General.

We would like to appreciate the editor and reviewers for providing the valuable comments and a better perspective on our work to improve the manuscript. In particular, we are very grateful to the editor and reviewers for giving us the opportunity to make revision. We have revised our manuscript by fully taking the reviewers' comments into account. Responses to specific comments raised by the reviewers are described below. All the changes made and appeared in the revised text are shown in red. All detailed answers to comments are displayed in blue.

Comments of Reviewer #2 and our responses to them

Comments:

This manuscript presents results from a detailed study of the chemical composition of aerosol particles collected at regular intervals over a year in Urumqi. The samples are characterized by soft ionization with UPLC-ESI-QToFMS and a focus is placed on the nitrogen containing molecular formulas identified in the mass spectra. The authors find differences in the composition of the CHON and CHN compounds between the colder and warmer periods and they attribute the majority of this difference to the variation in the fuels burned in the warmer period (wildfires) vs colder (combustion for heat). Overall, this is a very detailed and well carried out study that is clearly written. I have minor concerns about some of the data analysis and once these are addressed, I recommend acceptance of the manuscript.

Response: We appreciate the reviewer's valuable comments on our work. Our responses to the specific comments and changes made in the manuscript are given below.

Specific Comments:

1) *Thank you for providing the data for the figures. For the peak identification, how many of the measured peaks could not be identified? Was there a mass dependence to this (i.e., were there high mass peaks that were measured that could not be identified?).*

Response: We greatly thank you for your professional review of our article. These are very critical issues. In this study, the number of molecules that were excluded accounted for no more than 2% of the total number of molecules. Moreover, the signal intensity of these excluded peaks also accounted for no more than 2% of the total signal intensity.

In ESI+, the identified peaks were classified into several major compound categories based on their elemental compositions, including CHO, CHON, and CHN. The CH compounds were excluded because of their small number (0.43% of the total number of compounds in ESI+) and low signal intensity ($0.33 \pm 0.28\%$ of the total signal intensity in ESI+) being identified in this study. In ESI−, the identified peaks were classified into CHO, CHON, CHOS, and CHONS. S-containing compounds were not discussed in this work because of our focus on N-containing compounds.

For the compounds with high mass peaks (> 700 Da), their signal intensities accounted for 1.12% and 1.37% of total signal intensities in ESI+ and ESI−, respectively. Thus, these compounds were also excluded in discussion, as indicated by many previous studies

(Wang et al., 2021; Yuan et al., 2023; Xie et al., 2020). In general, the main conclusions of this study are unaffected by the exclusion of these compounds.

More descriptions have been added in **Sect S1**, which was shown below (Pages S4-S5).

Pages S4-S5: …The CH compounds were excluded because of their small number (0.43% of the total number of compounds in ESI+) and low signal intensity (0.33 ± 0.28% of the total signal intensity in ESI+) being identified in this study. For the compounds with high mass peaks (> 700 Da), their signal intensities accounted for 1.12% and 1.37% of total signal intensities in ESI+ and ESI−, respectively. Thus, these compounds were also excluded in discussion, as indicated by many previous studies (Wang et al., 2021; Yuan et al., 2023; Xie et al., 2020) …

2) *The sentence starting on line 55 is confusing and I recommend revising it: "Moreover, the modified forms of some nitrogen-containing organic compounds (NOCs) and volatile organic compounds (VOCs) by ozone (O3), hydroxyl radical (•OH), and nitrogen oxide (NOx) can lead to an increase in the health hazards of OA, among which nitrated amino acids and nitrated polycyclic aromatic hydrocarbons are two representative hazards (Franze et al., 2005; Bandowe and Meusel, 2017)." What does "modified forms" mean? The second half of the sentence (starting ...along with nitrated amino acids...) is also incomplete and may be better as its own sentence.*

Response: We apologize for the confusion caused by our expression and thank you for your suggestions. The expression "modified forms" refers to the products or intermediate products of the interactions of ozone (O_3), hydroxyl radical (•OH), and nitrogen oxide (NO_x) with some nitrogen-containing organic compounds (NOCs) and volatile organic compounds (VOCs). The revision has been made in the revised manuscript (Lines 55–60).

Lines 55–60: …Moreover, the further oxidation or nitrification of some nitrogen-containing organic compounds (NOCs) and volatile organic compounds (VOCs) by ozone (O_3), hydroxyl radical (•OH), and nitrogen oxide (NO_x) can lead to an increase in the health hazards of OA (Franze et al., 2005; Bandowe and Meusel, 2017). Nitrated amino acids and nitrated PAHs are two representative hazard NOCs (Franze et al., 2005; Bandowe and Meusel, 2017).

3) On line 238, the possibility for CHO compounds to be precursors for CHON compounds is raised. Please clarify if this is referring to possible reactions in the gas-phase, in the particle-phase, or both?

Response: The CHON compounds can be tightly associated with secondary formation processes involving the reactions of reactive nitrogen with gas-phase and particle-phase CHO compounds (Bandowe and Meusel, 2017; Zarzana et al., 2012; Laskin et al., 2014). For example, laboratory experiments have suggested that the oxidation of isoprene and α-/β-pinene in the presence of NO_x can form large amounts of CHON compounds (Surratt et al., 2010; Rollins et al., 2012; Nguyen et al., 2015). The reduced nitrogen species (e.g., NH_3 and NH_4^+) have been demonstrated to contribute to the formation of NOCs in particle-phase (Zarzana et al., 2012; Laskin et al., 2014). Xu et al. (2023) characterized the variations of molecular compositions in urban road $PM_{2.5}$, suggesting that organic nitrates increased largely through the interactions of atmospheric oxidants, CHO compounds, and aerosol liquid water in both gas-phase and particle-phase. In general, CHO compounds can be important precursors for the formation of NOCs (via reactions in the gas- and/or particle-phases).

More discussions are presented at Lines 71–89. Based on your suggestion, the revision was made in the Lines 250–252.

Lines 250–252: …This indicated that CHO compounds may be important precursors for the formation of NOCs (via reactions in the gas- and/or particle-phases) or that they have similar origins.

4) In Table S4: how were the identifications made that are in the footnote (a, b, c, d)? For this and other tables, how is “relatively high signal” defined?

Response: These compounds were identified or inferred based on their MS/MS fragments or the molecular formulae of the products obtained from **Figure 5.** More descriptions were added in the page S11.

Page S11: These compounds were identified or inferred based on their MS/MS fragments or the molecular formulae of the products obtained from Figure 5.

We apologize for the confusion caused by our expression. The correct expression is that "Characteristics of the observed CHON compounds with relatively high signal intensity compared to other CHON compounds in ESI+ mode in the warm period (Page S11). The titles of other tables (**Table S6** and **Table S7**) were also updated (Pages S13-S14).

5) *The mass error calculations here look to be a little incorrect (ppm values). I agree with the assignments and the errors I calculate are within the boundaries from the paper (5 ppm), but these values should be double checked.*

Response: The mass errors in **Table S5** were the results directly output by the data processing software. In general, mass error calculations for mass spectrometry (Brenton and Godfrey, 2010) can be expressed as follows:

$$\Delta m_i = \frac{(m_i - m_a)}{m_a} \times 10^6 \; in\; ppm\; (parts\; per\; million)$$

where m_i is the measurement value, m_a is the calculated mass value.

To check the accuracy of the mass errors obtained from the software, we added the theoretical masses of the ions detected by the instrument to **Table S5** (Page S12) and calculated the mass error for each compound according to the above equation. We found that the calculated results were consistent with the output values, indicating that the mass errors obtained from the instrument are reliable.

Once again, we deeply appreciate the time and effort you've spent in reviewing our manuscript.

Reference:

Bandowe, B. A. M. and Meusel, H.: Nitrated polycyclic aromatic hydrocarbons (nitro-PAHs) in the environment – A review, Sci. Total Environ., 581-582, 237-257, https://doi.org/10.1016/j.scitotenv.2016.12.115, 2017.

Brenton, A. G. and Godfrey, A. R.: Accurate mass measurement: Terminology and treatment of data, Journal of the American Society for Mass Spectrometry, 21, 1821-1835, 10.1016/j.jasms.2010.06.006, 2010.

Franze, T., Weller, M. G., Niessner, R., and Pöschl, U.: Protein Nitration by Polluted Air, Environ. Sci. Technol., 39, 1673-1678, https://doi.org/10.1021/es0488737, 2005.

Laskin, J., Laskin, A., Nizkorodov, S. A., Roach, P., Eckert, P., Gilles, M. K., Wang, B., Lee, H. J., and Hu, Q.: Molecular Selectivity of Brown Carbon Chromophores, Environ. Sci. Technol., 48, 12047-12055, https://doi.org/10.1021/es503432r, 2014.

Nguyen, T. B., Bates, K. H., Crounse, J. D., Schwantes, R. H., Zhang, X., Kjaergaard, H. G., Surratt, J. D., Lin, P., Laskin, A., Seinfeld, J. H., and Wennberg, P. O.: Mechanism of the hydroxyl radical oxidation of methacryloyl peroxynitrate (MPAN) and its pathway toward secondary organic aerosol formation in the atmosphere, Phys. Chem.

Chem. Phys., 17, 17914-17926, https://doi.org/10.1039/C5CP02001H, 2015.

Rollins, A. W., Browne, E. C., Min, K.-E., Pusede, S. E., Wooldridge, P. J., Gentner, D. R., Goldstein, A. H., Liu, S., Day, D. A., Russell, L. M., and Cohen, R. C.: Evidence for NO_x Control over Nighttime SOA Formation, Science, 337, 1210-1212, https://doi.org/10.1126/science.1221520, 2012.

Surratt, J. D., Chan, A. W. H., Eddingsaas, N. C., Chan, M., Loza, C. L., Kwan, A. J., Hersey, S. P., Flagan, R. C., Wennberg, P. O., and Seinfeld, J. H.: Reactive intermediates revealed in secondary organic aerosol formation from isoprene, P. Natl. Acad. Sci. USA, 107, 6640-6645, https://doi.org/10.1073/pnas.0911114107, 2010.

Wang, Y., Hu, M., Hu, W., Zheng, J., Niu, H., Fang, X., Xu, N., Wu, Z., Guo, S., Wu, Y., Chen, W., Lu, S., Shao, M., Xie, S., Luo, B., and Zhang, Y.: Secondary Formation of Aerosols Under Typical High-Humidity Conditions in Wintertime Sichuan Basin, China: A Contrast to the North China Plain, J. Geophys. Res.-Atmos., 126, e2021JD034560, https://doi.org/10.1029/2021JD034560, 2021.

Xie, Q., Su, S., Chen, S., Xu, Y., Cao, D., Chen, J., Ren, L., Yue, S., Zhao, W., Sun, Y., Wang, Z., Tong, H., Su, H., Cheng, Y., Kawamura, K., Jiang, G., Liu, C. Q., and Fu, P.: Molecular characterization of firework-related urban aerosols using Fourier transform ion cyclotron resonance mass spectrometry, Atmos. Chem. Phys., 20, 6803-6820, https://doi.org/10.5194/acp-20-6803-2020, 2020.

Xu, Y., Dong, X. N., He, C., Wu, D. S., Xiao, H. W., and Xiao, H. Y.: Mist cannon trucks can exacerbate the formation of water-soluble organic aerosol and $PM_{2.5}$ pollution in the road environment, Atmos. Chem. Phys., 23, 6775-6788, https://doi.org/10.5194/acp-23-6775-2023, 2023.

Yuan, W., Huang, R.-J., Shen, J., Wang, K., Yang, L., Wang, T., Gong, Y., Cao, W., Guo, J., Ni, H., Duan, J., and Hoffmann, T.: More water-soluble brown carbon after the residential "coal-to-gas" conversion measure in urban Beijing, npj Climate and Atmospheric Science, 6, 20, 10.1038/s41612-023-00355-w, 2023.

Zarzana, K. J., De Haan, D. O., Freedman, M. A., Hasenkopf, C. A., and Tolbert, M. A.: Optical Properties of the Products of α-Dicarbonyl and Amine Reactions in Simulated Cloud Droplets, Environ. Sci. Technol., 46, 4845-4851, https://doi.org/10.1021/es2040152, 2012.