Dear Editor and Reviewers,

Thank you for the comments to help improve the quality of the paper. We have revised the manuscript to address your comments and a detailed response to each comment is provided in this file. The comments are in regular font and the responses are in red.

Evolution of atmospheric age of particles and its implications for the formation of a severe haze event in eastern China
Manuscript #: acp-2023-11

RC3, Reviewer #3:

Summary:

The concept of atmospheric age, which refers to the time that has passed since the emission or formation of an air pollutant, has not received much attention in previous studies. This manuscript attempted to addresses this gap by updating an age-resolved UCD/CIT model and using it to simulate the evolution of age distribution for different PM2.5 components during a severe regional haze episode in eastern China. The particle age information presented in this manuscript is interesting and provides a unique perspective on the formation and evolution of regional haze event, which is meaningful for future research in atmospheric chemistry and physics. Moreover, the manuscript is well written and easy to follow. I recommend to accept this manuscript after some minor revisions.

Response: Thanks for the recognition of our study. Below is the response to each specific comment.

Comments:

1. Lines 67-68: How to define the atmospheric age of secondary particles? Is that based on the time since the emission of precursors or the formation of secondary aerosols?

Response: The atmospheric age of secondary particles is defined as the time they have been suspended in the atmosphere since they are formed.

2. Lines 79-80: The key factor contributes to the changes in the hydrocarbon ratios is the difference in reaction rates of different hydrocarbons with oxidants such as OH radicals. Please modify this sentence to make it more clear.

Response: Thanks for the suggestion. We've modified this sentence as "Since the oxidation rates of these hydrocarbons by hydroxyl (OH) radicals vary widely, the hydrocarbon ratios change with photochemical aging" in the revision (lines 79-80).

3. Lines 90-96: The author mentioned the dynamic age-bin updating algorithm used in the UCD/CIT model is based on the method used previously in the CMAQ model. Is there any difference between them? It's better to provide more details.

Response: Thanks for the comments. The dynamic age-bin updating algorithm is based on Zhang et al. (2019), who also used the UCD/CIT model. The dynamic age-bin updating algorithm is used to dynamically update the concentrations of particles between different age bins, which can be represented in Eq. 1 as mentioned in the Response to Reviewer #1's comment #1. The same algorithm has also been used in CMAQ to track the age distribution of primary and secondary inorganic aerosols (Ying et al., 2021). To clarify, we've modified the description in this paragraph in the revision. Please see lines 88-100.

4. Lines 123-126: How many age bins are used? What's the time interval between different age bins? Also, What's the highest explicit age?

Response: A total of 9 age bins were used in this study, and the age bin updating frequency is set to 12 h. So the explicitly tracked particle age is 96 h. For particles with age > 96 h, their concentrations were assigned to the last age bin.

5. Line 130: τ_i in equation (2) should be $\overline{\tau_i}$.

Response: Corrected.

6. Lines 145-146: How to convert total emissions into different size bins? Please provide more details.

Response: Thanks for the comments. The UCD/CIT model uses a sectional method to represent the size distribution of aerosols. In this study, a total of 15 size bins, ranging from 0.01 to 10 μm in diameter, are used. Because the existing emission inventories, such as MEIC, only provide the total particulate emissions without their size distribution. The total particulate emissions obtained from MEIC and FINN were transformed into size-resolved emissions based on a library of primary particle source profiles measured during actual source tests (Hu et al., 2015). The fraction of total particulate emissions assigned to each bin is shown in Table S1 (also shown as Table R3-1 below). Please see lines 151-152 in the revision.

Table R3-1. Fractional apportionment of particle-phase emissions across the 15 size bins in the UCD/CIT model.

Size bin	Diameter (nm)	Emission fractions				
		Industry	Residential	Power	Transportation	Wildfire
1	<10	0	0.0001	0	0.0046	0.0000
2	10-16	0	0.0004	0	0.0103	0.0007
3	16-25	0	0.0019	0	0.0140	0.0046
4	25-39.5	0.0004	0.0104	0.0004	0.0284	0.0437
5	39.5-63	0.0239	0.0295	0.0239	0.0839	0.1632
6	63-100	0.2542	0.0931	0.2542	0.2617	0.2601
7	100-160	0.5139	0.2366	0.5139	0.2194	0.2637
8	160-250	0.1902	0.2756	0.1902	0.0970	0.1017
9	250-395	0.0172	0.2225	0.0172	0.0732	0.0379
10	395-630	0.0002	0.0727	0.0002	0.0680	0.0343
11	630-1000	0	0.0163	0	0.0145	0.0308
12	1000-1600	0	0.0409	0	0.0305	0.0260
13	1600-2500	0	0	0	0.0325	0.0162

14	2500-3950	0	0	0	0.0312	0.0081
15	3950-10000	0	0	0	0.0310	0.0091

Reference: Hu, J., Zhang, H., Ying, Q., Chen, S. H., Vandenberghe, F., and Kleeman, M. J.: Long-term particulate matter modeling for health effect studies in California – Part 1: Model performance on temporal and spatial variations, Atmos. Chem. Phys., 15, 3445-3461, 10.5194/acp-15-3445-2015, 2015.

7. Lines 155-156: So there are a total of 5 cases? Why not directly use the high time resolution simulation, such as with a time interval of 1 hour?

Response: Yes. There are a total of 5 cases. In this study, a sectional method was used to represent the atmospheric age of particles, and the total number of age bins was fixed as 9 in the model. Thus, this method can only represent a limited range of particle age. For example, the simulation with a time interval of 12 h can explicitly represent particles within 96 h (12×8), and particles older than 96 h are assigned to the last age bin. Note that with an updating interval of 12 h, particles with ages of 0-12 h (12-24h, 24-36h, etc.) are assigned to the same age bin. However, in urban areas, some particles may have been emitted or formed just several hours ago due to intensive anthropogenic emissions. To get relatively detailed age information, we ran 5 cases with updating intervals of 1, 3, 6, 8, and 12 h, and then combined them together. Although the 1-hour simulation can provide the highest age resolution, the explicitly represented particle age is only 8 hours. Thus, it's not suitable to study the age distribution of particles older than 8 hours.

8. Line 191: Figure S1 should be Figure S2. So as Figure S2 in Line 196 and Figure S3 in Line 201. The authors should check this throughout the manuscript.

Response: Thanks for the comments. All the figure citations have been checked and revised.

9. Line 229: How to convert the measured OC to OM? Is that based on an assumption of OM/OC ratio? Please clarify.

Response: Yes. The measured OC is converted to OM by multiplying a factor of 1.64. The factor used in this study is obtained from Tan et al. (2018), which is based on measurements with an aerosol mass spectrometer. We've added this in the revision (Lines 182-183).

Reference: Tan, T., Hu, M., Li, M., et al.: New insight into $PM_{2.5}$ pollution patterns in Beijing based on one-year measurement of chemical compositions, Sci. Total Environ., 621, 734-743, 10.1016/j.scitotenv.2017.11.208, 2018.

10. Figure 3: What time period does the result shown in Figure 3e correspond to? Additionally, the colors used for SO_4^{2-} and NH_4^+ are too similar, making it difficult to differentiate between them. Please change.

Response: Thanks for the comments. Figure 3e shows the IMR (incremental mass ratio) of the major PM2.5 chemical compositions, which is calculated based on the changes in the mass concentration of particles during the $PM_{2.5}$ growth stage. Thus, the time periods are different for different cities. For Beijing and Jinan, $PM_{2.5}$ concentration began to increase on Dec. 25 and reached its peak on Dec. 29, so the corresponding time period is Dec.25–29. For Nanjing and Shanghai, the corresponding time periods are Dec. 27–31 and Dec. 28–31, respectively. The colors in Figure 3 have been modified (see Figure R3-1 below).

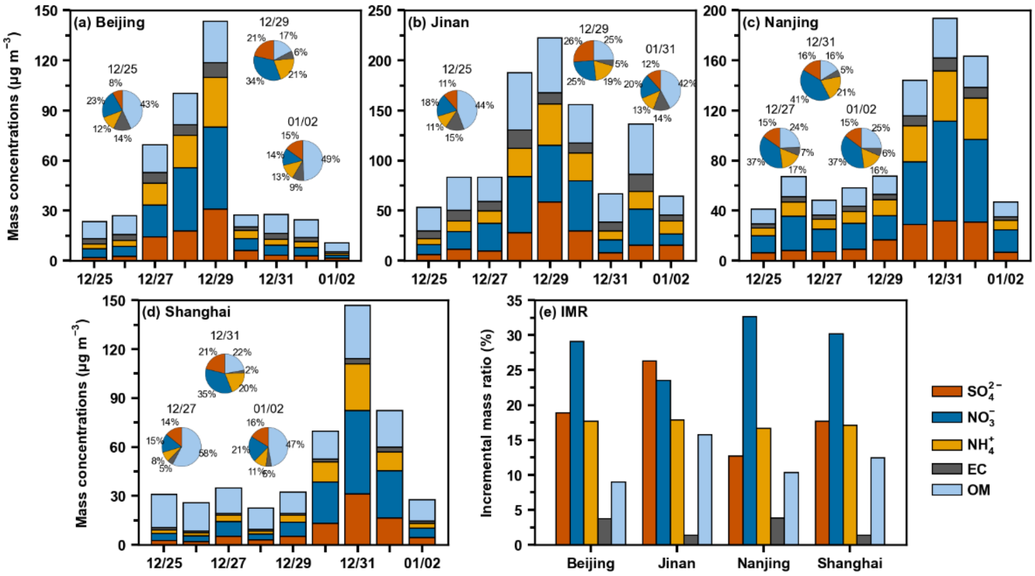

Figure R3-1. Mass concentrations and fractions **(a–d)** and incremental mass ratio (e) of the major $PM_{2.5}$ chemical compositions (SO_4^{2-}, NO_3^-, NH_4^+, EC, and OM) in Beijing, Jinan, Nanjing, and Shanghai.

11. Lines 316-317: I cannot find a "more uniform" vertical distribution for the period of accumulation stage from Figure 7. On the contrary, during the regional transport stage, the vertical distribution in the NCP region appears to be more uniform. This is because the air pollutants ahead of the cold front was lifted from the ground to the upper level.

Response: Thanks for the comments. From Figure 7, a "more uniform" vertical distribution in the NCP can be found during the accumulation stage when compared to that in the YRD. To be clear, we modified this sentence to "Compared to YRD, the vertical distribution of particle ages was more uniform in the NCP due to the accumulation of aged pollutants in the atmosphere under stable weather conditions" in the revision. We do agree that the vertical distribution in the NCP region appears to be more uniform during the regional transport stage when compared to that during the accumulation stage. We've added some more discussion in the revision (please see lines 333-339).

12. Lines 335-336: What is "fresh" particle? In Line 272, the author mentioned fresh particles refer to particles with atmospheric age < 24 h, while they are particles with an age of less than 12 h here. Please clarify.

Response: Thanks for the comments. Fresh particles refer to particles with a low atmospheric age. In this study, we defined fresh particles as particles with atmospheric age < 24 h. To be clear, we added a definition in the revision. Please see lines 275-276. Also, in line 357, we've removed the word "fresh" to avoid misunderstanding.

13. Line 375: It should be "NO_3^- formed locally in YRD".

Response: Corrected.

14. Mistake in the reference in Line 558.

Response: Corrected.