

Impact of low-sulfur marine fuel regulations on PM_{2.5} in the Kyushu region of Japan using the PMF model

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Abstract. PM_{2.5} negatively impacts on human health and results from various domestic and transboundary sources, including shipping emissions. Maritime transport, which relies on heavy fuel oil, is one of the primary sources to air pollution. In response, the new marine fuel regulation (the IMO Sulfur Cap 2020) was introduced to reduce sulfur oxide emissions from ships. This study quantified PM_{2.5} contributions from maritime transport in the Kyushu region of western Japan. Seasonal PM_{2.5} chemical composition data from six monitoring sites (2016–2022) were utilized for the Positive Matrix Factorization model. Seven factors were identified: secondary sulfate and biomass burning (49.2%), road transportation (19.3%), heavy oil combustion (8.9%), secondary nitrate (6.6%), coal combustion (6.4%), soil (6.1%), and sea salt (4.8%). Secondary sulfate showed a gradual decrease, reflecting the Chinese governments' air pollution mitigation measures. In contrast, heavy oil combustion, strongly linked to shipping emissions, exhibited a sharp reduction after 2020. During 2017–2019 and 2020–2022, the contributions decreased by 65–97% across all sites, with the largest declines in Fukuoka and Oita. These results indicate that the IMO Sulfur Cap 2020 significantly reduced ship-related PM_{2.5} in Kyushu, highlighting the importance of international emission regulations for regional air quality improvement.

1 Introduction

PM_{2.5} has adverse influence on air pollution, climate change, and human health [1]. PM_{2.5} arises from diverse and complex emission sources, including both domestic activities (e.g., transportation, shipping, industry, and other local activities) and transboundary pollution [2]. Identifying source contributions is essential for developing effective mitigation strategies.

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Maritime transport, handling approximately 80% of global trade, primarily uses heavy fuel oils and generates substantial quantities of air pollutants and greenhouse gases each year [3-4]. For instance, international shipping released for approximately 4% of global anthropogenic PM_{2.5} emissions in 2018 [5]. These pollutants can degrade air quality and pose health risks not only in coastal zones but also in inland regions [6-7]. On 1 January 2020, the new marine fuel regulations (widely known as “the IMO Sulfur Cap 2020”) were issued for reducing sulfur oxide emissions from maritime transport. In the East Asian region, several studies have examined its impacts on PM_{2.5} [8-9]. However, existing research has primarily focused on the impacts in western areas in Japan, particularly the Seto Inland Sea [7, 10], yet few studies have specifically addressed those in the Kyushu region.

Previous source apportionment studies assessing the impact of shipping emissions have often employed the specific tracer compounds of PM_{2.5}, namely nickel (Ni) and vanadium (V) [6-10]. However, they are not unique to shipping and also substantially released from oil-fired power plants as well as industrial facilities, owing to their predominant origin in heavy fuel oil combustion. In the Kyushu region, large-scale industrial facilities and oil-fired power plants are located only in limited areas of the northern part, suggesting that source apportionment of ship-oriented PM_{2.5} is feasible.

The study quantifies the PM_{2.5} contribution from shipping emissions in the Kyushu region using observational data. The Positive Matrix Factorization (PMF) analysis was conducted using seasonal PM_{2.5} composition data from 2016 to 2022, for a quantitative assessment of the IMO 2020 Sulfur Cap.

2 Materials and Methods

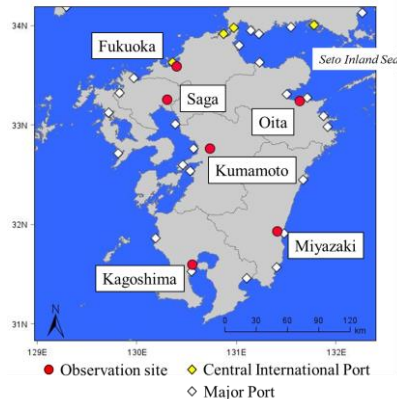


Fig.1. PM_{2.5} monitoring sites and ports in the Kyushu region, Japan

2.1 Study area and measurement data

We selected the Kyushu region as the case study. The region is located in western Japan, bordering the Seto Inland Sea, where the marine vessel traffic is among the highest in Japan. The region has multiple air pollution sources, including shipping activities near two major international ports in Fukuoka and other ports across Kyushu, industrial areas, high-traffic roads, residential villages, and agricultural zones. Fig. 1 shows the six PM_{2.5} monitoring sites in the Kyushu region, Japan.

Chemical composition data at the six PM_{2.5} monitoring sites in the Kyushu region (2016-2022) were sourced from the official website of the Government of Japan (MOE;

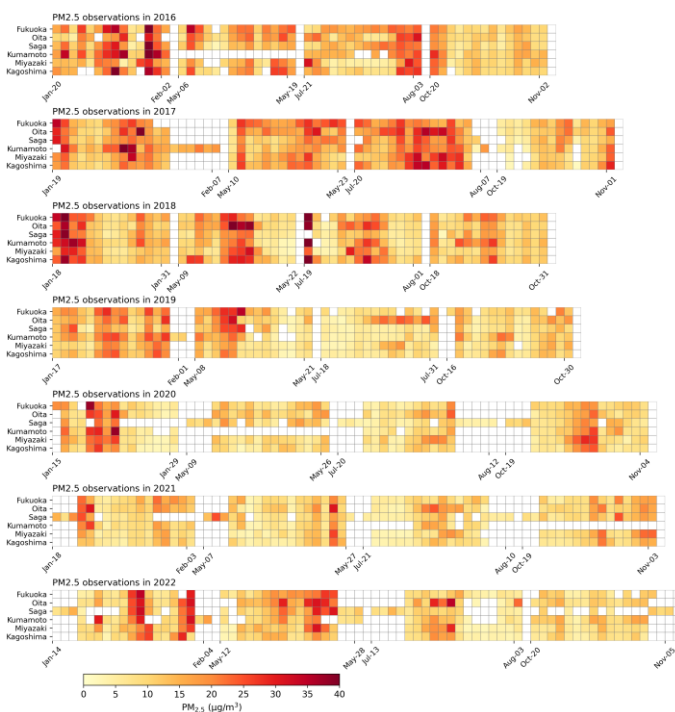


Fig.2. Sampling date and daily mean $PM_{2.5}$ variations in this study

<http://www.env.go.jp/air/osen/pm/monitoring.html> ;accessed September 20, 2025) and used for is used for PMF analysis. The $PM_{2.5}$ chemical composition measurements were conducted by the local authorities at the monitoring sites and based on the standardized measuring procedures for $PM_{2.5}$ components defined under the Air Pollution Control Act [11]. At each site, $PM_{2.5}$ samples were collected over 24-hour periods with polytetrafluoroethylene and quartz filters. The samples were analysed for weight of total mass, concentrations of $PM_{2.5}$ components (metallic elements, organic and elemental carbon (OC and EC), and the water-soluble ionic species) [11]. More than 14-day sampling campaigns were carried out per season (Fig.2).

2.2 Configuration of the PMF model

EPA PMF v5.0 was applied for estimating $PM_{2.5}$ source contributions using the chemical composition dataset aggregated across all six sites in the Kyushu region. Systematic analysis procedures using the model can be found in the PMF user guide and previous studies [11-12]. The PMF model was applied for all six sites. Note that aggregating data from multiple sites may limit the ability to resolve PMF factors influenced by local emissions, which could contribute only partially to $PM_{2.5}$ at individual sites. Dataset preparation for the PMF model followed our previous practice [11]. For example, outliers in total $PM_{2.5}$ mass and $PM_{2.5}$ component concentrations were excluded from the dataset. $PM_{2.5}$ component concentrations identified as having issues were removed based on ion balance and the chemical mass closure model. The analysis included 21 variables across 2,236 samples: $PM_{2.5}$, Ca^{2+} , K^+ , Mg^{2+} , Na^+ , NH_4^+ , Cl^- , NO_3^- , SO_4^{2-} , Al, As, Ba, Cu, Fe, Ni, Pb, Sb, V, Zn, OC, and EC. EPA PMF v5.0 provides error estimation tools (Displacement and bootstrap). The analyses were examined to assess the robustness of the model solution. Displacement did not show any factor swapping, and bootstrap results showed mapping rates exceeded 88% for all factors.

3 Results and Discussion

3.1 Characteristics of PM_{2.5} composition

Temporal changes in daily mean PM_{2.5} concentrations during the study period are shown in Fig.1. Between 2016 and 2022, the percentages of days exceeding 25 $\mu\text{g}/\text{m}^3$ were 12.1%, 13.9%, 12.0%, 4.9%, 4.0%, 1.9%, and 8.9%, respectively. Overall, the values exhibited a decreasing trend, with the exception of 2022. The earlier studies suggested that reductions in PM_{2.5} levels across Japan between 2013 and 2016 were partly driven by decreased transboundary pollution associated with China's air pollution control measures [13]. These measures have continued beyond 2016, and their influence may have contributed to the decreasing trend from 2016 to 2021.

Fig. 3 shows the interannual variations in mean concentrations of PM_{2.5}, V, and Ni (indicators of heavy oil combustion) in the Kyushu region. At all six sites, both V and Ni concentrations dropped from 2016 to 2020, followed by a slight increase in 2021–2022. In particular, Fukuoka and Saga, which are located near the Seto Inland Sea with high ship traffic, showed markedly higher concentrations of these elements compared with the other four sites. Moreover, the decreasing trends of these elements remained till 2020, when the marine fuel regulations in 2020 was implemented, was also more pronounced at Fukuoka and Saga. Based on these results, although a substantial decrease in PM_{2.5} concentrations were observed till the marine fuel regulations issued in 2020, the influence of other sources, such as transboundary pollution, was also present, making it difficult to accurately estimate the effect of the fuel oil regulation.

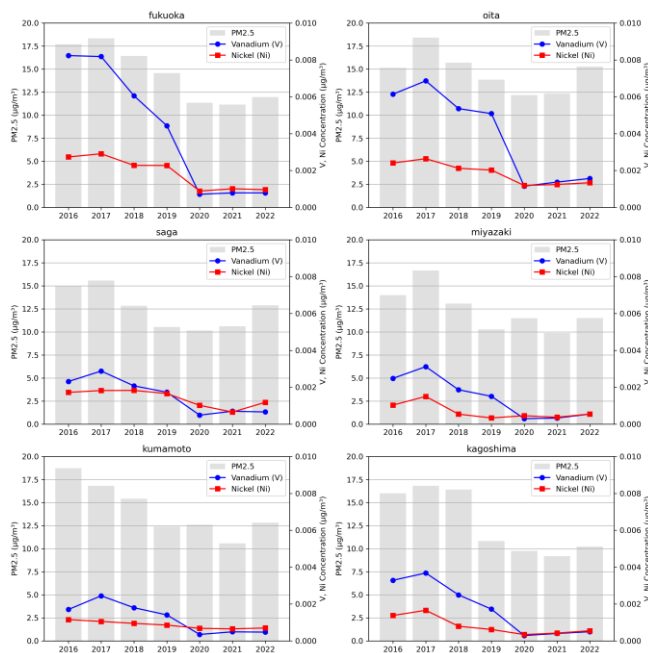


Fig.3. The interannual variations in mean concentrations of PM_{2.5}, V, and Ni at this study

3.2 PM_{2.5} source apportionment

We executed the PMF analysis to quantify PM_{2.5} contributions from emission sources. The PMF model extracted seven factors. These factors were identified by considering their chemical compositions, seasonal variations in their contributions, and previously reported studies [7, 11]. The factors were interpreted to the following source categories: heavy oil combustion, secondary nitrate, secondary sulfate mixed with biomass burning, soil and industrial emissions, sea salt, coal combustion, and road transportation. The factors' PM_{2.5} component profiles are shown in Fig.4. Fig.5 illustrates the annual and seasonal mean PM_{2.5} source contributions averaged across the six sites in the Kyushu region.

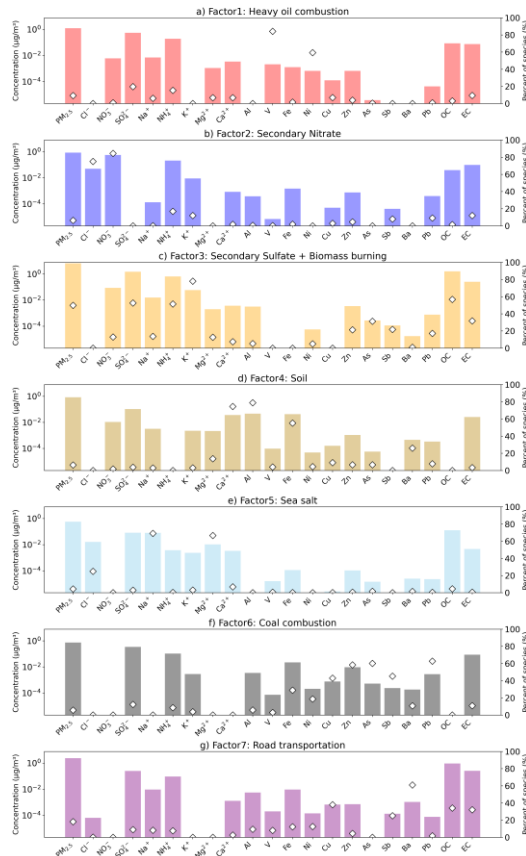


Fig.4. PM_{2.5} source concentrations (bars) and percent of species (circle) classified by the PMF model

Factor 1's chemical profile showed higher loadings of NH_4^+ , SO_4^{2-} , Ni, and V (Fig. 4a). In Japan, Ni and V are commonly recognized as indicators of heavy oil combustion, particularly, ship emissions at the sites near ports [7, 11]. Factor 1 is thus primarily influenced by ship traffic in coastal areas, with an average contribution of 8.9%.

Factor 2 contained significant fractions of NO_3^- , NH_4^+ , and several other components (Fig. 4b). Higher contributions were observed during cold season (winter) at all sites (Fig. 5). This factor is interpreted as being dominated by NH_4NO_3 , a semi-volatile species that volatilizes at higher temperatures, consistent with previous studies using similar tracers [7, 11]. The average contribution of "Secondary nitrate" was 6.6%.

Factor 3 accounted for a substantial fraction of SO_4^{2-} , and NH_4^+ (Fig. 4c). With an ion balance of 0.87, this factor represents a source associated with $(\text{NH}_4)_2\text{SO}_4$, which is stable

and can undergo long-range transport. The previous study indicated that the Kyushu region is strongly affected by transboundary pollution [2, 13]. Factor 3 also exhibited high proportions of K^+ , EC, and OC, markers of biomass burning. Accordingly, this factor was classified as “Secondary sulfate mixed with biomass burning,” with an average contribution of 49.2%.

The $PM_{2.5}$ chemical components for factor 4 consisted of crustal elements (Ca^{2+} , Al, and Fe) (Fig. 4d), and has been recognized as the “Soil” factor in the previous study [11]. Its higher contribution in spring suggests an influence of Asian dust events (Fig. 5). The average contribution of the “Soil” factor was 6.1%.

Factor 5 was composed of large amounts of Cl^- , Na^+ , and Mg^{2+} (Fig. 4e). The Mg^{2+}/Na^+ ratio (0.25) was close to the seawater reference value (0.23–0.24) reported by the previous study [14], indicating that this factor can be interpreted as originating from sea salt. The equivalent ratio of Cl^-/Na^+ was 0.20, which is close to the previous study [7], suggesting the occurrence of chlorine loss from sea salt. The average contribution of “Sea salt” was 4.8%.

Factor 6 accounted for a substantial levels of As, and Pb (Fig. 4f). Moreover, SO_4^{2-} , NH_4^+ are primary components in Factor 6, suggesting the contribution of $(NH_4)_2SO_4$. Arsenic (As)-containing atmospheric particles are generally considered tracers of coal combustion [7, 11]. The presence of EC further suggests contributions from fossil fuel use. Considering the widespread use of coal in industrial sources and power generation in continental Asia, this factor is interpreted as “Coal combustion” and had an average contribution of 6.4%.

Factor 7 exhibited high contributions of Cu, Sb, Ba, OC, and EC (Fig. 4g). While OC and EC indicate combustion-related emissions, Cu, Sb, and Ba are typically associated with brake wear [7]. These combined features suggest traffic-related emissions “Road transportation”, including both exhaust and non-exhaust sources from road vehicles. This factor accounted for 19.3% of $PM_{2.5}$ on average.

Across the Kyushu region, a marked decrease in yearly mean $PM_{2.5}$ concentrations was observed between 2017 and 2021, primarily attributable to reductions in Factor 1 (heavy oil combustion: $2.3 \mu\text{g}/\text{m}^3$ in 2017 vs $0.3 \mu\text{g}/\text{m}^3$ in 2021) and Factor 3 (secondary sulfate + biomass burning: $8.7 \mu\text{g}/\text{m}^3$ in 2017 vs $5.3 \mu\text{g}/\text{m}^3$ in 2021). While Factor 3 exhibited a gradual decline in $PM_{2.5}$ contribution each year, Factor 1 showed a sudden decrease in $PM_{2.5}$ contribution beginning in 2020. Factor 3 is characterized by $(NH_4)_2SO_4$, largely originating from transboundary pollution. Given that Kyushu is located at the western edge of Japan and is particularly susceptible to the influence of transboundary pollution from China and other regions [2], it is reasonable to infer that China’s air pollution control measures, which have mitigated transboundary transport, have strongly influenced the variations in Factor 3 and contributed to the decline in $PM_{2.5}$ concentrations in western Japan [13].

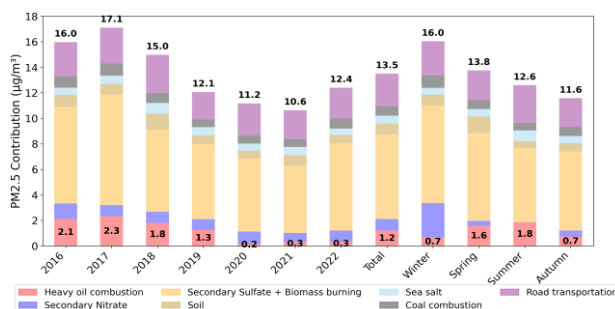


Fig.5. Mean source contributions estimated by the PMF model from 2016 to 2022 in the Kyushu region. See Fig. 2 for details of the observation days used for the seasonal mean contributions.

3.3 Evaluation of low-sulfur marine fuel regulations on PM_{2.5}

Fig. 6 shows the PM_{2.5} contribution from heavy oil combustion at the six sites in the Kyushu region identified by the PMF analysis, comparing the three years preceding (2017-2019) and following (2020-2022), low-sulfur marine fuel regulations in 2020. During 2017-2019, heavy oil combustion factor showed relatively high contributions in Fukuoka and Oita (2.8 and 3.1 $\mu\text{g}/\text{m}^3$, respectively), whereas the other four sites exhibited lower contributions, ranging from 1.0 to 1.4 $\mu\text{g}/\text{m}^3$. This elevated contribution in Oita is likely attributable to its location facing the Seto Inland Sea, which has heavy ship traffic, while Fukuoka hosts a major port, resulting in a higher density of vessel navigation compared to the other sites. In contrast, during 2020-2022, PMF-estimated PM_{2.5} contributions decreased dramatically at all sites, with reductions ranging from 65% to 97%. Although the magnitude of reduction varied among the sites, the consistent decline in contributions from heavy oil combustion after 2020 clearly indicates that low-sulfur marine fuel regulations in 2020 affected, at least in part, PM_{2.5} level drop in the Kyushu region.

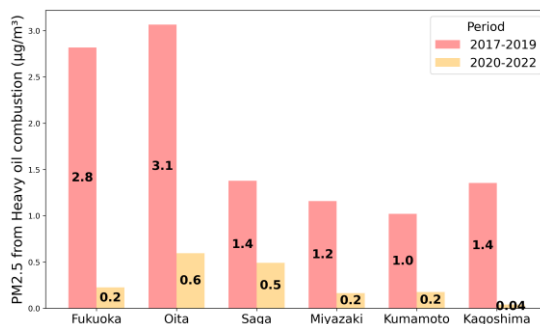


Fig.6. Comparison of the 3-years mean contributions from heavy oil combustion in the Kyushu region

4 Conclusion

This study quantified the PM_{2.5} contributions from shipping emissions in the Kyushu region, Japan, using the PMF model with chemical composition data from six monitoring sites between 2016 and 2022. The model identified seven major source factors: secondary sulfate and biomass burning (49.2%), road transportation (19.3%), heavy oil combustion (8.9%), secondary nitrate (6.6%), coal combustion (6.4%), soil (6.1%), and sea salt (4.8%). Among these, secondary sulfate showed a steady decrease throughout the study period, consistent with the influence of strengthened air quality control measures in China, which have reduced long-range transported pollution. In contrast, heavy oil combustion, strongly associated with shipping emissions, exhibited a sharp reduction beginning in 2020. During 2017-2019 and 2020-2022, contributions from heavy oil combustion declined by 65-97% across all sites. The most pronounced decreases were observed in Fukuoka and Oita, where shipping traffic density is particularly high. These reductions are consistent with low-sulfur marine fuel regulations in 2020, suggesting that international maritime fuel regulations contributed substantially to lowering PM_{2.5} level in the Kyushu region. This study highlights the importance of integrating long-term observational data with receptor modeling to assess the impacts of international emission policies. By distinguishing between transboundary influences and domestic sources, we demonstrated that global-scale regulatory measures (the IMO Sulfur Cap 2020), could yield tangible regional benefits for air quality. Continued monitoring and analysis are essential to evaluate the persistence of these improvements and

to understand their interactions with broader air quality management and climate mitigation strategies. Future research should incorporate higher-resolution emission inventories and monitoring networks to better capture spatial variations in shipping influences. Integrating receptor-based approaches with chemical transport models will provide a more comprehensive assessment of policy impacts.

References

1. Zhang Q., Jiang X., Tong D., Davis S.J., Zhao H., Geng G., Feng T., Zheng B., Lu Z., Streets D.G., et al., Transboundary health impacts of transported global air pollution and international trade. *Nature* **543**(7647):705–709 (2017).
2. Shimadera H., Kojima T., Kondo A., Evaluation of Air Quality Model Performance for Simulating Long-Range Transport and Local Pollution of PM_{2.5} in Japan. *Advances in Meteorology* (2016).
3. IMO (2020) Introduction to IMO. International Maritime Organization. <https://www.imo.org/en/About/Pages/Default.aspx> (accessed September 20, 2025)
4. Liu, H., Fu, M., Jin, X. et al. Health and climate impacts of ocean-going vessels in East Asia. *Nature Clim. Change* **6**, 1037–1041 (2016). <https://doi.org/10.1038/nclimate3083>
5. Shi, Z., Endres, S., Rutgersson, A., Al-Hajjaji, S., Brynolf, S., Booge, D., et al., Perspectives on shipping emissions and their impacts on the surface ocean and lower atmosphere: an environmental-social-economic dimension. *Elem. Sci. Anthropol.* **11**:00052 (2023).
6. Lv, Z., Liu, H., Ying, Q., Fu, M., Meng, Z., Wang, Y., et al., Impacts of shipping emissions on PM_{2.5} pollution in China. *Atmos. Chem. Phys.* **18**:15811–15824 (2018).
7. Nakatsubo R., Oshita Y., Aikawa M., Takimoto M., Kubo T., Matsumura C., Takaishi Y., Hiraki T., Influence of marine vessel emissions on the atmospheric PM_{2.5} in Japan's around the congested sea areas. *Sci. Total Environ.* **702**:134744 (2020).
8. Yu G., Zhang Y., Yang F., He B., Zhang C., Zou Z., Yang X., Li N., Chen J., Dynamic Ni/V ratio in the ship-emitted particles driven by multiphase fuel oil regulations in coastal China. *Environ. Sci. Technol.* **55**(22):15031–15039 (2021).
9. Zhang, X., Zhang, Y., Liu, Y., Zhao, J., Zhou, Y., Wang, X., Yang, X., Zou, Z., Zhang, C., Fu, Q., Xu, J., Gao, W., Li, N., Chen, J., Changes in the SO₂ Level and PM_{2.5} Components in Shanghai Driven by Implementing the Ship Emission Control Policy. *Environ. Sci.* **53**:11580–11587 (2019).
10. Tauchi M., Yamaji K., Nakatsubo R., Oshita Y., Kawamoto K., Itano Y., Hayashi M., Hiraki T., Takaishi Y., Futamura A., Evaluation of the effect of Global Sulfur Cap 2020 on a Japanese inland sea area. *Case Stud. Transp. Policy.* **10**(2):785–794 (2022)
11. Uranishi K., Ikemori F., Nakatsubo R., Shimadera H., Kondo A., Kikutani Y., Asano K., Sugata S., Identification of biased sectors in emission data using a combination of chemical transport model and receptor model. *Atmos Environ.* **166**:166–181 (2017)
12. Norris, G., Duvall, R., Brown, S., Bai, S., EPA positive matrix factorization (PMF) 5.0 Fundamentals and user guide. *U.S. Environ. Prot. Agency.* EPA/600/R-14/108 (2014).
13. Uno, I., Wang, Z., Yumimoto, K., Itahashi, S., Osada, K., Irie, H., Yamamoto, S., Hayasaki, M. and Sugata, S., Is PM_{2.5} Trans-boundary Environmental Problem in Japan dramatically improving?, *J. Jpn. Soc. Atmos. Environ.*, **52**: 177-184 (2017)
14. Keene, W.C., Pszenny, A.A.P., Galloway, J.N., Hawley, M.E., Sea-salt corrections and interpretation of constituent ratios in marine precipitation. *J. Geophys. Res.* **91**: 6647–6658 (1986).