



International Journal of Environment and Geoinformatics (IJECEO) is an international, multidisciplinary, peer reviewed, open access journal.

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A Modeling Framework to Quantify Routine VOC Emissions and Concentrations from Organic Liquid Tanks

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Received: 28.05.2021

Accepted: 04.09.2021

How to cite: Kocak, T.K. (2022). A Modeling Framework to Quantify Routine VOC Emissions and Concentrations from Organic Liquid Tanks. *International Journal of Environment and Geoinformatics (IJEGEO)*, 9(1):011-017. DOI:10.30897/ijegeo. 943706

Abstract

The oil industry has been a primary source of energy for years but it can also lead to the emission of Volatile Organic Compounds (VOC). VOCs play a major role in the formation of photochemical oxidants and can be harmful to the ecosystem. Thereupon, effective mitigation and control strategies of air pollution have recently become more prominent for the oil industry. To orchestrate these strategies, the understanding of how air pollutants disperse from organic storage tanks should be improved. In this study, a modeling framework was developed to estimate in-field two-month average VOC concentrations caused by crude oil tanks. Firstly, United States Environmental Protection Agency's (US-EPA) Tanks 9b software was used to estimate emission rates from tanks. Then, Gaussian Dispersion Formulation was applied to simulate VOC dispersion. Following this, an in-house equation was used to represent the average VOC concentration at selected receptor locations. Moreover, in-field VOC measurement (passive sampling method) was also conducted to evaluate model performance. The normalized root-mean-square deviation between the measured and estimated VOC concentration was found to be 0.15. There was also a strong correlation between the two data with a correlation coefficient of 0.96. Overall, the results suggest the model statistically performed well with a 95% confidence interval. Due to its effectiveness and time-saving application, the method described in this study can be used to develop air pollution mitigation plans for organic storage facilities.

Keywords: Organic Storage Tanks, Volatile Organic Compounds, Air Pollution Modeling, Crude Oil

Introduction

The oil industry has globally become an indispensable part of the energy sector. With being the leading source in the energy business, oil products provided 34% of energy consumption in 2019 (Spencer, 2019). Moreover, the primary energy consumption requirement (total energy consumption and losses) in the world is estimated to increase by 2.23% from 2020 to 2040 (Ahmad, 2020). In addition to this, the world crude oil reserves are predicted to provide raw material at least for 3 decades (Barthe, 2015). As a result, ever-increasing dependence on the oil industry seems most likely to remain for a foreseeable future. In the meantime, every stage from extraction to produce in the oil industry may have potential impacts on the environment (O'Rourke, 2003; Ngene, 2016). One of these stages is storing crude oil in hydrocarbon storage tank farms which emit volatile organic compounds (VOCs) to the ambient air. VOCs are a large group of hydrocarbons that have low water solubility and high vapor pressure, having members from BTEX (benzene, toluene, ethylbenzene, and xylene) to other toxic chemicals such as formaldehyde, trichloroethylene, styrene, and perchloroethylene (Anand, 2014). Impacts of VOCs induced by the petroleum sector on public health have been systematically well-reviewed in Rajabi (et al., 2020) study. The detrimental effects of VOCs on human health are various from acute to long-term exposures result in

eye irritation, dizziness, headache, cancer, decreased mental capacity, respiratory diseases, immune and central nervous system disorders. VOCs also play a vital role in the formation of tropospheric ozone and photochemical oxidants that adversely influence the air quality (Finlayson-Pitts, 1993). Therefore, VOCs caused by hydrocarbon tanks should be managed delicately to minimize their adverse effects on the environment and public health. Accordingly, in order to implement effective mitigation strategies and comply with legal regulations, accurate emission and concentration estimations of VOCs are required.

Several air pollution studies regarding hydrocarbon tanks were conducted to have a better understanding of the associated factors that can be used for the emission mitigation strategies. However, they are either limited with the emission estimation only or use a high resolution for concentration receptors.

Lu (et al., 2013) estimated VOC emissions from 3 different liquid tanks to commentate their emission characteristics. They mainly focused on standing and working emission losses and found out that the working losses showed better agreement with the measured emissions compared to the standing losses. However, their study did not include the concentration estimates of VOCs. In another study, United States Environmental Protection Agency (US-EPA) Tanks (a software

specifically designed to estimate emissions from liquid tanks) was used to analyze emission reduction after a renovation work of 27 refinery tanks in a city of Serbia (Jovanovic et al., 2010). Due to the purpose of their study, their modeling only considered the emission estimation. Similarly, Invernizzi (et al., 2018) proposed a method to correlate emission mass from hydrocarbon tanks with odor emission rate. This study may be useful to quantify odor flow rates resulting from the tanks but lacks in estimating pollutant concentration near the sources.

Large-scale air dispersion studies provide a comprehensive look at the environmental impacts of VOCs caused by tank farms and petroleum refineries in cities or municipalities. Jackson (2006) developed a modeling framework to estimate VOC emissions and the concentrations of benzene, toluene, and xylene from the organic liquid storage tanks of eight different companies in the city of Dar-es-Salam. He used US-EPA Tanks and CALLPUFF (a non-steady-state air quality dispersion modeling system) for the estimation of emissions and concentrations caused by the tanks, respectively. His study covers a 100 km²-area with a 1 km resolution. He found out that the type of the tanks highly influences the emission rate of VOCs. He concluded the study that the VOC concentrations on the downwind were below tolerable limits according to standards set by the World Health Organization (WHO) and the United States Occupational Safety and Health Administration (US-OSHA) but the carcinogenic risks associated with the maximum benzene exposure were higher than the US-EPA standard. In a similar study, Carletti (et al., 2014) applied a modeling framework to quantify fugitive VOC emission from an oil refinery in Falconara Marittima, Italy. They used emission factors from the emission inventory of the European Environment Agency to estimate VOC emission caused by the refinery. Later, they applied AERMOD (A steady-state plume model) to simulate air dispersion. Their study suggests that underestimation of VOC emission caused by petroleum refineries may lead to a misdetection of pollutant concentration in the nearby area. AERMOD was also used in a similar study for simulating air pollution dispersion caused by a tank farm of a petroleum refinery

to evaluate its odor and health impact (Saikomol et al., 2019). In the study, emission mass from the EPA Tanks model was used as input to AERMOD but receptors were selected from outside of the farm and the dispersion model was not validated with measured data. Correspondingly, Howari (2015) and Ashafi (et al., 2014) used dispersion models to provide a further understanding of the issue. However, the above-mentioned studies mainly focus on the pollutant concentrations outside of the tank farm. The fact remains that VOCs in cities are highly influenced by the other anthropogenic VOC sources such as transportation, paint industry, chemical industry, and industrial manufacturing processes (Carletti, 2014; Ling and Guo, 2014; Huang and Hsieh, 2020). Furthermore, most government regulations, similar to Turkey's, usually set permissible limits for VOC concentration inside the field of a storage facility. Therefore, the current study aims to develop a modeling framework to quantify VOC emission caused by crude oil tanks in a tank farm.

Study Area

The crude oil storage facility used in this study is located in the southern part of the Eğil district of Diyarbakır Province, Turkey (fig.1). Similar to Diyarbakır, summers are very hot and dry, while winters are cold with little rain in Eğil. During the study period, maximum and minimum temperatures were 13.6 and -1 Celsius, respectively.

The storage facility covers an 83000-meter square area in the district. The facility is 26 km away from Diyarbakır city center, as the crow flies. There is an organized industrial site 5.5 km south of the facility. The sectors that carry out their businesses in the organized industrial site include food, metal, plastic, furniture, textile and construction. In an area of 4 km around the facility, there are also textile, furniture factories, and another hydrocarbon storage facility. In order to minimize contribution of these sources on ambient VOC concentration, dispersion modeling of the current study focuses on the VOCs inside of the facility.

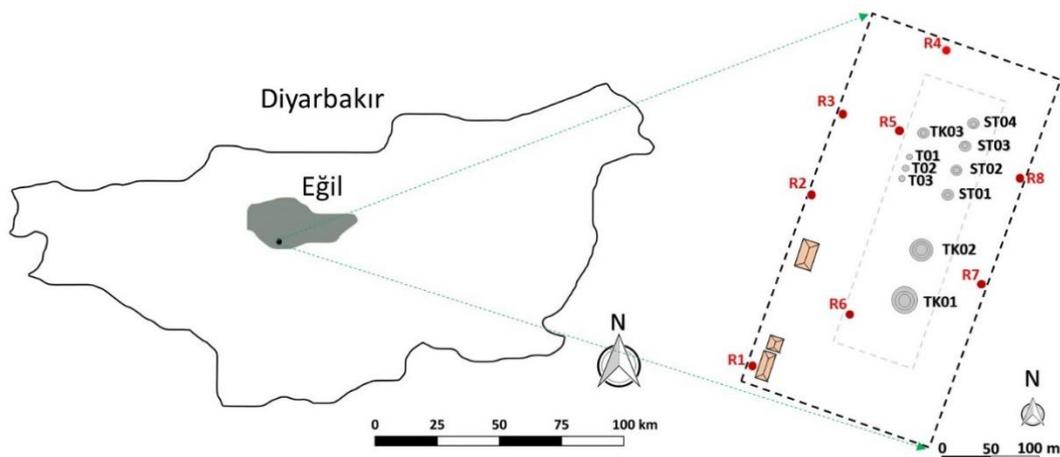


Fig. 1. The study area: Diyarbakır Province and Eğil district (small grey area) on the left side, crude oil storage facility with the tank (grey circles) and VOC receptor (red dots) locations on the right side.

Materials and Methods

The modeling framework applied in this study to quantify VOC pollution consists of three major parts: emission estimation, concentration estimation, and model performance evaluation. Emissions from crude oil tanks were estimated by using EPA Tanks 4.09d software. Then, estimated emission rates were used as input to simulate pollutant dispersion. The passive sampling method was chosen to conduct VOCs field measurement since it is a regulatory measurement method in Turkey. The samples were collected from January 22th to March 22th for the year 2021. Lastly, in order to evaluate model performance, measured VOC concentrations were statistically compared with the modeled ones. Details are described below.

Emission Estimation

Ten tanks given in figure 1 are fixed roof tanks. The fixed roof tank is a type of tank that has a cylindrical shape with a steel shell and a permanently affixed roof. Routine fugitive emissions from a fixed roof tank occur in two ways; these are working loss and breathing loss. The working loss is seen during liquid filling and emptying episodes. The breathing loss, on the other hand, mainly occurs due to weather conditions; the change in temperature and pressure expands and contracts the vapor pressure of the liquid in the tanks. Consequently, the differences between vapor pressure and atmospheric pressure cause VOCs to emit into the atmosphere. Therefore, total fugitive emission (L_t) from an organic liquid tank is the sum of working and breathing losses. The basic equation for L_t (mass/time) is given in equation 1. Its details are extensively explained in the EPA document, Compilation of Air Pollutant Emissions Factors (AP-42) Chapter 7: Liquid Storage Tanks (US EPA, 1995).

$$L_t = V_Q K_N K_p W_V K_B + 365 K_E \left(\frac{\pi}{4} D^2\right) H_{V_O} K_S W_V \quad (\text{Eq.1})$$

Where V_Q is the working loss throughput (volume/time), K_N is the working loss saturation factor (dimensionless), K_p is the working loss product factor (dimensionless), W_V is the vapor density (mass/volume), K_B is the vent setting correction factor (dimensionless), K_E is the vapor space expansion factor (per day), D is the tank diameter (length), H_{V_O} is the vapor space outage (length), K_S is vented vapor saturation factor (dimensionless) and W_V is the stock vapor density (mass/volume). The first term on the right side of equation 1 gives the working loss ($V_Q K_N K_p W_V K_B$), while the second is breathing loss ($365 K_E ((\pi/4) D^2) H_{V_O} K_S W_V$). The total loss (L_t) was separately calculated for each tank shown in figure 2 by using EPA Tanks 9b software.

Meteorological parameters such as diurnal temperatures, atmospheric pressure, the intensity of solar energy, and wind speed for the software were obtained from the Eğıl weather station where is roughly 15 km away from the

study area (*Meteoroloji Genel Müdürlüğü*, n.d.) Other major parameters for the emission estimation include tank characteristics (dimensions, condition, type etc.), number of turnovers, tank capacities, and stored liquid features such as vapor pressure and molecular weight. The stored liquid in this study was crude oil for each tank. Some other main features of the tanks which were used as inputs to EPA Tanks 9b are given in table 1.

Table 1. Main features of storage tanks

Tank Name	Tank Purpose	Working Capacity (bbl)	Number of Turnovers*
TK01	Overflow	35000	3
TK02	Overflow	25000	3
TK03	Overflow	5000	6
ST01	Stock	5000	5
ST02	Stock	5000	5
ST03	Stock	5000	5
ST04	Stock	5000	5
T01	Test	1000	52
T02	Test	1000	50
T03	Test	1000	54

* The number of turnovers was taken during the study period (2 months)

EPA Tanks software is capable of estimating tank emissions for a year or month. In this case, total emissions for each tank were estimated for a time period between January 22th and March 22th for the purpose of this study.

Concentration Estimation

In order to estimate VOC concentrations from crude oil tanks, the steady-state Gaussian dispersion equation was used. The VOC contribution (C, mass/volume) of a puff caused by a tank to a receptor location (x, y, z) is given in equation 2 (Turner, 1994).

$$C_{(x,y,z)} = \frac{Q/u}{\sigma_y \sqrt{2+\pi}} \exp\left(\frac{-y^2}{2\sigma_y^2}\right) \left\{ \frac{1}{\sigma_z \sqrt{2+\pi}} * \left[\exp\left(\frac{-(z-H)^2}{2\sigma_z^2}\right) + \exp\left(\frac{-(z+H)^2}{2\sigma_z^2}\right) \right] \right\} \quad (\text{Eq.2})$$

Where Q is the emission rate (mass/time) and H is the effective stack height or the breathing height of a tank (length). The wind speed (length/time) here is u which has the same direction as x. Respectively, σ_y and σ_z represent the lateral dispersion (length) and vertical deposition coefficient (length). Spline fit of σ_y and σ_z illustrate the downwind concentration profiles.

The emission rates (Q) of each tank for equation 2 were obtained from EPA Tanks software. Two-month total emissions (January and March) were averaged as gram per second. Similarly, wind speeds for the same time period were averaged based on their directions. Average wind speeds and their respective directions are illustrated in figure 2.

stock tanks (ST01, 02, 03, and 04) were found to have the same amount of emissions due to the fact that their working volume and number of turnovers were the same (see Table 1). Lastly, the least amount of VOCs was estimated to be emitted by the test tanks (T01, 02, and 03) as a result of their smaller sizes, even though their turnover counts were highest among other tanks. This may be attributed to their low capacity.

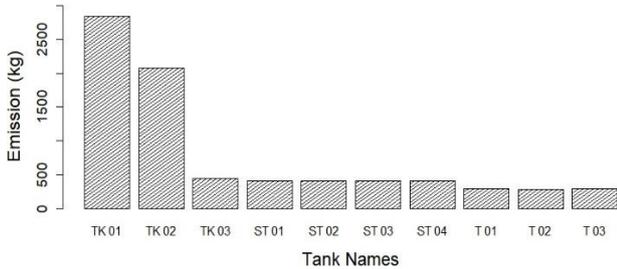


Fig. 3. Two-month cumulative emissions (kg) of each tank from January 22th and March 22th (2021)

Concentration Results

Gaussian dispersion simulation for a few tanks is given in Figure 4 to represent how VOCs spread. The direction and speed of wind in the figure are West North West (prevailing wind direction in the study area) and 2.5 m/s (average wind speed in the prevailing wind direction), respectively. The estimated VOC concentration of each

tank showed variety depending on the effective stack height and the emission rate. TK01 was seen to emit the highest concentration ($266 \mu\text{g}/\text{m}^3$) as expected because of having the highest emission. VOC concentrations caused by test tanks were estimated to be lower compared to the other tanks. Overall, VOC concentrations were attenuating with the distance from the tanks.

Table 2 shows the measured (C_m) and estimated (C_e) wind-adjusted average VOC concentrations ($\mu\text{g}/\text{m}^3$) at receptor locations. Both measured and estimated concentration at R2 were the highest, compared to other receptor locations. The lowest concentrations, on the other hand, were seen at R1 and R6. This pattern could be caused by the wind features and the tank locations in the study area. R2 is located northwest of TK01 and TK02 which had the two highest emission rates during the study period (see Figure 3). Because of the prevailing wind direction being West North West (see Figure 2), R2 was the most affected receptor location by TK01 and TK02 in terms of VOC emission. R1 and R6, in contrast to R2, are located southwest of the facility where the wind blows the least. Therefore, this positioning could be why R1 and R6 had the lowest concentrations, while R2 had the highest. Similarly, concentration differences at other receptors could be due to the wind features and tank locations.

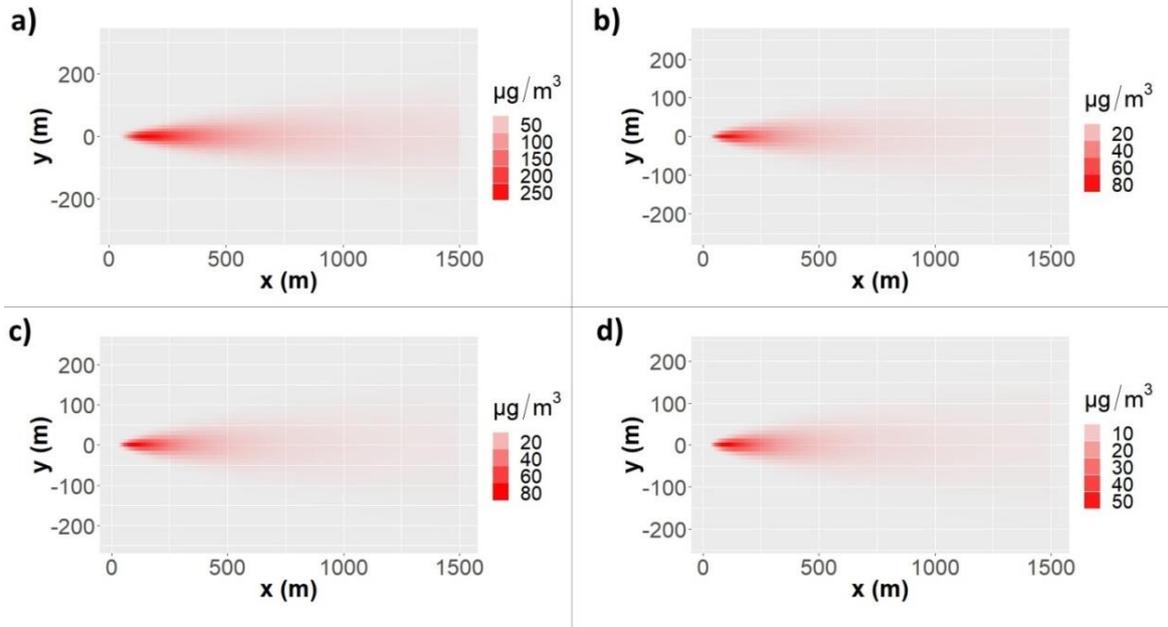


Fig. 4: Gaussian dispersion plots for a) TK01, b) TK03, c) ST01, and d) T02. Plots are given in coordinate system (X, Y) and distances in meters. Origins (0, 0) of each plot represent the tank location. X direction lies on the same direction with the prevailing wind direction (West North West)

Table 2: Measured (C_m) and estimated (C_e) wind-adjusted average VOC concentrations ($\mu\text{g}/\text{m}^3$) at receptor locations (R_i)

Receptor	R1	R2	R3	R4	R5	R6	R7	R8
C_m	16.3	62.9	38.1	58.8	34.2	18.2	34.3	37.2
C_e	16.2	83.1	37.8	58.3	32.9	17.8	33.3	36.8

According to these results, the modeling framework tends to underestimate VOC concentration except for R2. This slight underestimation trend was expected since the current study only accounted for the organic liquid

tanks' contribution to VOC concentration in ambient air. Nonetheless, there might be other VOC sources that contribute the VOC concentration outside of the storage facility near the study area. As for the overestimation of

VOC concentration at R2, the structure of Equation 3 could be the reason. According to the equation, the amount of wind blows from a specific direction highly impacts to average concentration. In this situation, R2 was significantly fed by TK01 and TK02 in terms of VOC concentration because of the West North West wind direction. This resulted in overestimation. Whether these differences between the measured and estimated values have a statistical meaning is discussed in the next section.

Lastly, study findings for the average VOC concentration was also evaluated from a regulatory point of view. The fact remains that many countries have not set any acceptable limit for total VOC concentration in the ambient air. Instead, they mostly focus on individual VOCs such as benzene (European Commission, n.d.) and toluene (OSHA, n.d.). However, a few countries determine an acceptable limit for average VOC concentration (also known as long-term VOC concentration) in the ambient air such as Turkey (500 $\mu\text{g}/\text{m}^3$) and the United Arab Emirates (20000 $\mu\text{g}/\text{m}^3$) (Howari, 2015). The findings of the current study were far below these limits.

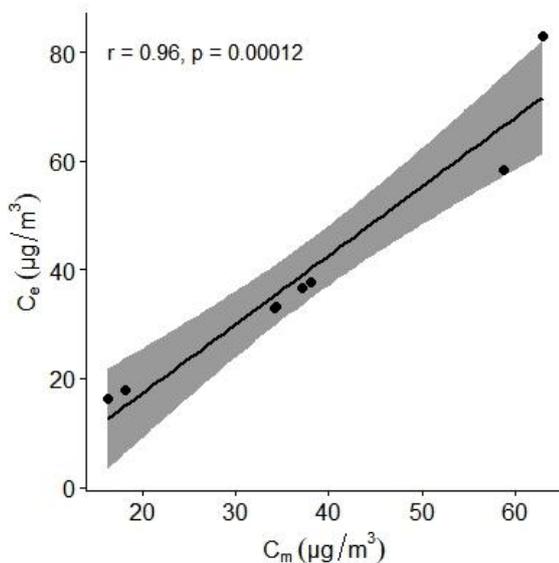


Fig. 5. Scatter plot of estimated [$C_e(\mu\text{g}/\text{m}^3)$] and measured [$C_m(\mu\text{g}/\text{m}^3)$] VOC concentration at receptor locations. Shaded areas along the fitted line shows 95 % confidence interval, while r is the Pearson correlation coefficient and p is the p -value of the test.

Model Evaluation Results

The normalized root-mean-square deviation (NRMSD) between the measured and estimated concentration was 0.15. Low NRMSD such as in this study indicates less residual variance. The output of the Shapiro-Wilk test showed p -values of 0.31 and 0.18 for the measured and estimated concentrations, respectively. P -values being greater than the level of significance (0.05) imply that the distribution of data is not significantly different from a normal distribution. Accordingly, they both were from a normal distribution and suitable for Pearson Correlation. As for the Pearson Correlation coefficient, the p -value ($p=0.00012$) was found to be less than the

significance level alpha (0.05). Thus, it can be concluded that measured and estimated values were significantly correlated with a correlation coefficient (r) of 0.96. The Correlation has also been visualized with a scatter plot in Figure 5. As seen in the figure, the covariation is linear since the scatter plots show linear patterns. Overall, the modeling approach statistically performed well with a 95% confidence interval. In other words, the estimated concentrations showed consistency with the measured ones.

Conclusion

Energy demand in the world has been growing exponentially over the years. The oil industry has globally been supplying this demand by being the leading source of energy need. However, working with petroleum products might have detrimental impacts on the environment including through air pollution. Therefore, effective mitigation strategies should be conducted to minimize these impacts.

In this study, a modeling framework was developed to estimate in-field VOC concentration in an organic liquid storage facility. Specifically, existing methods of emission and concentration estimation were combined to simulate VOC dispersion from the crude oil tanks. Later, average VOC concentrations at selected receptor locations were estimated by using the wind features in the study area. Last but not least, the results of the VOC passive sampling measurement were found to be in statistical accordance with the study findings.

The modeling framework described in this study can be used as a tool for the plants that have hydrocarbon storage tanks to plan their VOC management strategies. It does not require a complex set of data such as terrain information since it focuses on the pollutant concentration nearby the storage tanks. Thus, its application is fairly easy compared to the large-scale dispersion models. Additionally, the modeling framework eases to investigate alternative tank layout plans in terms of air quality control because it does not require excessive time such as in the passive sampling method. Overall, the method described in this study provides a practical approach to oversee the impacts of organic storage tanks on air quality.

Acknowledgements

The author gratefully acknowledges support provided by the Turkish Petroleum Corporation.

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