

Comments

This manuscript presents an investigation into the sources and formation mechanisms of nitrogenous organic aerosols (ON) in an urban atmosphere, leveraging a new analytical technique for bulk ON quantification and high-resolution source apportionment. The study's strengths lie in its measurement approach, comprehensive source marker analysis, and identification of secondary ON formation pathways, including nitrate formation, photochemical oxidation, and DCA-associated reduced-N species. Several critical gaps in scientific rigor, data interpretation, and presentation weaken the manuscript's impact. Key concerns include incomplete source apportionment due to unclear presentation in methodology, insufficient mechanistic evidence for DCA_ON formation. Addressing these issues would significantly strengthen the manuscript's contribution to atmospheric chemistry and air quality research.

1 The method used for total nitrogen estimation in this study is not commercially available. Additional details regarding the calibration procedure for total nitrogen quantification using this instrument should be provided. The IN and ON datasets should be cross-validated using independent measurements. For instance, it would be important to demonstrate whether water-soluble inorganic nitrate concentrations correlate strongly with the inorganic nitrogen values quantified in this study.

2 The terms "Significant" and "significantly" are overused throughout the manuscript (lines 181, 339, 463, 353), yet no quantitative data or statistical evidence (e.g., percentage) are provided to substantiate these claims. For example, when stating that a result is "significant," the authors should clarify *how significant* it is (e.g., "a 30% increase with 95% confidence") rather than relying on qualitative assertions. Note that the term "significant" carries strong inferential weight in scientific writing; its use in the main text should be reserved for findings supported by rigorous statistical analysis.

3 The assertion in line 194 that the primary sources of OC and ON "did not change across seasons" is questionable. For example, biomass burning and coal combustion are known to contribute seasonally variable OC/ON contributions. The authors should provide supplemental data or additional evidence (e.g., time-resolved source apportionment results) to support this claim in this study, particularly given evidence from prior studies demonstrating no seasonal shifts of ON and OC.

4 The uncertainty associated with the 18 identified factors requires further verification. (a) The correlation matrix between factors should be presented to evaluate their interdependence.

(b) The observation that no organic nitrogen (ON) was attributed to biomass burning or isoprene/ α -pinene SOA factors (lines 232-233) is unexpected and conflicts with extensive evidence. Biomass burning is a well-documented source of nitrogen-containing organic aerosols (OA) in both rural and urban environments (Li et al., 2023). Similarly, biogenic VOC (BVOC) oxidation under high-NO_x or NO₃ radical-dominated conditions has been shown to produce ON (Xu et al., 2014). The authors should reconcile these discrepancies by either (1) re-examining their receptor modeling framework, (2) acknowledging limitations in source apportionment resolution.

(c) The proposed formation pathway for ON from oxidized cooking emissions lacks mechanistic clarity and experimental support. The authors should either (1) provide chemical speciation data (e.g., HRMS)

linking cooking-derived organic compounds to ON precursors, (2) reference chamber studies demonstrating this pathway, or (3) propose a plausible reaction mechanism (e.g., radical-induced coupling of cooking-derived VOCs with NO_x). Without such evidence, this conclusion remains speculative.

5 The strong correlation between DAC_ON and NH₄⁺ (line 324) requires further scrutiny. While demonstrating agreement between these variables is important, the authors must also rule out potential correlations with other PMF factors (e.g., Nitrate_ON) to confirm the specificity of this relationship.

6 The statement in lines 393–396 that higher relative humidity (RH) promotes DCA_ON formation directly contradicts the evidence presented in Figure 3b, which shows that higher pH enhances DCA_ON formation. Higher RH usually leads to higher pH values.

7 Methodology sections switch between past and present tenses (e.g., "We have developed... All measurements were carried out..."). Standardize to past tense for consistency.

References:

Li, Y., Fu, T.-M., Yu, J. Z., Yu, X., Chen, Q., Miao, R., Zhou, Y., Zhang, A., Ye, J., Yang, X., Tao, S., Liu, H., and Yao, W.: Dissecting the contributions of organic nitrogen aerosols to global atmospheric nitrogen deposition and implications for ecosystems, *National Science Review*, 10, 10.1093/nsr/nwad244, 2023.

Xu, L., Guo, H., Boyd, C. M., Klein, M., Bougiatioti, A., Cerully, K. M., Hite, J. R., Isaacman-VanWertz, G., Kreisberg, N. M., Knote, C., Olson, K., Koss, A., Goldstein, A. H., Hering, S. V., de Gouw, J., Baumann, K., Lee, S.-H., Nenes, A., Weber, R. J., and Ng, N. L.: Effects of anthropogenic emissions on aerosol formation from isoprene and monoterpenes in the southeastern United States, *Proceedings of the National Academy of Sciences*, 112, 37-42, 10.1073/pnas.1417609112, 2014.