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ORIGINAL PAPER

THE CONTRIBUTION OF AIR EMISSION TO THE ENVIRONMENT OF GEELONG BASED ON THE PMF RECEPTOR MODEL*

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ABSTRACT

The research aimed to evaluate the gas pollutant emission in a relatively closed and stable environment, specifically in the Geelong area on Corio Bay, Victoria state, Australia, under the circumstances of the global respiratory epidemic. During this pandemic, owing to the small local population and a limited number of travelers, industrial emissions have become the only vital factor to interfere with air quality, excluding the impact of ordinary daily traffic. PM₁₀ (ug/m³) data were measured every hour by the Victorian Government, uninterruptedly from 2017 January 1st to 2020 December 31th. The emission of industrial waste gas and the leakage of fossil oil jointly determine local air conditions, which can be reflected in the numerical value of the PM₁₀ in the Geelong area. Sample data of PM₁₀ were analyzed through the US EPA Positive Matrix Factorization model (PMF 5.0) to show the tested factor and contribution output. The results demonstrate the actual air condition in a local environment, which reflects the impact of regional factories on the environment. In addition, the model result will be compared to the annals data to testify its accuracy and precision; moreover, it will forecast the next year's air condition, which is of practical significance for treatment of waste gas emissions from factories and for the development of environmental protection.

Keywords: air pollution sources, oil spill, receptor model, COVID-19, PMF 5.0 model.

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INTRODUCTION

The COVID-19 epidemic, which is especially severe in Australia, broke out in late 2019 and spread all over the world, despite the governments taking several measures to control it. Since March 2020, the Australian government has denied entry to anyone without the resident status (CHANG et al. 2020), resulting in Australia's ecological environment being affected solely by domestic production and macro-climate change; and the consequences remain unknown so far. Based on this consideration, the influence of the oil processing factory in Corio Bay in Geelong (VIC) on the surrounding bay environment will be assessed. Through the official data, the specific impact of the oil industry on air quality in the region can be analyzed and compared, aiming at predicting the air situation in 2021 and putting forward constructive opinions and suggestions.

For many years, the air quality prediction method has been based on the statistical analysis of meteorological data of air pollution, taking the physical mechanism of air pollution as the starting point and using the knowledge of atmospheric conditions, geographical sciences and mathematics as well as computer simulation technology to predict the spatial and temporal distribution of concentrations of air pollutants in a region with certain pollution sources (YAMAZAKI et al. 2017). It has a broad prospect for development. The data about air conditions will help understand the current situation of the environmental quality, providing a decision-making basis for the government to formulate environmental policies and pollution control measures, so as to achieve the ultimate goal of improving the ambient air.

Massive data need to be processed with professional and effective analytical methods. Different models are implemented to distinguish levels of air quality in order to evaluate the environmental circumstances. Traditional comprehensive indexes of environmental quality mainly include an elementary mathematical index model, simple addition, arithmetical average, and weighted arithmetic mean. For a weighted arithmetic mean, the contribution of each factor presented in the database to the average is not equal, and some factors are more important than others, which can be represented by the following formula:

$$\bar{x} = \frac{\sum_{i=1}^{n} w_i x_i}{\sum_{i=1}^{n} w_i}$$
(1)

where, x represents the weighted mean of a non-empty finite multiset of data, with corresponding non-negative weights, w (Bor et al. 1989).

However, most research has abandoned this method, having considered several reasons, such as low precision and inclusion of extreme data. The Nemerow index is a weighted multi-factor environmental quality index that takes into account the extreme value or prominent maximum value, and which avoids the influence of subjective factors on a weight coefficient in the process of weighting (L_I et al. 2009); however, it overemphasizes the weight value of the highest data in an analysis. As a result, it is still not an ideal model for air quality analysis and assessment.

The receptor model, or the receptor-oriented model, is a digital model and a method to identify and analyze different sources and contribution rates of air pollutants at receptors (HOPKE et al. 2000). It is better than the traditional approaches, and therefore it was employed in this research, carried out in Victoria state, Australia. The basic mechanism of the receptor model is the mass balance relationship between the source of pollution and the receptor (WATSON et al. 1984). Pollutants are discharged from a source and evenly distributed in the atmosphere after diffusing and mixing. Technically, the source and mass of atmospheric particulate matter and its elements of the receptor can be considered as the result of the superposition of different pollution sources around, which can be expressed by a simple numerical expression.

The difference between numerous receptor models is the richness of source data. Due to the disordered pollution sources from official environmental data, a non-data-exacting and operating-friendly receptor is wanted. Therefore, the research on the Corio Bay area adopts the Positive Matrix Factorization version 5.0 (PMF 5.0). Comparing to the old version, EPA PMF 5.0 adds two key components to EPA PMF 3.0, two additional error estimation methods and source contribution and profile constraints (Lu et al. 2018). Many other changes have been made to ensure that the software is easier to use, e.g. the ability to read data from multiple sites.

PMF 5.0 uses matrix decomposition to solve practical problems (PAATERO et al. 2014). The limitation of the receptor model is that all elements in a matrix are supposed to be non-negative, which fits the air data used in this research. The PMF uses the least square method for iterative operation, which can determine the pollution source components and contribution at the same time, and can also be directly compared with the original data matrix without conversion. In the process of solving, it imposes non-negative constraints on factor load and factor score to avoid negative value in the result of matrix decomposition, which makes factor load and factor score have explicability and clear physical meaning.

The aim of the research was to employ PMF 5.0 to evaluate the pollution sources of the primary and secondary pollutants in industrial exhausts, NO_x , CO, SO_x , O_3 , and other metals (WHEELER et al. 2020), in relation to PM_{10} in a suburban region of Geelong in Corio Bay, in Victoria state, situated close to the VIVA petroleum refinery. The accuracy and precision of the PMF 5.0 model will be tested through annual data. Moreover, according to the simulation results, some prospective estimation and discussion of 2021 will be made and compared with the official year annual year statistics.

MATERIAL AND METHODS

Material

The research was launched in the area of Corio Bay, in Geelong, located at 38°06'S-38°16'S / 144°36'E-144°44'E, in Victoria state, Australia (Figure 1). According to the Australian Bureau of Statistics, this specific region covers an area of 50.457 km² and has a total population of 23,959 (Illawarra 2016).



Fig. 1. Location of the air quality monitor station: Geelong, Victoria, Australia (YAMAZAKI et al. 2017)

The reason why this place was chosen as a sampling site is that the population composition of the region is uncomplicated at this moment, consisting of residents and workers. Due to the special epidemic prevention measures and the control period (CHANG et al. 2020), the air quality is only affected by means of transportation and the oil refining industry, excluding the traditional tourism industry and foreign trade. Under these circumstances, the pollution sources can be traced and identified in terms of components.

The air quality data were collected by the EPA Victoria hourly from January 1st, 2015 to December 31st, 2020. The whole annual data set includes dry bulb temperature (DBT), carbon monoxide (CO), nitrogen oxide (NO_x), ozone (O₃), sulfur dioxide (SO_x), scalar wind speed (SWS), vector wind direction (SWD), vector wind speed (VWS), trace metals, all being comprehensive air qualified indicators, needed to discuss the contribution of the less than 10 μ m>s Particulate Matter (PM₁₀), which will be used in the research, or as random variables of reference.

Another pollution information source is the Annual Sustainability Report by VIVA Energy Australia. The company holds the only crude oil refinery in the area, which is the main air pollution source locally. The report chronicles the volume of Greenhouse Gas (GHG) Emission (Table 1), and the crude Oil and Derivative Products leakage, from 2017 to 2020 (Table 2). These data will be used to compare and verify the result of the PMF 5.0 receptor. Predictions and suggestions for the future will be put forward based on the final conclusion.

	Greenhouse Gas (GHG) Emissions				
Year	2017	2018	2019	2020	
Total (t CO_2 e)	1,328,985	1,392,568	1,430,837	1,282,597	
Total scope 1 (tCO ₂ e)	1,032,422	1,061,632	1,113,911	1,000,445	
Refining $(tCO_2 e)$	1,020,905	1,020,846	1,101,920	985,025	
Other $(tCO_2 e)$	11,517	10,786	11,991	15,420	
Total scope 2 (tCO ₂ e)	296,563	330,936	317,082	282,152	
Refining $(tCO_2 e)$	258,586	290,158	276,423	246,632	
Other $(tCO_2 e)$	37,977	40,778	40,659	35,520	
Percent of total (%)	24.45	25.62	26.33	23.59	

Statistical data of refinery oil gas emissions

Table 2

Table 1

Specification	Oil spill data				
Year	2017	2018	2019	2020	
Spill value (kg)	7,200	6,200	5,200	5,500	
Total value (kg)	24,100				
Percent of Total (%)	29.3	25.9	21.8	23.0	

Statistical data of refinery oil spills in Geelong

Methods

Positive Matrix Factorization (PMF) is a recent type of a receptor model. The PMF model has been implemented to handle non-representative data, missing data, and outliers. This is an important property as it prevents the rejection of such values and hence the reduction of the initial data set (PAATERO al. 2014). It is as a weighted factorization problem with non-negativity constraints which, given the matrices X (input data matrix) and o

(uncertainties data matrix) and a selected rank p, is defined in a 2-dimensional (COMERO et al. 2009) case by the following equations:

$$X = GF + E(G_{ik} \ge 0, F \ge 0) \tag{2}$$

$$G = n \times p \tag{3}$$

$$F = m \times p \tag{4}$$

$$Q(E) = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{E_{ij}}{\sigma_{ij}} \right]^2$$
(5)

$$\{G, F\} = \underset{G, F}{\operatorname{argmin}Q} \tag{6}$$

The series of equations remains difficult to solve for its two different non-linearities, inequalities and products of unknowns. To deal with these issues, we use PMF2 and ME-2 (Multilinear Engine) as the main algorithms. To achieve clear distinction between PMF as a model and the name of the programs, the model is designated as PMF while the programs used to solve the model are designated PMF2, and ME-2 (PAATERO al. 2014).

PMF2 is used to solve 2-dimensional problems by means of the following model, and formula (1),(2),(3), (4), and (5) mentioned above:

$$X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} (i = 1...n; j = 1...n)$$
(7)

where: X is the measured data matrix, G and F are the matrices to be determined, and E is the residual matrix (the unexplained part of X). Due to the factor linearity, the matrices G and F can be exchanged without changes in matrix X. In a practical example, X can be viewed as a matrix containing the measured value of a certain variable, G – the contributions matrix of the identified sources, and F – the matrix characterizing each source. The elements of G and F are constrained to assume a positive value and this corresponds to the idea that no sources may emit negative amounts of physical substances. Where X_{ij} means the concentration of component j in sample I, g_{ik} means the weight of the emission source k to the sample i, f_{kj} means the concentration of component j in the emission source k, e_{ij} means residual. In this way, the PMF problem is then identified as minimization of Q(E) with respect to G and F, and under the constraint that each element of matrices G and F is to be non-negative (PAATERO al. 2014).

The Q value should have consistency and relevance to the theoretical Q value to guarantee the desired availability by following equation:

$$Q = mn - p(m+n) \tag{8}$$

In this paper, the PMF 5.0 version was used for data computing aimed to analyze the collected data of PM_{10} . According to data quality and chemical

characteristics, ten sets of data of air compositions and their respective uncertainty as the sample are used for data analysis. The based model runs are the twenties, and the factor model starts with random seed numbers. Five factors of Geelong were chosen to explore the relationship between air status and environmental pollution emission. The Q values, and the ratio of these Q values will be discussed.

RESULTS AND DISCUSSION

The PMF Time Series shows that the PAHs content, comparing the total amounts from the past four years, was 29.3% in 2017 and 25.9% in 2018. In 2019, the PAHs made up 21.8% and rose to 23.0% in 2020 (Table 2). Of all of the petroleum products, unrefined crude oil has the highest PAHs component concentration (MINAI-TEHRANI et al. 2009); thus, in the research, we use the PAHs as the tracer to represent various leakage problems during the crude oil refining process. Actual leakage accidents were recorded and we can calculate the number of spills and containment. Comparing with these dual sets of data, it becomes explicit that the similarity reaches 97.8% (Table 3, Figure 2), which means that the PMF Time Series reflects the Table 3

Specification	Oil spill data				
Year	2017	2018	2019	2020	
Percent of true (%)	29.3	25.9	21.8	23.0	
Percent of exp. (%)	28.2	26.3	22.1	23.4	
Percent differences (%)	1.1	0.4	0.3	0.4	
Percent diff total (%)	2.2				

Statistical data of refinery oil spill between true and experimentally determined events



Fig. 2. Species concentrations of refinery oil spills between from 2017 to 2020

actual oil leakage and related accidents well. Therefore, the PAHs data and the oil spill events can be components in an analysis carried out to judge the contribution of petroleum refineries to the air pollution. However, comparing the data of the Times Series with the total greenhouse gas (GHD) emissions (Table 1), we can find out that there is no exact correspondence between the two data sets. For instance, most crude oil spills occurred in 2017, but were the least numerous in 2019. In terms of GHG emissions, it can be seen that the emissions in 2017 are not as large as those in 2019, which means that the small number of oil spills had no decisive influence on the annual air pollution, contrary to what we might have expected. This research result has encouraged us to plan a new study with the neo miniaturized near-infrared reflectance spectrometer technology (TANG et al. 2019), in order to explore soil contaminated with spilled crude oil so as to understand if there is any influence for those oil spills to crop yields.

As the PMF factor contribution analysis results show (Figure 3), the $\rm PM_{_{10}}$ pollution source mainly comes from six different main components, while other components occur in a very low content (accounting for approximately 1%). The largest pollution source are crude oil refineries in the region. Power generation and blast furnace smelting consume much coal; as a result, Cl can be regarded generally as the tracer characteristic component of coal (DUAN et al. 2006). The second largest pollution source is the vehicle exhaust emission in this area. Zn and Cu are the main additives in automobile lubricating oil, and Mn and Ba are also widely used in brake pads and tires. In addition, Pb also appears in gasoline. Therefore, these three elements are used as tracer characteristic components to judge the pollution caused by motor vehicles (Yu et al. 2013). The third largest pollution source are ocean shipping freighters. The fuel used by large ocean-going cargo ships is usually heavy diesel or heavy oil; thus S, Ni, V, Fe, and Cu can be used as tracer characteristic components. The fourth largest source of pollution comes from the construction industry in the city. Mg, Ca, Na, Al, Fe, which are used as tracer characteristic components, are related to cement, lime, and other building materials. The fifth largest pollution is light pollution in cities. SO_4^{-2} , NO_3^{-1} ions in the light pollution material can often transform to SO_2 and NO_v as a tracer characteristic group (TANG et al. 2006). The sixth largest pollution source comes from airborne dust in the area. Al, Ca, Fe and other earth crust elements are used as tracer characteristic components (Table 4). The remaining 1% of the pollution source components will be discussed in the future research if there are special reasons.

We can see that the atmospheric pollution caused by crude oil refineries ranks the highest, followed by the exhaust emissions from motor vehicles and sea freighters. The average value of pollution due to refineries in 2017-2019 is 31.6%, compared to 56.3% in 2020, which was a year affected by the epidemic (Table 4). The value for the whole year 2020 increased by 24.7%. Does it mean that the expansion of refinery production last year made the environmental pollution more serious? The data show that greenhouse gas



Fig. 3.a,b,c,d The relative contributions of PM₁₀ sources in Geelong

	Monitoring air quality pollution sources							
Year	refinery	vehicle	freight- er	construc- tion	light	dust	others	
2017	25.8	25.1	23.2	12.6	10.0	1.6	1.0	
2018	35.1	26.3	12.3	12.2	7.9	4.0	1.2	
2019	34.0	25.3	21.6	13.8	2.5	0.6	0.4	
2020	56.3	15.1	8.3	7.9	7.8	2.8	1.3	
Ave 17-19	31.6	25.6	19.0	12.9	6.8	2.1	0.9	
Diff 20-Ave	+24.7	-10.5	-10.7	-5.0	+1.0	+0.7	+0.4	

Statistical data of the relative contribution proportions of PM_{10} sources in Geelong

emissions were almost the same in the past four years, even decreasing in 2020 by 2.74% compared with 2019 (Table 1). This shows that the refinery capacity is almost close, which reflects there are some proportion changes of other gas pollution sources in the research. When we observe the data regarding the second pollution source, motor vehicles, it is not difficult to find that the average percentage from 2017 to 2019 is 25.6% (Table 4). In 2020, however, it fell by 10.5% down to 15.1% (Table 4). The reason why this happened is that, with the outbreak of COVID-19 in 2020, the Australian government adopted a closed-door policy, which made it impossible for almost all foreign tourists, foreign students, and green card holders to enter the country. In addition, due to the repeated epidemic waves, multiple urban blockades, and the rise of remote work (CHANG et al. 2020), the utilization rate of motor vehicles gradually decreased, resulting in the decrease of total vehicle exhaust emissions, which explains the mentioned change. Moreover, the experimental results show that the third largest pollution source is the traffic around the sea of large cargo ships connected with the shipping industry. The average weighted contribution from 2017 to 2019 is 19% (Table 4). In 2020, it decreased by 10.3% down to just 8.3%, which is similar to the decline of daily urban transportation (Table 4). We have reason to believe that this was also a consequence of COVID-19. The global transportation industry was almost stagnant in 2020. All customs departments investigated imported products, especially cold chain products, to prevent any incidental invasion of the virus. Australia has almost exclusively domestic shipping and a small amount of trade with New Zealand (Lee et al. 2020). Similarly, the sharp reduction in tourism also greatly reduced the probability of large cruise ships, arriving, anchoring and refueling in the local port. Due to the change of the shares of the three main pollution sources, the percentage contributions of the remaining pollution sources also change in 2020.

The fear of the COVID-19 epidemics in Australia and around the world will not disappear soon. It will deter many people from living in hotels and staying in crowded scenic spots. It will also restrain people's willingness to

travel. During the epidemic, the suspension of work and the loss of family income are visible to the naked eye (BLUSTEIN et al. 2020). Many families are inevitably forced to reduce their expenses. As luxury consumption, tourism and catering will be reduced. The vehicle exhaust emission might continue to decrease in 2021. In terms of shipping logistics, the impact of the epidemic on exports has become evident. Calculated by volume, 82% of the global cargo trade is transported by sea (CORBETT et al. 2003). The change of the marine shipping business can reflect the impact of the epidemic on trade in real-time. As the pandemic progresses, China, Singapore and other countries have tightened the regulations on berthing in Hong Kong. Maersk, Mediterranean Shipping, and other international shipping company groups have announced that they have reduced the number of ships on some routes to Australia. The average charter price in the Pacific region has fallen to the lowest level in the last three years in the first week of February 2020 (FERNANDES et al. 2020). This index reflects the impact of the epidemic on export trade in real-time from the perspective of the shipping market. The pollution caused by shipping might keep decreasing in 2021. Because of the changes above, contribution of the air pollution sources might increase in 2021.

For the whole EMP 5.0 receptor, the optimal solution ratio determined by self-diagnosis methods such as Q (robust), Q (true) and Q (true) / Q (exp) ratio is more reliable than the logical conclusion from an analysis of the temporal and spatial variation of factor source contribution (Table 5) – REFF Table 5

Year	Q(robust)	Q(true)	Q(true) / Q(exp)	Converged
2017	$3.23645 \cdot 10^{-3}$	$1.77373 \cdot 10^{-6}$	$2.94953 \cdot 10^{-6}$	yes
2018	$1.16403 \cdot 10^{-3}$	$2.00327 \cdot 10^{-6}$	$3.33123 \cdot 10^{-6}$	yes
2019	$1.79052 \cdot 10^{-3}$	$1.87183 \cdot 10^{-6}$	$3.11266 \cdot 10^{-6}$	yes
2020	$2.52546 \cdot 10^{-3}$	$1.69588 \cdot 10^{-6}$	$2.82007 \cdot 10^{-6}$	yes

Statistical parameters	s of the	PMF	5.0	model
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et al. (2007). Through a rigorous self-diagnosis of the PMF simulation results, the optimal solution can objectively identify the average status of each type of source contribution; as a result, all data in this research appear reliable.

CONCLUSIONS

The direct emission of PAHs due to leakage of crude oil in the refinery has become one of the air pollution sources. However, it has no too direct impact on the air quality, affected by the overall exhaust emission. According to the data collected in recent years, the main source of air pollution for the whole Geelong area is the exhaust emission from local petroleum plants, while the second most important source are exhaust fumes generated by motor vehicles and large ships.

The epidemic has greatly damaged the tourism and shipping industry in some areas, thus considerably reducing the contribution of these air pollution sources. It can be predicted that the relatives shares of these pollution sources will remain low in 2021.

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