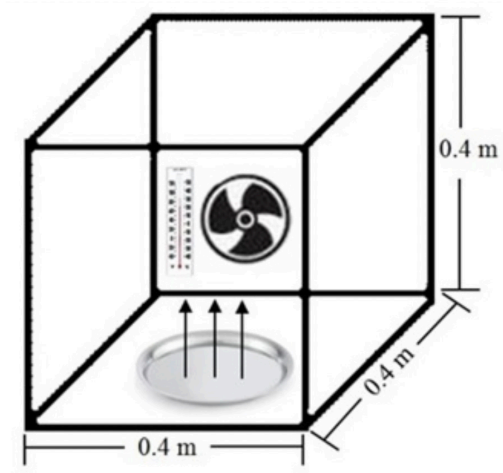

Figure 3. Chamber experiment study

under the study scenarios and then evaluated the spatial distribution by AERMOD model.

3. Result and discussion

During the wastewater treatment process, volatile organic compounds can release from liquid-phase to gas-phase. The amount of VOCs emitted to the atmosphere depending upon biological, physical, chemical and atmospheric conditions. The emission rates of emitted VOCs in this study (Table S2) were coincided with those from previous studies (Atasoy *et al*., 2004; Wu *et al*., 2006; Yang *et al*. 2012). It should be noted that these emission rates were also fluctuated during daytime and nighttime. Higher emission rates of VOC from wastewater during daytime were found in this study than those during nighttime period. Temporal variations of the emission rate of volatile organic compounds are greatly depend on atmospheric and meteorological conditions such as wind speed, temperature and humidity. In this study, acetaldehyde emission is highest among other compounds (Table S2).

3.1 Predicted maximum and calculated peak concentration of acetaldehyde

Based on the chamber experiment and modeled simulation described above, the predicted concentration of acetaldehyde (78.08 μg/m2/s) can be obtained. In this study, the predicted maximum and calculated peak concentrations of acetaldehyde were elucidated under two different scenarios, scenario 1 before the WWTP improvement and scenario 2 after the WWTP improvement.

Table 3.1 presents the predicted maximum 1h concentration of acetaldehyde under two different scenarios. The average, lowest and highest concentrations were illustrated in this table. Predicted maximum concentrations were illustrated under scenario 1 and 2. Scenario 1; before the wastewater treatment plant improvement, the range of maximum concentration was 3.8-35,286 μg/m^3. As for scenario 2; after the wastewater treatment plant improvement, the range of maximum concentration was 0.2-9,303 μg/m^3.

Equation 1 was applied to calculated peak concentration which means a distinct odor perception over several breaths and the peak concentrations were evaluated under scenario 1 and 2 as shown in Table 3.2. Scenario 1; before the wastewater treatment plant improvement, the range of peak concentration was 7.0-64,241 μg/m^{3}can be expected. Under scenario 2; after the wastewater treatment plant improvement, the range of peak concentration was 0.3-16,937 μg/m^3.

Even though, the wastewater treatment plant of this tapioca factory has changed to the modified covered lagoon. The average maximum and peak concentration of acetaldehyde under scenario 2 were higher than the odor threshold (450 μg/m^3). The highest of predicted maximum and calculated peak concentration were higher than odor threshold more than 78 times and 142 times, respectively. Their predicted maximum and calculated peak concentrations of acetaldehyde were exceeded the odor thresholds which these results designate that the odor nuisance to the residential living in the vicinity of this tapioca factory. However the magnitude of odor was significantly decreased and the amount of people who affect this odor nuisance problem were also lessen due to the improvement of the WWTP.

3.2 Spatial distribution of acetaldehyde

For scenario 1 (the case of before the WWTP improvement), the highest value of maximum average 1-h concentration of acetaldehyde over the modeling domain during the simulation period was 35,287 μg/m^3. For scenario 2 (the case of after the WWTP improvement), the highest value of acetaldehyde over the modeling domain decreased to 9,646 μg/m^3. The maximum spatial distributions of acetaldehyde were illustrated in Figure 4.1 and 4.2.

As for the peak concentration, the case of before the WWTP improvement (scenario 1), the highest value of peak concentration of acetaldehyde over the modeling domain during the simulation period was 64,241 μg/m^3. As for scenario 2 (after improvement of the WWTP) the highest peak concentration was significantly

decreased to 17,560 μg/m^3. Spatial distributions of the peak concentration during 3 min for both scenarios are illustrated in Figure 5.1 and 5.2. Totally, there were 15,626 grid cells in the modeling domain. There were 6,717 and 189 grid cells having predicted maximum concentrations higher than odor threshold of acetaldehyde under scenario 1 and 2, respectively. The number of exceedance grid cells were expected to increase to 10,443 (scenario 1) and 379 (scenario 2) grid cells when the peak concentrations were estimated. Results under this study revealed that the affected area was significant decreased more than 96% from the business as usual scenario (scenario 1) as a result from modification of the wastewater treatment from it opened lagoon to the modified covered lagoon.

Table 3.1 Predicted maximum 1-h concentration of acetaldehyde under two scenarios

Scenario	concentration (μg/m^3)	
	Average	Range
1	771.3	3.8-35,286
2	44.9	0.2-9,303

Table 3.2 Calculated peak concentration of acetaldehyde under two scenarios

Scenario	concentration (μg/m^3)	
	Average	Range
1	1,404.1	7.0-64,241
2	81.8	0.3-17,560

Table S1 The most volatile organic compounds emitted from tapioca wastewater treatment plant

VOCs	Concentration (μg/m^3)	Emission rate (μg/m^2/s)	Odor
Acetaldehyde	483.87	78.08	Strong pungent
Acetone	422.88	68.23	Fruity
Isopropyl alcohol	6.97	1.13	Mild
2-Butanone	33.93	5.47	Sweet

Remark: The concentrations and emission rates are daytime values used in model application

Table S2 The concentrations and emission rates of 21 volatile organic compounds emitted from tapioca wastewater treatment plant

VOCs	Daytime		Nighttime	
	Concentration (μg/m^3)	Emission rate (μg/m^2/s)	Concentration (μg/m^3)	Emission rate (μg/m^2/s)
Acetaldehyde	483.87	78.08	155.20	25.04
Acetone	422.88	68.23	181.27	29.25
Acrylonitrile	0.07	0.01	0.08	0.01
Benzene	0.15	0.03	0.09	0.02
Carbon disulfide	0.18	0.03	0.10	0.02
Carbon tetrachloride	0.02	<0.01	0.02	<0.01
Chloroform	0.12	0.02	0.06	0.01
Chloromethane	0.13	0.02	0.10	0.02
Cyclohexane	0.02	<0.01	0.02	<0.01
Dichloromethane	0.39	0.06	0.27	0.04
Hexane	0.02	<0.01	0.02	0.03
Isobutane	0.16	0.03	0.06	0.01
Isoprene	0.07	0.01	0.09	0.01
Isopropyl alcohol	6.97	1.13	12.74	2.06
Methacrolein	0.56	0.09	0.22	0.04
Pentane	0.15	0.03	0.08	0.01
Propene	0.40	0.06	0.11	0.02
Toluene	0.17	0.03	0.11	0.02
1,2-Dichloroethane	1.13	0.18	0.35	0.57
2-Butanone	33.93	5.47	14.19	2.29
3-Pentanone	0.85	0.14	0.28	0.05

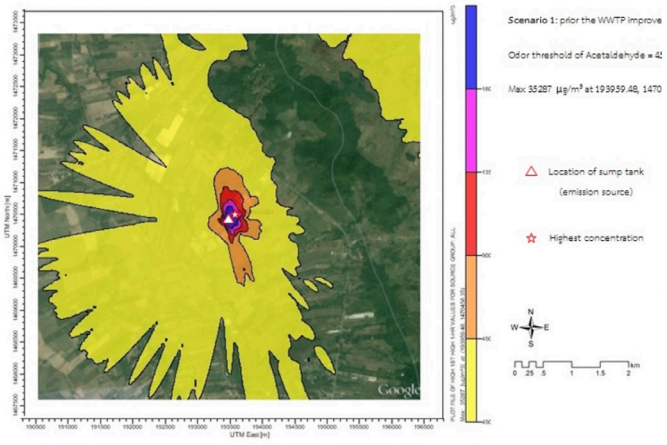

Figure 4.1. Spatial distribution of maximum (1 h) concentration of acetaldehyde under scenario 1

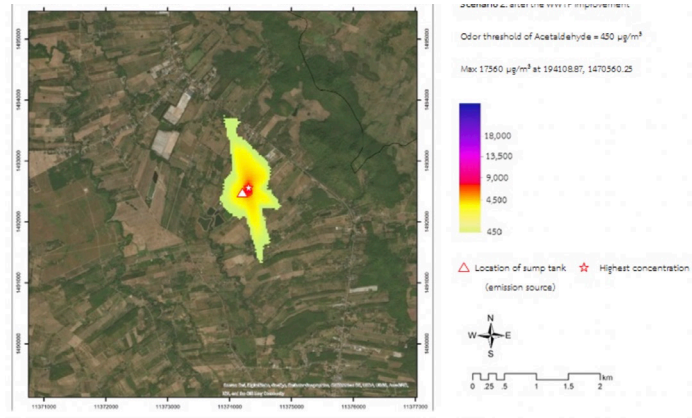

Figure 4.2. Spatial distribution of maximum (1 h) concentration of acetaldehyde under scenario 2

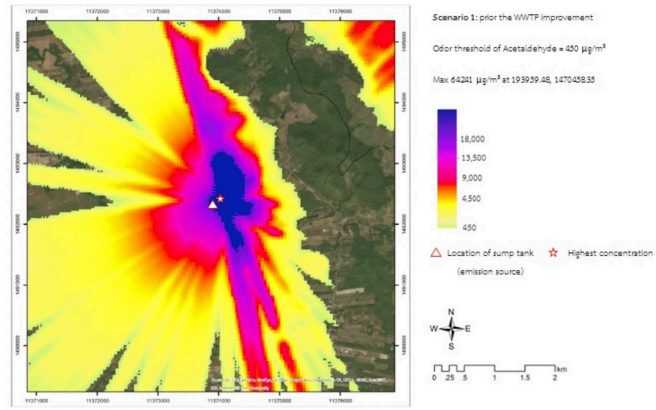

Figure 5.1 Spatial distribution of peak concentration (3 min) of acetaldehyde under scenario 1

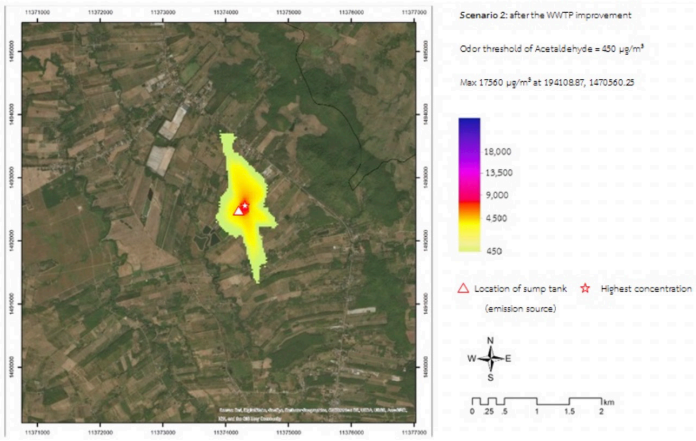

Figure 5.2. Spatial distribution of peak concentration (3 min) of acetaldehyde under scenario 1

4. Conclusions

Wastewater treatment system is one of the major emission sources of chemicals released from the tapioca starch industry. Results from the direct measurements of VOCs released to the air from wastewater treatment plant indicated that acetaldehyde was one of the chemical significantly released from this emission source. The extent and magnitude of acetaldehyde concentrations in the ambient air in the vicinity of the factory were evaluated using the AERMOD dispersion model. Predicted maximum 1h concentrations were further calculated for their peak concentration over 3 min using the peak to mean ratio approach. The effectiveness of measure to control emission from wastewater treatment by modifying the business as usual system (opened lagoon) to modified covered lagoon was quantified through the determination of areas affected from dispersion of odor of acetaldehyde. Results clearly illustrated the success of mitigation measure put on the wastewater treatment toward air pollution control. This study demonstrated the procedures to evaluate the impact of air pollutions by considering both their potential health impacts and odor nuisance effects. It also provide the quantitative data in evaluating the success of measures implemented for environmental pollution control which can be further applied to other cases in order to serve for evaluation of the success and the appropriateness of mitigation control scenarios.

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