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# $NO_x$ reductions paradoxically hinder the decline of secondary organic aerosol in Eastern China via particulate organic nitrates chemistry

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**Dear Editor,**

Since 2013, the Chinese government has enacted robust measures to mitigate severe air pollution, achieving significant reductions in $PM_{2.5}$ levels by approximately 50%.[1] However, observational evidences reveal a critical divergence that secondary organic aerosol (SOA), a major $PM_{2.5}$ component, has declined at a significantly slower rate (-30%) than that of other $PM_{2.5}$ components.[2] This lagging decline has progressively increased SOA's contribution to total $PM_{2.5}$, establishing it as a critical bottleneck for further air quality improvement.[3]

Given the complex physicochemical nature of SOA, understanding the key components of SOA is essential to advancing $PM_{2.5}$ mitigation.[4] Particulate organic nitrates (pON) represent a significant fraction of SOA mass (10%–30%), formed primarily from biogenic VOCs reacting with $NO_3$ radicals at night or with OH under $NO_x$-rich conditions.[5] Recent studies underscore the persistent uncertainties regarding the chemical mechanisms of SOA oxidation by $NO_3$ radicals and its important role in aggravating urban air pollution and affecting radiation forcing across China.[6,7] However, long-term analyses differentiating SOA components by their distinct chemical mechanism remains scarce. Focusing on critical species like pON provides a more targeted foundation for optimizing control strategies.

Here, we analyzed observational data and conducted model simulations to investigate SOA trends in Eastern China during summer (June~August) from 2014 to 2019. Observations included daily $PM_{2.5}$ composition from Beijing and Shanghai, aggregated monthly SOA data from 20 sites across China, and gridded $PM_{2.5}$ component reanalysis products. The formation of key SOA components was simulated using the CMAQ model (v5.4) driven by meteorology from the WRF model, with anthropogenic emissions from the MEIC inventory, biogenic emissions from MEGAN, and biomass burning emissions from FINN. To attribute changes, we performed sensitivity simulations: one holding 2014 emissions constant to isolate meteorological effects, and another with progressive reductions (10%~50%) in $NO_x$ and VOC emissions from a 2019 baseline.

## TRENDS OF SOA

Since 2014, various data channels—including urban observations, $PM_{2.5}$ component reanalysis, and model simulations—indicate a rapid decline of secondary $PM_{2.5}$ (SPM) concentration in China at a rate of -2.6 $\mu g \cdot m^{-3} \cdot yr^{-1}$ (-10% $yr^{-1}$), driven by the swift response of inorganic components like $SO_4^{2-}$ to precursor reductions. As a consequence of energy structure transformation, the adoption of advanced emission control technologies, and stringent regulations, $SO_2$ and $NO_x$ emissions decreased by 61% and 17%, respectively, during this period. However, the decline rate of SOA concentrations is significantly lower than other SPM compositions. Observations in some Chinese cities show that SOA concentrations decreased slowly at an average rate of only −0.6 $\mu g \cdot m^{-3} \cdot yr^{-1}$, corresponding to approximately 60% of the SPM reduction rate (Figure 1A).

pON, a significant product of anthropogenic-biogenic interactions, constitute a substantial fraction (1.40~2.73 $\mu g \cdot m^{-3}$, 15%~28%) of SOA concentrations in the Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD) and South China (SC) regions, based on both model simulations and surface observations in China (Figure 1B). The model captured the observed pON quite well, with a correlation coefficient of 0.87 and a normalized mean bias of 13%. Among the specific SOA components, pON displayed a distinct upward trend across most of Eastern China, contrary to conventional expectations, and contributed +3% to +13% to SOA trends in the BTH, YRD and SC regions (Figure 1C). This phenomenon highlights the escalating significance of pON, a previously underappreciated component in regional SOA dynamics.

## DRIVING FACTORS FOR INTERANNUAL VARIABILITY OF PON

We further elucidate these phenomena through a chemical mechanistic perspective. Our study considered three distinct pathways for the formation of pON. The contribution of the monoterpenes (TERP) + $NO_3$ reaction channel is the highest, primarily depending on the fate of peroxyl radicals, which can react with $RO_2$, $HO_2$, $NO_3$, and NO producing volatile organic vapor-monoterpene nitrates, with the yields ranging from 42.2% to 100%. TERP can also react with OH in the presence of NO to produce volatile organic vapor, with a yield of 20.1%. In addition, isoprene can react with $NO_3$ and produce another volatile organic vapor-isoprene nitrates, with a relatively lower yield. Volatile organic vapors condense into semi-volatile substances after gas-particle partitioning, subsequently hydrolyzing at a fixed rate in the particulate phase for three hours to produce non-volatile substances.[8] We define the sum of semi-volatile substances and their hydrolysis products as pON. TERP and its hydrolysis products contribute nearly 99% of pON.

Based on the understanding of chemical mechanism, we investigated the relationship between pON and their major precursors $NO_3$ radical and TERP. Observations and simulations of the pON diurnal cycle pattern also demonstrate the dominant role of $NO_3$. pON trend is more strongly correlated with $NO_3$ radical variation in urban areas (Figures 1D-E). $NO_3$ is primarily a night time oxidant that is formed by the reaction of $NO_2$ and $O_3$, and exhibited a fluctuating upward trend across all three major regions, consistent with the observed $NO_3$ radical production rate ($PNO_3$) results.[9] This suggests that the reduction in anthropogenic emissions and adverse meteorological conditions appear to have increased the generation of $NO_3$ radicals, resulting in higher urban pON concentrations. This trend is even more pronounced at night (+0.04~+0.10 $\mu g \cdot m^{-3} \cdot yr^{-1}$).

Contrastingly, TERP is biogenic and its emissions are modulated by air temperature and may influence pON concentrations in specific years, as evidenced by the abnormally high values observed in the YRD region during the hot summer of 2017 (Figures 1D & F). However, no significant interannual trend has been observed in the BTH and YRD regions during 2014 and 2019 (Figure 1F).

This study employs the $NO_2/O_3$ ratio (calculated by MDA8 $O_3$ and daily average $NO_2$) as a key indicator for pON variation, due to its control over $NO_3$ radical formation and subsequent pON production. As shown in Figure 1G, despite significant baseline $NO_2$ differences, three distinct regions exhibit a

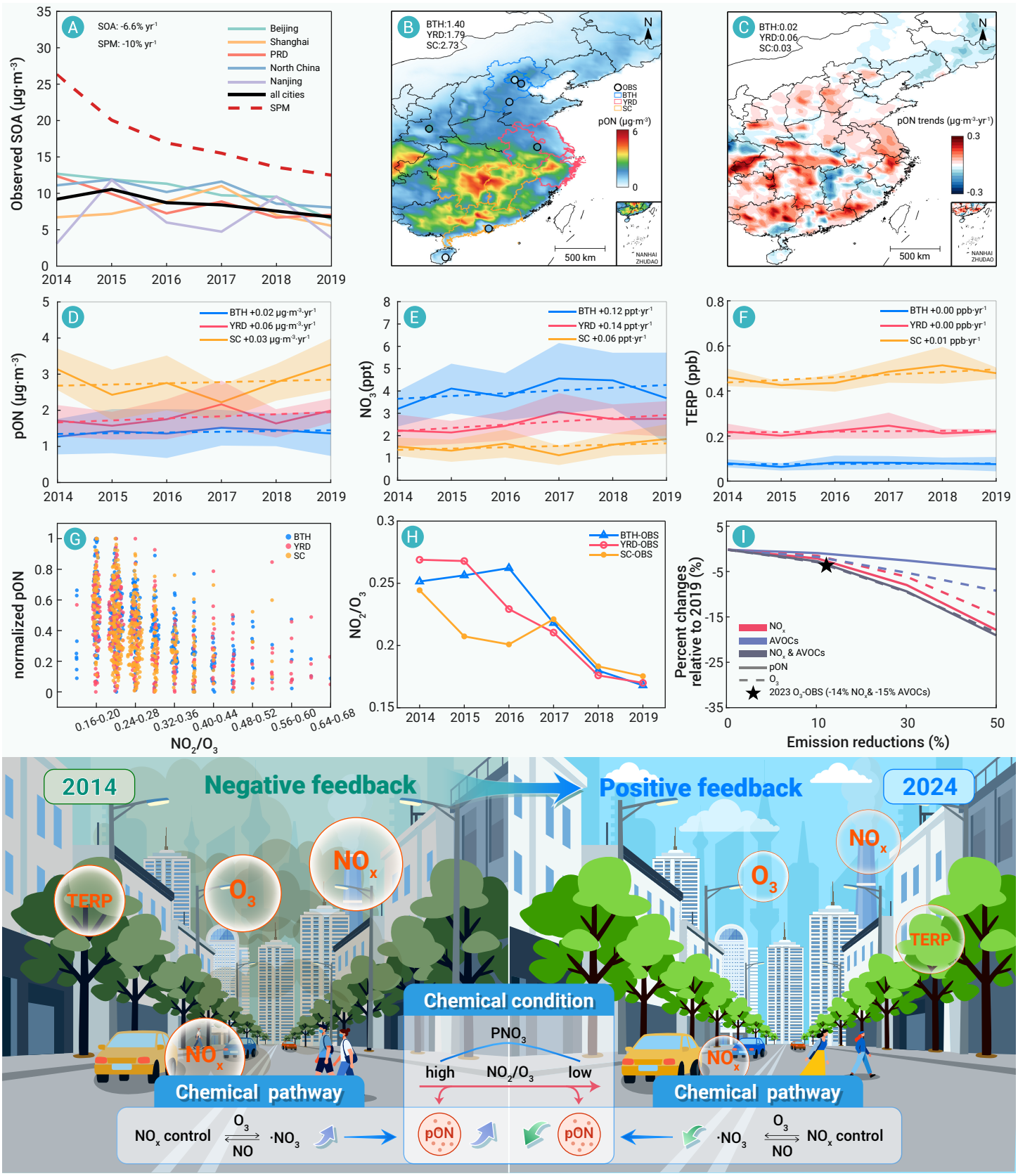


**Figure 1. Trends and driving factors of particulate organic nitrates (pON) in China** (A) Trends in observed SOA concentrations and secondary $PM_{2.5}$ (SPM) concentrations. (B) Observed and simulated surface pON concentrations. Observations shown as symbols. (C) Simulated pON trends. (D)-(F) Interannual variations in pON, $NO_3$, and TERP concentrations across the BTH, YRD and SC regions. The shaded areas denote standard deviations. (G) Relationships among normalized pON and the ratio of $NO_2/O_3$. (H) Interannual variations in the ratio of $NO_2/O_3$ from observation data. (I) Changes of pON and $O_3$ in response to emission reductions relative to 2019. Results are from CMAQ simulations for the July 2019 meteorological year, with 10%~50% individual emission reductions of $NO_x$, VOCs and combined emission reductions of $NO_x$ + VOCs. The distinct colors correspond to different emission reduction scenarios: red denotes $NO_x$ reduction, blue represents AVOCs reduction, and gray corresponds to the combined reduction of $NO_x$ and AVOCs. Solid lines indicate changes in pON, while dashed lines depict changes in $O_3$. Black stars mark the observed changes in $O_3$ in 2023 relative to 2019, and values in brackets indicate the relative changes in $NO_x$ and AVOCs emissions in summer 2023 compared to the same period in 2019.

consistent nonlinear relationship between pON and the $NO_2/O_3$ ratio. Specifically, when the $NO_2/O_3$ ratio exceeds approximately 0.20, pON increase as the ratio decreases, e.g. negative feedback regime, driven by weakened NO titration of $O_3$ under high emissions. Increased $O_3$ levels react with freshly emitted $NO_x$ during nighttime, resulting in the formation of numerous $NO_3$ radicals, which subsequently enhances the production of pON. When $NO_2/O_3$ drops below approximately 0.16, further reductions in the ratio suppress pON formation, e.g. positive feedback regime, resulting from diminished daytime $O_3$ in low-emission areas.

Observations reveal a significant decline in the $NO_2/O_3$ ratio from 0.26 (2014) to 0.16 (2019) in Eastern China, strongly correlated with nationwide $NO_2$ reductions (Figure 1H). This trend signifies China's transition from the negative feedback regime toward the positive feedback regime. Crucially, during 2017–2019, China traversed the peak-impact transition regime, where pON production maximized, exerting the strongest influence on SOA and $PM_{2.5}$. Continued $NO_x$-focused emission reductions post-2020 are projected to potentially decrease pON formation.

## COLLABORATIVE CONTROL OF SOA AND $O_3$

With sustained and substantial $NO_x$ reduction after 2020, observed $PNO_3$ level began to decline.[9] The continued decrease in both $NO_2$ and $PNO_3$ implies that pON formation entered a $NO_2$ positive feedback regime starting in 2020. Therefore, sustained $NO_2$ emission reductions are effective for controlling pON. Recent studies demonstrate a gradual transition in urban $O_3$ formation regime in China, from VOC-limited toward $NO_x$-limited regimes or joint-control regimes.[10] This shift suggests potential for synergistic mitigation of $O_3$ and pON through $NO_x$ reduction.

Further emission reduction experiments have also confirmed the potential for synergistic control of pON and $O_3$ pollution, providing a scientific basis for developing targeted mitigation strategies. Using July 2019 as the baseline period, we quantified changes in pON and $O_3$ concentrations resulting from 10%, 30%, and 50% reductions in $NO_x$ and AVOCs emissions. As shown in Figure 1I, pON and $O_3$ exhibit coherent variation across all scenarios. The simulated $O_3$ response to precursor reductions aligns closely with observed trends, as exemplified by measurements from the year 2023. $NO_x$ emission reductions effectively decrease pON concentrations; a 50% reduction in $NO_x$ emissions lead to an average 25% reduction in pON concentrations. The concurrent reduction of $NO_2$ and $O_3$ significantly suppressed the formation of pON. In contrast, AVOCs reductions exert negligible effects on pON pollution levels, primarily due to the reduced sensitivity of $O_3$ to VOCs in these regions, with the slight decrease in $O_3$ leading to only a marginal reduction in $NO_3$ concentrations. However, the benefits of emission reductions in the heavily polluted BTH region may be significantly lower than in other regions. Consequently, tailoring emission reduction policies to local conditions represents the optimal strategy. Considering the substantial costs associated with the synergistic control of $NO_x$ and VOCs, prioritizing $NO_x$ reductions represents a more pragmatic approach.

In recent years, observational and modeling evidences elucidate that the decline rate of SOA is significantly slower than that of other $PM_{2.5}$ components, leading to its increasing proportion in $PM_{2.5}$ and posing a critical bottleneck for further air quality improvement. This study advances the mechanistic understanding of SOA composition variability in Eastern China by quantifying the coupled roles of chemical pathways. Our results show that this anomaly is primarily attributable to pON, whose concentrations increased by 6%~21% across Eastern China, partially offsetting overall SOA reductions. We further demonstrate that urban $NO_x$ reductions unexpectedly elevated $NO_3$ radical concentrations, thereby enhancing pON formation. Our analysis reveals a dual-phase $NO_2$-driven feedback mechanism: prior to 2020, declining $NO_x$ weakened $O_3$ titration, elevating $NO_3$ radicals and promoting pON formation (negative feedback); after 2020, sustained $NO_x$ mitigation shifted this balance toward positive feedback and enabled coordinated SOA-$O_3$ control. These findings provide critical insights into managing key SOA components and regulating $PM_{2.5}$ levels in the future.

In summary, our study underscores the complex mechanisms governing changes in pON and its important role in the synergistic control of SOA and $O_3$ pollution. Future work should prioritize a deeper understanding of the precursors, oxidants, mechanisms and environmental impacts of key SOA components within complex atmospheric pollution systems.

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## AUTHOR CONTRIBUTIONS

F.C. and N.L. compiled the data and drafted the manuscript. N.L. and H.L. initiated the study and designed the research. All authors contributed to the manuscript and approved the final version.

## DECLARATION OF INTERESTS

The authors declare no competing interests.