

## **A: Procedures for PMF Analysis**

The following describes the procedures for PMF analysis. These procedures were developed based on the *EPA PMF 5.0 User Guide*. It should be noted that this document does not cover all aspects of PMF analysis, and the responsibility for the analysis results remains with the analyst. This document must be used with sufficient understanding of the EPA PMF 5.0 User Guide. Since the User Guide is assumed to have been thoroughly reviewed beforehand, distinctions between standardized interpretations and official procedures are not indicated here.

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### **(1) Software and Analytical Procedures**

The software used for analysis was *EPA PMF 5.0*, which is publicly available free of charge by the U.S. EPA. Analysts are required to read the EPA PMF 5.0 User Guide thoroughly and follow procedures consistent with the Use Guide. Records should be maintained regarding data cleaning, parameter decisions (e.g., number of factors), and other analytical settings to ensure traceability and reproducibility.

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### **(2) Creation of the Data Matrix**

#### **① Preparation of Concentration Data**

Dataset of concentration will be constructed using the measured chemical components. It is essential to design the chemical analysis carefully to ensure that source-specific marker compounds are included in the dataset.

Measurements *below the detection limit* were replaced with **one-half of the detection limit**.

If only a specific component is missing in a sample, a value of **-999** is assigned, and later converted to the median value through software processing to increase analytical uncertainty.

#### **② Preparation of Uncertainty Data**

There are two approaches to preparing uncertainty dataset: entering a single detection limit value and measurement error for each species, or calculating uncertainty using the detection limit and an error fraction assigned to each species.

The software parameter *Extra Modeling Uncertainty* is set to 0% by default, but it can be increased if necessary, such as when the model fails to converge.

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### **(3) Weighting of Components**

**PM2.5 should be set as “Weak”.**

(Software setting: *Total Variable (Defaults to Weak)*)

Based on the **Signal-to-Noise (S/N) ratio**, component weights are adjusted:

<b>S/N Range</b>	<b>Treatment</b>
$0 \leq S/N < 0.5$	“Bad” (excluded from analysis)
$0.5 \leq S/N < 1$	“Weak” (uncertainty $\times 3$ )
$S/N \geq 1$	“Strong” (no adjustment)

It may be reasonable to classify components with more than 50% of values below detection limits as “Bad” and exclude them. Excluding them during the dataset construction stage is generally more practical.

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#### **(4) PMF Analysis**

##### **① Preliminary Calculation for Component Weighting**

Using the weighting rules described in section (3), factor numbers are varied between **3 and 12**, and approximately **20 runs** are performed. Fixing the random seed is advisable to ensure reproducibility.

The model outputs **Q(true)** and **Q(robust)** are compared with the theoretical **Q(Theory)** values, and the relative standard deviation across runs is examined.

-Since Q(true) and Q(robust) depend on parameter settings, deviations from Q(Theory) at this stage are not considered critical.

-The reproducibility of each species in the model output can be used to inform decisions regarding parameter weighting, which may improve the accuracy of the calculation. However, since modifying the weighting can substantially alter the analytical results, it may not be necessary at this stage to discuss the number of factors or the interpretation of source profiles in detail.

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##### **② Recalculation with Updated Weighting (recommended when higher accuracy is desired; although, based on existing reports, it is not always performed)**

For components with a coefficient of determination ( $R^2 < 0.5$ ) between observed and predicted values, weighting is changed to “Weak” and the analysis is rerun.

As in step ①, the Q metrics are evaluated to determine the number of factors. Source identification is attempted based on factor profiles.

-Multiple candidate solutions may exist at this stage.

-A **Bootstrap run (approx. 20 iterations)** is conducted to confirm the robustness of included components.

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### ③ Final Computation Toward the Best Solution

Based on the appropriate analytical conditions identified in step ②, the model is rerun **100 times**.

Additional evaluations are conducted using model functions, including:

**Bootstrap**

**F<sub>peak</sub>**

**DISP**

**BS-DISP**

These are used to assess stability and robustness before finalizing the factor number.

Example Bootstrap evaluation criteria:

Mapping rate of  $\geq 80\%$  with zero unmapped factors

PM<sub>2.5</sub> and indicator components aligning with interquartile ranges (no missing key markers)

**(recommended when higher accuracy is desired; although, based on existing reports, it is not always performed)**

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### ④ Final Solution Determination

Factor profiles are examined, and source identification is performed based on major contributing tracer species.

As described in the next section (*Evaluation of Interpretation Validity*), results are validated. If many inconsistencies are found, the analysis is repeated, including dataset reconstruction if necessary.

As described in the next section, factor interpretation must be validated using multiple approaches. The final solution should be consistent with the general understanding of atmospheric aerosol sources. If discrepancies arise, the results may still be presented; however, sufficient justification must be provided to demonstrate that the interpretation is valid.

### **B: Methods for Validating the Appropriateness of Factor Interpretation**

The following describes the methods for validating the appropriateness of the PMF results. The validity of factor interpretation is evaluated using the criteria listed below. These criteria are developed through reference to published literature. Although it is not mandatory to strictly follow these criteria, they can serve as indicators during interpretation. However, in cases where the interpretation clearly contradicts these criteria, justification must be provided. If many items do not match, analysts should reconsider the dataset composition and the number of factors.

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### (1) Items to Check for All Factors

Whether the average contribution ratio of each extracted factor deviates significantly from expected contributions.

Whether consistency is observed with geographical or site characteristics (particularly when multi-site datasets are analyzed).

#### Examples:

Sea salt → higher at coastal sites than inland

Road traffic → higher near roadside sites than background sites

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### (2) Items to Check for Each Factor

The characteristics of each factor, including factor profiles, component ratios, and seasonal behavior in contribution levels, are evaluated using the reference criteria below. The factors listed here are examples and will not necessarily be extracted in every analysis. When unidentified factors are extracted, analysts must verify whether unique emission sources exist at the sampling location. Additionally, reviewing previous studies that reported similar factor types is recommended to assess consistency. It should also be noted that these criteria are based on a limited number of published studies and may not apply in all cases.

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#### Road Traffic

- Detected loads of OC and EC.
- If loads of inorganic elements such as Mn, Fe, Cu, Ni, Zn, and Sb are observed in addition to OC and EC, the factor may include the influence of brake wear, tire wear, road dust, or engine oil (Cheng et al., 2015; Bressi et al., 2014; Sahu et al., 2011; Wang et al., 2013; Waked et al., 2014).
- The OC/EC (or EC/OC) ratio in the factor profile is close to reported values in tunnel studies or other primary emission datasets (calculated using concentration-based values) (Cheng et al., 2015; Bressi et al., 2014; Waked et al., 2014).
- OC/EC ratio is often **less than 1** when the factor represents direct automobile exhaust emissions (Cheng et al., 2015; Waked et al., 2014).
- Lower EC/OC ratio indicates a higher contribution from gasoline vehicles, while a higher ratio indicates greater diesel influence (Sahu et al., 2011; Wang et al., 2013).
- No clear seasonal pattern is typically observed (Wang et al., 2013).

- Contribution values tend to be higher on weekdays and lower on weekends (Waked et al., 2014).
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### Sea Salt

- Fresh sea salt typically shows high loads of  $\text{Cl}^-$ ,  $\text{Na}^+$ , and  $\text{Mg}^{2+}$  (Waked et al., 2014).
  - For fresh sea salt, the  **$\text{Cl}^-/\text{Na}^+$  ratio** and  **$\text{Mg}^{2+}/\text{Na}^+$  ratio** are close to seawater composition values (1.17 and 0.11 respectively) (Bressi et al., 2014; Cohen et al., 2012).
  - Aged sea salt may exhibit replacement of Cl by S ( $\text{SO}_4^{2-}$ ) or N ( $\text{NO}_3^-$ ) (Wang et al., 2013; Waked et al., 2014).
    - If the  $\text{Cl}^-/\text{Na}^+$  ratio is lower than the seawater ratio **and** the  $\text{SO}_4^{2-}/\text{Na}^+$  ratio exceeds **0.060**, aging via acid displacement (chlorine loss) is suspected (Bressi et al., 2014).
    - If  $\text{NO}_3^-$ , EC, V, or Ni are present, the factor may contain a mixture of shipping emissions (Bressi et al., 2014).
    - Particularly in Pacific coastal regions, contributions tend to increase in summer when sea breeze activity is frequent (Wang et al., 2013; Waked et al., 2014).
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### Sulfate

- Ion balance between anions and cations is close to 1:1.
  - When OC and EC are present, the OC/EC ratio tends to be high (indicating secondary formation) (Bressi et al., 2014; Wang et al., 2013).
  - As a secondary aerosol, contributions tend to be high from spring to summer and lower in winter (Sahu et al., 2011; Mantas et al., 2014; Wang et al., 2013).
  - In autumn or winter, peaks may occur concurrently with nitrate aerosol peaks (Waked et al., 2014).
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### Sulfate (Heavy Oil Combustion)

- Elevated V, Ni, and  $\text{SO}_4^{2-}$  loads are observed (Bressi et al., 2014; Waked et al., 2014).
- When classified as a combustion source, EC should ideally be present.
- **V/Ni ratio:**
  - **2.1–3.1** → **ship emissions dominant**
  - **0.9–1.9** → **influence from stationary combustion sources** (Bressi et al., 2014; Pra et al., 2014)

- **Ni/V ratio indicators:**
    - **0.96 or 0.75** → **stainless steel or ceramic industry influence** (Waked et al., 2014)
    - **0.2–0.4** → **shipping or power plant influence** (Waked et al., 2014)
  - Absence of Na<sup>+</sup>, Cl<sup>-</sup>, and Mg<sup>2+</sup> suggests non-marine combustion sources (Waked et al., 2014).
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### **Industrial Dust**

- When Cd is included in the analysis, a **Pb/Cd ratio  $\leq 50$**  may indicate industrial influence (Bressi et al., 2014).
  - If Na, S, or EC loads are present, Na/S ratios should be checked to assess possible formation of Na<sub>2</sub>SO<sub>4</sub> from aged sea salt reactions (Cohen et al., 2012).
  - If affected by nearby industrial facilities, concentration patterns may lack clear periodicity (Cohen et al., 2012).
  - As primary particles, OC and EC loads are low or absent (Pandolfi et al., 2011).
  - Contributions may increase during periods of low boundary layer height due to regional emission characteristics (Pra et al., 2014).
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### **Soil or Resuspended Dust**

- Typical elevated components include Al, Ca, Ti, Si, and Fe (Cheng et al., 2015; Bressi et al., 2014; Sahu et al., 2011; Cohen et al., 2012; Mantas et al., 2014).
  - When Si is present, the **Al/Si ratio often falls between 0.25–0.35** (Cohen et al., 2012).
  - Where strong resuspension occurs, road-dust indicators (OC, EC, Ni, V, Cr, etc.) may also appear (Mantas et al., 2014; Waked et al., 2014).
  - Rb may appear due to erosion of soil and rock (Waked et al., 2014).
  - If sampling includes Asian dust events (Kosa), elevated contributions may occur during those periods.
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### **Nitrate**

- **NO<sub>3</sub><sup>-</sup>/NH<sub>4</sub><sup>+</sup> ratio  $\approx 4.0$**  (Pandolfi et al., 2011).
- Higher in winter (Wang et al., 2013).
- Cl<sup>-</sup> may also appear in some cases.

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## Biomass Burning

- High loads of **OC, EC, and K+ (or K)** are observed (Sahu et al., 2011; Cohen et al., 2012; Wang et al., 2013).
- Contribution peaks during agricultural burning periods such as straw burning (Sahu et al., 2011; Cohen et al., 2012; Mantas et al., 2014; Wang et al., 2013; Waked et al., 2014).
- OC/EC ratio is typically **2–4** (Waked et al., 2014).
- In open burning (including waste burning), components such as As, Rb, and Pb may be elevated in addition to K (Cheng et al., 2015; Waked et al., 2014).

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