



VOCs emission rate estimate for complicated industrial area source using an inverse-dispersion calculation method: A case study on a petroleum refinery in Northern China[☆]



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ABSTRACT

This study aimed to apply an inverse-dispersion calculation method (IDM) to estimate the emission rate of volatile organic compounds (VOCs) for the complicated industrial area sources, through a case study on a petroleum refinery in Northern China. The IDM was composed of on-site monitoring of ambient VOCs concentrations and meteorological parameters around the source, calculation of the relationship coefficient γ between the source's emission rate and the ambient VOCs concentration by the ISC3 model, and estimation of the actual VOCs emission rate from the source. Targeting the studied refinery, 10 tests and 8 tests were respectively conducted in March and in June of 2014. The monitoring showed large differences in VOCs concentrations between background and downwind receptors, reaching 59.7 ppbv in March and 248.6 ppbv in June, on average. The VOCs increases at receptors mainly consisted of ethane (3.1%–22.6%), propane (3.8%–11.3%), isobutane (8.5%–10.2%), *n*-butane (9.9%–13.2%), isopentane (6.1%–12.9%), *n*-pentane (5.1%–9.7%), propylene (6.1–11.1%) and 1-butylene (1.6%–5.4%). The chemical composition of the VOCs increases in this field monitoring was similar to that of VOCs emissions from China's refineries reported, which revealed that the ambient VOCs increases were predominantly contributed by this refinery. So, we used the ISC3 model to create the relationship coefficient γ for each receptor of each test. In result, the monthly VOCs emissions from this refinery were calculated to be 183.5 ± 89.0 ton in March and 538.3 ± 281.0 ton in June. The estimate in June was greatly higher than in March, chiefly because the higher environmental temperature in summer produced more VOCs emissions from evaporation and fugitive process of the refinery. Finally, the VOCs emission factors (g VOCs/kg crude oil refined) of 0.73 ± 0.34 (in March) and 2.15 ± 1.12 (in June) were deduced for this refinery, being in the same order with previous direct-measurement results (1.08–2.65 g VOCs/kg crude oil refined).

An inverse-dispersion calculation method was applied to estimate VOCs emission rate for a petroleum refinery, being 183.5 ton/month (March) and 538.3 ton/month (June).

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1. Introduction

Volatile organic compounds (VOCs) adversely impact air quality and human health, through initiating the formation of ground-level ozone (O₃) and fine particulate matter (PM_{2.5}) in atmosphere. In eastern China, the influence of VOCs on atmospheric chemical

reactions became more and more significant for their huge emissions (Ou et al., 2015; Pan et al., 2011; Ran et al., 2012; Xing et al., 2011; Yamaji et al., 2012). Numerous emission inventories showed that China annually emitted about 16.5–23.2 Mt of anthropogenic VOCs in 2005–2013 (Bo et al., 2008; ESS, 2012; Wei et al., 2008; Zhang et al., 2009). Among various emission sources, petro-chemical industries contributed about 17.9%–39.6%, which mainly included petroleum refining industry, petrochemical industry, coking industry, chemical pharmacy industry, etc. And VOCs emitted from these petro-chemical industries usually involved many emission processes, including exhausted gas from stacks, leakage from valves and lines, evaporation from tanks, and fugitive

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emission from waste water treatment, etc. Consequently, these petro-chemical manufacturing plants were usually treated as area sources in the research fields of emission inventory and air quality modelling.

When estimating VOCs emissions for the petro-chemical industries, United States and European countries recommended the VOCs emission factors for these direct-process emissions, which were expressed in units of kilograms VOCs per cubic meters feed (EEA, 2006; U.S. EPA, 1995). So the detailed process information, such as the consumption of feeds by various units, the storage and transportation of raw materials and finished products, the treatment of waste water, etc., was essential. Then, summing up these direct-process emissions, and determining the integrated VOCs emissions. According to this bottom-up approach, the integrated VOCs emission factor of 1.08–2.65 (g VOCs/kg crude oil refined) was concluded for the petroleum refining industry in developed countries. However, this bottom-up approach is infeasible in China, because the detailed process information has not been gathered by China's environmental protection departments or statistic departments. And Chinese VOCs emission inventories had to employ the integrated VOCs emission factors from the developed countries for the petro-chemical industries (Fu et al., 2013; Huang et al., 2011; Mo et al., 2015; Qiu et al., 2014; Wei et al., 2011; Wu et al., 2015; Yin et al., 2015; Zheng et al., 2009). Undoubtedly, it will bring about some estimate uncertainty, for there were the differences in processing flow scheme, arrangement of emission processes, raw materials and management between developed countries and China.

Facing this difficulty, we tried to initiate an inverse-dispersion calculation method (IDM) to estimate VOCs emissions for these complicated industrial area sources by the following steps: (1) monitoring ambient VOCs concentrations and meteorological parameters around an industrial source; (2) quantifying the relationship coefficient γ between the source's VOCs emission rate and the ambient VOCs concentration under the actual meteorological conditions measured; (3) combining step 1 and step 2 to calculate the actual VOCs emission rate for this source. This inverse technique was also employed in previous studies for source apportionment of air pollutants in industrial complex regions (Flesch et al., 2005; Khlaifi et al., 2009; Patrick and O'Shaughnessy, 2011; Shaughnessy and Altmair, 2011).

In this study, we constrained the IDM application within the several-kilometers zone surrounding the source, to ensure the notable influence of the studied source on its ambient concentrations. Thus, the relationship coefficient γ could be achieved by air pollution dispersion model in medium and small scale, when only the studied source contributes to the ambient concentration. There are three typical steady-state Gaussian plume dispersion models that can well predict near-field concentrations from most types of sources, including ISC3 model, AERMOD model, and ADMS model. The ISC3 model mainly uses sequential hourly surface meteorological data to calculate values of average concentration with the time resolution of 1 h. The AERMOD model is the next generation which incorporates planetary boundary layer concepts. Both surface and upper air meteorological data are required by the AERMOD model. In addition, the AERMOD model also has a better performance in regions of complex terrain, which leads to its dependence on terrain data and land use data. Both the ISC3 model and the AERMOD model were proposed for regulatory use by U.S. Environmental Protection Agency.

The ISC3 model had been widely used in the field of air pollution. For example, Banerjee et al. (2011) used the ISC3 model to apportion sources' contributions to ambient NO_2 concentration in an integrated industrial estate Panthnagar of India. Huertas et al. (2012) used the ISC3 model and the AERMOD model to simulate the dispersion of total suspended particulate emitted from multiple

open pit coal mines in northern Colombia. Mofarrah and Husain (2010) used the ISC3 model to trace the pollutant's transport and transformation according to air quality monitoring network in Canada. The previous studies approved that the ISC3 model could well estimate ambient concentrations of air pollutants in small scale space, with the normalized mean error (NME) of about 15.6%–52.9%. Considering the simplicity of the preparation of the input data, the ISC3 model was selected here to quantitatively link the source's emission rate to the ambient concentration.

Comparing with the traditional direct-measurement method, the IDM was subject to a certain uncertainty, for the inevitable simulation error in the dispersion model. However, we still believed that the IDM would be an effective approach to estimating VOCs emissions for a large number of complicated industrial area sources, through scientifically designing its application. Moreover, the IDM would also provide a good verification to improve the understanding of VOCs emissions from these important area sources.

2. Methodologies

2.1. Application of the IDM

Combining with the characteristics of the ISC3 model, the IDM application in this study was constrained in following aspects: (1) the terrain of the area where the measured source is located should be relatively flatter; (2) the IDM should be conducted within the 2-km zone surrounding the source, to ensure the obvious contribution of this source to ambient VOCs concentrations; (3) there should be no other potential sources in the studied zone, especially eliminating residential districts, factories and roads with heavy traffic; (4) meteorological conditions of calm winds should be avoided when carrying out the IDM; (5) VOCs concentrations at several different downwind receptors should be simultaneously measured and used to estimate the source's VOCs emission rate, which comparison can evaluate the uncertainty of the IDM application.

Moreover, we also made two criterions before using the IDM for QA/QC. Criterion (1), the ambient downwind VOCs concentrations around the source are obviously higher than background level. Criterion (2), VOCs increases through subtracting background VOCs from downwind VOCs are consistent in chemical composition with the source's VOCs emissions which might be reported by literatures. Both criterions could assure that the studied source and only the studied source predominantly contributes the ambient VOCs concentrations. The similarity between two different chemical compositions was evaluated by coefficient of divergence (COD), which calculation as shown by formula (1) (Chen et al., 2014; Massoud et al., 2011). COD value is in the range of 0–1. And 0 means two chemical compositions are totally same, and 1 means two chemical compositions are totally different.

$$COD_{kl} = \sqrt{\frac{1}{p} \sum_{s=1}^p \left(\frac{x_{sk} - x_{sl}}{x_{sk} + x_{sl}} \right)^2} \quad (1)$$

where COD_{kl} was coefficient of divergence between chemical composition k and chemical composition l ; x_{sk} and x_{sl} was the proportion of compound s in chemical composition k and in chemical composition l respectively; p was the number of chemical compounds.

2.2. Schedule of field monitoring around a petroleum refinery

According to the application conditions of the IDM, a petroleum

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