

1 We are very grateful for the anonymous reviewer's positive assessments of the manuscript and
2 insightful comments for further improvement. We have revised the manuscript by fully taking the
3 reviewers' suggestions into account. Please find our point-to-point replies below in blue, and the
4 specific changes in the revised manuscript and SI are highlighted here in red.

5 **Reviewer 1**

6 Investigating the water solubility of SOA is a highly significant topic, because it has a
7 significant impact on its climatic effects. This work utilized carbon isotopic techniques and mass
8 spectrometry method to evaluate the water solubility of SOA with varying degrees of aging, basing
9 on one-year ambient PM_{2.5} data and established stable carbon isotope profiles of fresh and aged
10 SOA. This work found that SOA has high water solubility, and the solubility of aged SOA is higher
11 than that of fresh SOA. The finding of this work is of great significance for us to deeply understand
12 the climatic effects of SOA. There are certain issues that need to be addressed before considering
13 this work for publication.

14 1. The source apportionment based on offline data involves water-soluble ions and heavy metal
15 components. The relevant analysis methods should be briefly introduced in the main text and
16 described in detail in the Supplementary Information (SI). Quality control should also be briefly
17 explained.

18 **Response:**

19 Thank you very much for your valuable suggestions. The analysis methods of water-soluble
20 ions and heavy metal components have been added in line 105-110 in the revised manuscript, as
21 presented below:

22 'In addition, the samples collected by Teflon filter in this study were analyzed for water-soluble
23 ions (mainly SO₄²⁻, NO₃⁻, NH₄⁺, and Cl⁻) within PM_{2.5}, and the mass concentrations of twenty-three
24 metallic elements (primarily Na, Mg, Al, K, Ca, V, Fe, Ni, Zn, Pb, and Cd) within PM_{2.5} were also
25 determined using an inductively coupled plasma mass spectrometer (ICP-MS, Aurora M90; Bruker,
26 Germany). Relevant quality control information is described in the Supplementary Information
27 (Text S1).'

28 The detailed quality control methods for PM_{2.5} components have also been included in line 32-
29 41 of the revised Supplementary Information (SI).

30 'The measuring methods for each component are described in the main text, and the
31 measurement processes were subjected to strict quality control as follows, which are also available
32 in our previous studies (Huang et al., 2019; Yan et al., 2022). The OC/EC analyzer was calibrated
33 using eight standard concentration gradients of sucrose solution prior to each sample analysis of the
34 carbonaceous fractions, with all standard curves achieving an R² value exceeding 0.999. The charge
35 concentration balance of water-soluble ions (R² = 0.98, slope = 0.87) confirmed the validity of the
36 measurement results for water-soluble ions. The spiked recoveries for all metal elements ranged
37 between 80 % and 120 % in this study. Furthermore, the background concentration of blank samples
38 and the reproducibility of the measurement results were evaluated during the determination of each
39 component, and all the results met the experimental requirements.'

41 2. The results of the PMF model should be explained in greater detail, including the explanation
42 of the source profiles identified by PMF and the evaluation of the model results.

43 **Response:**

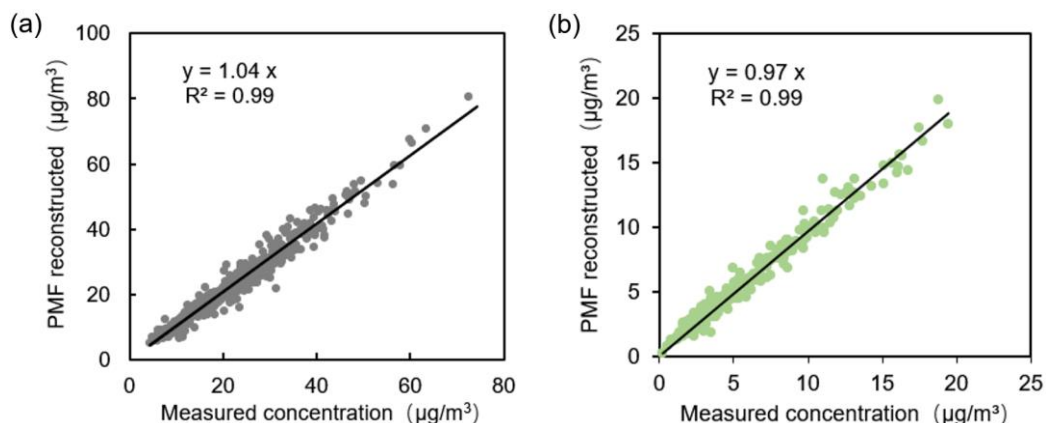
44 Thanks for your suggestion. Detailed explanations of the source profiles identified by PMF
45 and the evaluation of both PM_{2.5} and WSOC model results have been added to Text S1 of SI. The
46 added PMF results and evaluation for PM_{2.5} (line 42-59 in the revised SI) are as follows:

47 ‘To find out the optimal solution, factor numbers ranging from 5 to 11 were evaluated using
48 the PMF model. Among them, the nine-factor solution exhibited a notable covariance between
49 vehicle emissions and biomass burning sources, while the eleven-factor solution displayed a
50 dispersed distribution of Pb, Fe and Cd. Subsequently, the ten-factor solution was identified as
51 optimal due to its highly interpretable factor profiles (Fig. S2), with scaled residuals demonstrating
52 a generally symmetrically distribution between -3 and +3. There was a strong correlation between
53 the total mass of the input species and the total mass of all the model-reconstructed factors ($R^2 =$
54 0.99 , slope = 1.04) (Fig. S3), and favorable correlations were also observed between the source
55 contributions and their corresponding source markers ($R^2 = 0.83 \sim 0.96$), suggesting robust
56 performance of PMF model. According to Fig. S2, factor 1 exhibited high percentage explained
57 variation (EV) values for SO_4^{2-} (66 %) and NH_4^+ (59 %). In factor 2, not only OM and EC displayed
58 substantial EV values (49 % and 62 %), Zn and Fe also contribute notably. Factor 3 demonstrated
59 the highest EV values for the elements Na and Mg. Cl^- in factor 4 had an EV value of up to 82 %.
60 NO_3^- (67 %) and NH_4^+ (25 %) exhibited the highest EV values in factor 5. Factor 6 showed the
61 highest EV values for Pb, Cd and Zn, while factor 7 demonstrated the highest EV values for V and
62 Ni. Factors 8-10 exhibited the highest EV values for Ca (73 %), K (72 %) and Al (76 %), respectively.
63 Consequently, the ten factors were identified as secondary sulfate, vehicle emissions, aged sea salt,
64 coal combustion, secondary nitrate, industrial emissions, ship emissions, construction dust, biomass
65 burning, and fugitive dust, respectively.’

66 The added PMF results and evaluation for WSOC (line 68-81 in the revised SI) are as follows:

67 ‘In the source apportionment of WSOC, the mass concentration and uncertainty matrixes of
68 five species (CO_2^+ , C_4H_9^+ , $\text{C}_2\text{H}_4\text{O}_2^+$, WSOC, and WSOO) were put into the PMF model to identify
69 and calculate source contributions to WSOC. Following examination of a range of 2 to 4 factor
70 numbers, a three-factor solution output by the PMF model was determined to be optimal. The scaled
71 residuals exhibited a generally symmetrical distribution between -3 and +3 as well. Moreover, there
72 was also a strong overall correlation between the total factor concentrations reconstructed by the
73 PMF model and the total mass concentrations of the measured species ($R^2 = 0.99$, slope = 0.97) (Fig.
74 S3). According to Fig. S5, factor 1 displayed the highest percentage of EV values for m/z 44 (CO_2^+)
75 and WSOO (73 % and 63 %, respectively), with an oxygen-carbon ratio (O/C) of 1.01, which is
76 highly oxidized and identified as aged SOC source. Factor 2 exhibited EV values of 64% for m/z 57
77 (C_4H_9^+), 29% for WSOC, 27% for m/z 44, and 23% for WSOO. In addition, factor 2 had a lower
78 level of oxidation with an O/C ratio of 0.43, and was therefore identified as fresh SOC source. Factor
79 3 demonstrated a 100 % EV value for m/z 60 ($\text{C}_2\text{H}_4\text{O}_2^+$) and a low O/C ratio of 0.36, indicating that
80 factor 3 represented the biomass burning source (BB).’

81 In addition, Figure S3 was also added to SI to verify the PMF results.



82

83 **Figure S3.** Comparison between the measured total mass of species and the PMF reconstructed total
 84 mass of sources of (a) $\text{PM}_{2.5}$, (b) WSOC.

85 3. The uncertainty assessment of the Bayesian model is crucial, it is better to move Figure S5 to
 86 the main text.

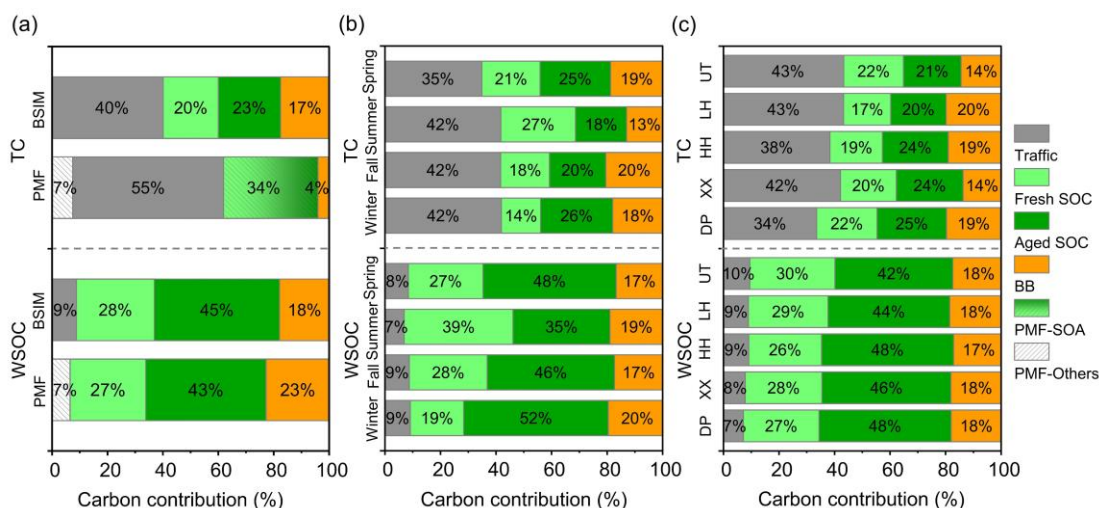
87 **Response:**

88 Thanks for your suggestion. Figure S5 has been moved to the main text (Now Figure 5 in the
 89 revised manuscript), as you suggested.

90 4. To ensure consistency and clarity, it is advisable to arrange the various sources in Figure 4(a)
 91 in a uniform order.

92 **Response:**

93 Thanks for your suggestion. The TC sources identified by PMF in Figure 4(a) have been
 94 arranged in a uniform order in the revised manuscript according to your advice.



95

96 **Figure 4.** (a) Comparison of source apportionment results between BSIM model and PMF model for TC
 97 and WSOC, (b) seasonal and (c) spatial distributions of source apportionment results for TC and WSOC
 98 based on the BSIM model.

99 5. Lines 305-306, the meanings of "[c]_{water-soluble}" and "[c]_{water-insoluble}" should be clearly
 100 explained to avoid any ambiguity.

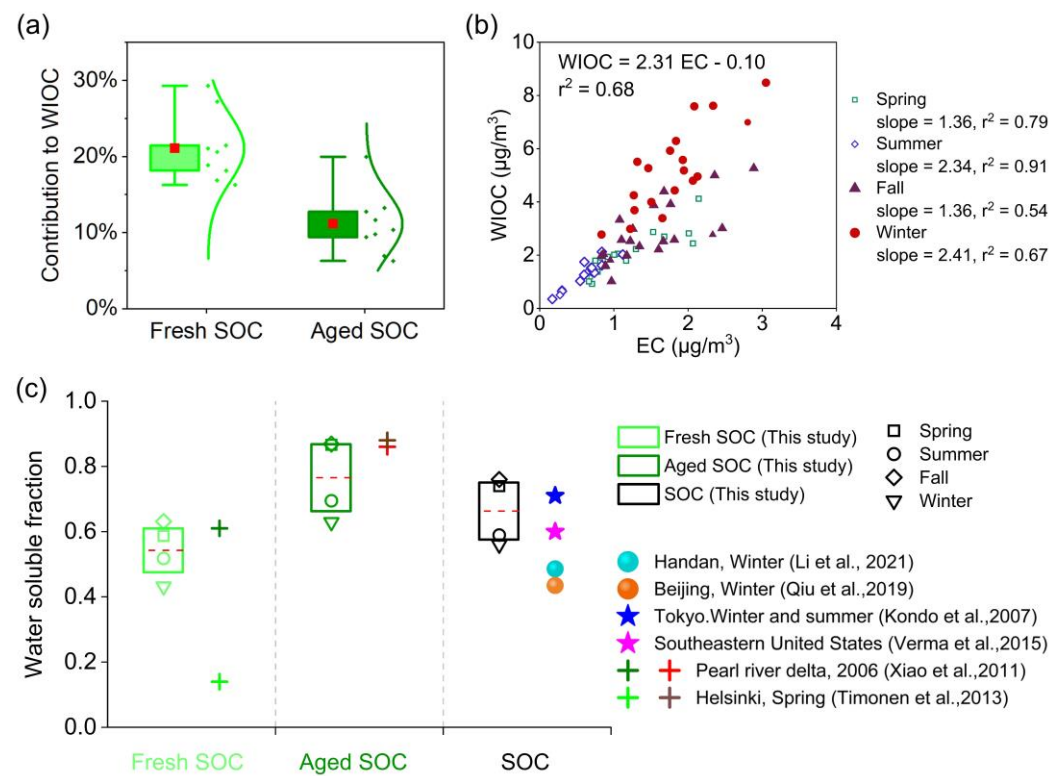
101 **Response:**

102 Thanks for your suggestion. Detailed explanations of the meaning of "[c]_{water-soluble}" and
 103 "[c]_{water-insoluble}" have been added in line 334-337 in the revised manuscript, as presented below:
 104 ‘we then calculate their water-soluble fraction by comparing their water-soluble portion to the
 105 ambient fraction ($[c]_{\text{water-soluble}} / ([c]_{\text{water-soluble}} + [c]_{\text{water-insoluble}})$), where $[c]_{\text{water-soluble}}$ and $[c]_{\text{water-insoluble}}$
 106 are the concentrations of fresh SOC or aged SOC in WSOC and WIOC, respectively) (Li et al.,
 107 2021).’

108 6. Figure 5(c) appears redundant as it overlaps with Figure 5(b) in terms of information
 109 presented. To streamline the content, it is advisable to include the slope information within Figure
 110 5(b).

111 **Response:**

112 Thanks for your comments. We have followed your suggestions and made corresponding
 113 adjustments to Figure 5 (Now Figure 6 in revised manuscript).



114 **Figure 6.** (a) Left is the box and whisker plots of fresh and aged SOC contributions to WIOC, the upper
 115 and lower of the box representing the 75th and 25th percentiles, and the red squares featuring mean
 116 values. The dots on the right show the contribution of fresh and aged SOC to WIOC across seasons and
 117 sites, the curve demonstrates its normal distribution. (b) Scatterplot of WIOC versus EC by season, (c)
 118 Comparison of the water-soluble fraction of SOC (fresh SOC, aged SOC, SOC) in this study (box and
 119 whisker plots) with those in other related literature (colored markings on the right). The upper and lower
 120 of the box represent the 75th and 25th percentiles and the dashed red lines indicate mean values.
 121

122 7. Line 38, the full name of CCN should be clearly listed at the first mention in the main text.

123 **Response:**

124 Thanks for your suggestion. The full name of CCN (cloud condensation nuclei) has been added
125 in line 38.