Response to the reviewers:

We are deeply grateful to the reviewers for their thoughtful evaluations and valuable recommendations, which have substantially enhanced the quality of our work. Below, we present our point-by-point responses to each reviewer's comments, with the original feedback displayed in black and our replies in blue. For clarity, we have included the modified passages to demonstrate how each recommendation has been addressed and integrated into the revised manuscript.

Reviewer 2:

Comments

1. In Section 3.4.2 and Figure 11, the authors present mass-normalized OP across sites for each identified source and show that sources such as biomass burning, secondary aerosol, and dust have very different toxicities across locations. This comparison is potentially misleading because the factors come from separate PMF solutions; identically named factors (e.g., "secondary aerosol") may reflect different mixes of organics, inorganics, and metals at each site. When discussing the cross-site variability in toxicity, the manuscript lists possible explanations without evidence, making the discussion speculative rather than evidence-based. The authors should integrate differences in source profiles (Figure 10) with site characteristics to explain why similarly named aerosols exhibit different OP at different sites.

Response： **Thank you for this valuable comment.** We have revised Section 3.4.2 to provide more detailed, evidence-based explanations for the cross-site variability in source-specific OP by integrating differences in source profiles (Figure 10) with site characteristics. Specifically, we have enhanced our discussion for each source category:

Secondary aerosols: Zero OP at NN and ZZ sites due to different local VOC emission patterns and oxidation processes affecting chemical composition.

Dust: Zero contribution at ZZ attributed to inherent differences in local dust particle composition and trace metal content.

Agricultural activities: Zero OP at LFS likely due to NH_4^+ neutralization effects reducing redox activity at this rural site.

Chloride-rich combustion: OP contribution at GC (0.006 nmol H_2O_2·μg^{-1}) but not at LFS, likely due to GC's location in the Beijing-Tianjin-Hebei pollution transport corridor where industrial activities co-emit transition metals that catalyze ROS formation through Fenton reactions, while the remote LFS site lacks these catalytically active components.

We have specifically corrected in **Lines 595-603, Lines 604-617, Lines 634-641, Lines 642-650,** and **Lines 687-694**.

Lines 595-603: "Secondary aerosols

Secondary aerosols exhibited distinctly different patterns at different sites. At the LFS and GC sites, OP_m values of secondary aerosols are 0.005 and 0.014 H_2O_2·μg^{-1}, respectively, while the OP contributions of secondary aerosols were completely absent at NN and ZZ sites. PMF analysis revealed that OC at NN and ZZ sites was dominated by primary emissions with negligible SOA contributions, explaining their zero OP values. The photochemical oxidation of volatile organic compounds (VOCs) generates redox-active products (aldehydes, ketones, peroxides) that contribute to aerosol OP through ROS generation (Kong et al., 2023; Chen et al., 2022; Wei et al., 2022; Lin and Yu, 2011). The absence of secondary aerosol OP at NN and ZZ sites reflects their PMF-resolved source profiles: these sites were characterized by primary sources with low oxidation states, indicating limited photochemical processing."

Lines 604-617: "Dust

The minerals contained in dust can participate in the ROS generation process (Nishita-Hara et al., 2019; Lodovici and Bigagli, 2011). The OP_m values of dust at NN, LFS, and GC, are 0.004, 0.009, and 0.016 nmol H_2O_2·μg^{-1}, respectively. The dust source at the GC site exhibited the highest toxic efficiency, which may be attributed to its location in the highly industrialized Beijing-Tianjin-Hebei region, resulting in elevated TMs emissions (Li et al., 2022). Notably, the dust source at the ZZ site contributed zero to the OP, suggesting the possible presence of chemically inert mineral phases in this region. This absence of oxidative activity may be attributed to limited TMs content in the local soil and the unique chemical characteristics of dust particles. The oxidative activity of dust particles is primarily associated with their complex mineral composition and surface chemical properties. Specifically, iron-containing minerals can catalyze ROS formation through Fenton reactions, while clay minerals provide large specific surface areas that facilitate metal ion adsorption and subsequent redox reactions (Saffari et al., 2014; Guo et al., 2020; Liu et al., 2022). Due to differences in geological background, dust from different regions exhibits significant variations in mineral composition, leading to differences in their oxidative activity. Furthermore, quartz particles demonstrate certain oxidative activity through surface catalytic reactions, with surface silanol groups and defect sites promoting the generation of ·OH (Konecny et al., 2001)."

Lines 634-641: "Agricultural activity

The OP of agricultural activities was only detected at the GC site, with OP_m of 0.004 $H_2O_2 \cdot \mu g^{-1}$, while no OP contribution from agricultural activities was observed at the LFS site. PMF analysis showed contrasting patterns: agricultural factors at LFS were dominated by NH_4^+ while GC showed minimal NH_4^+ contributions. At LFS, the predominance of NH_4^+ in agricultural factors likely forms stable ammonium salts that neutralize particle acidity and reduce redox activity. In contrast, the lower NH_4^+ at GC may allow agricultural particles to maintain higher acidity and preserve the redox activity of trace metal components or organic matter (Tong et al., 2017; Wei et al., 2022). The presence of NH_4^+ can alter particle pH and ionic strength, affecting the solubility and reactivity of redox-active species (Zhang et al., 2021; Zhang et al., 2025)."

Lines 642-650: **"Chloride-rich combustion**

The OP of chloride-rich combustion was only detected at the GC site, with an OP_m of 0.006 nmol $H_2O_2 \cdot \mu g^{-1}$, while no OP was observed at the LFS site. This difference may be mainly attributed to the different pollution source characteristics and geographical locations of the two sites. The GC site is located in the Beijing-Tianjin-Hebei atmospheric pollution transport corridor and, as an industrial agglomeration area, the chlorine-rich combustion processes may be accompanied by co-emission or formation of more transition metals and other catalytically active components (Li et al., 2022). These metal ions can catalyze the generation of ROS through Fenton reactions and other pathways, thereby possibly exhibiting significant OP (Saffari et al., 2014; Guo et al., 2020). In contrast, LFS as a remote site may lack catalytically active metal components, and therefore showed no detectable OP."

Lines 687-694: "Other sources displayed notable regional variations. Secondary aerosols contributed zero OP at NN and ZZ sites, likely due to local VOC emission patterns and oxidation processes. Dust sources showed zero contribution at ZZ, potentially related to the inherent composition of local dust particles. Agricultural activities exhibited zero OP at LFS, possibly due to NH_4^+ neutralization effects that reduced redox activity. At GC, chloride-rich combustion demonstrated OP contribution (0.006 nmol $H_2O_2 \cdot \mu g^{-1}$), likely attributed to elevated TMs emissions in Beijing-Tianjin-Hebei region that promote ROS formation. Sea salt aerosols at NN showed an OP_m of 0.016 nmol $H_2O_2 \cdot \mu g^{-1}$, which was attributed to the catalytic effect of halogen compounds on ROS. In contrast, industrial emissions at ZZ exhibited zero OP due to minimal organic carbon content."

2. In addition, some statements appear arbitrary. For example: "Meteorological conditions also significantly influence the oxidative activity of dust (Joshi et al., 2017; Ma et al., 2023). The NN site is located in southern China, where higher precipitation levels compared to northern regions may contribute to reduced oxidative reactivity." The cited references are not relevant to this specific claim, and the linkage between precipitation and intrinsic dust toxicity is unsupported, thus failing to meet scientific standards.

Response: We thank for this important observation. We have removed these statements from the revised manuscript. **Lines 604-617** have been modified to:

"**Dust**

The minerals contained in dust can participate in the ROS generation process

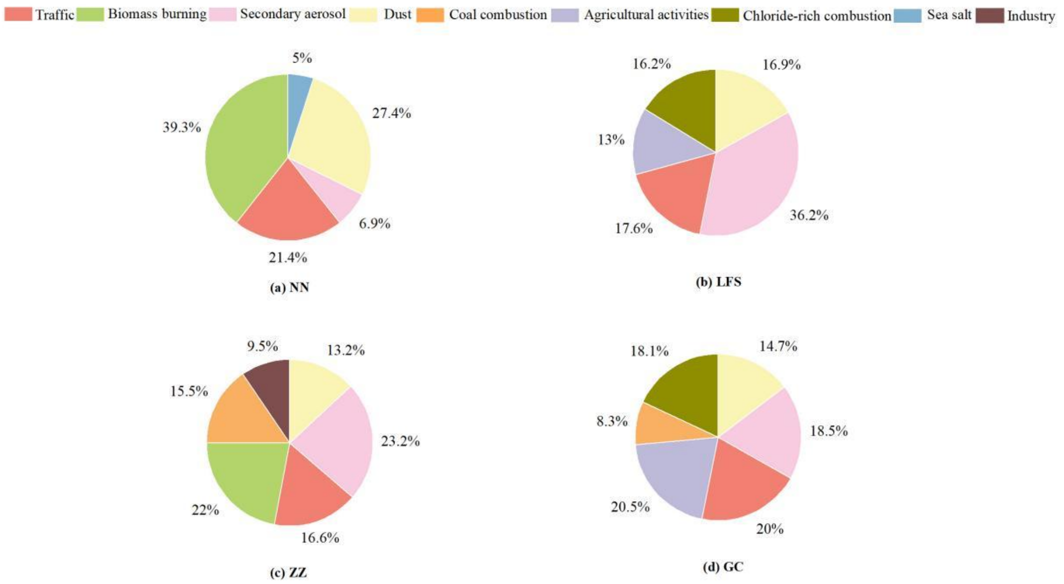
(Nishita-Hara et al., 2019; Lodovici and Bigagli, 2011). The OP_m values of dust at NN, LFS, and GC, are 0.004, 0.009, and 0.016 nmol $H_2O_2 \cdot \mu g^{-1}$, respectively. The dust source at the GC site exhibited the highest toxic efficiency, which may be attributed to its location in the highly industrialized Beijing-Tianjin-Hebei region, resulting in elevated TMs emissions (Li et al., 2022). Notably, the dust source at the ZZ site contributed zero to the OP, suggesting the possible presence of chemically inert mineral phases in this region. This absence of oxidative activity may be attributed to limited TMs content in the local soil and the unique chemical characteristics of dust particles. The oxidative activity of dust particles is primarily associated with their complex mineral composition and surface chemical properties. Specifically, iron-containing minerals can catalyze ROS formation through Fenton reactions, while clay minerals provide large specific surface areas that facilitate metal ion adsorption and subsequent redox reactions (Saffari et al., 2014; Guo et al., 2020; Liu et al., 2022). Due to differences in geological background, dust from different regions exhibits significant variations in mineral composition, leading to differences in their oxidative activity. Furthermore, quartz particles demonstrate certain oxidative activity through surface catalytic reactions, with surface silanol groups and defect sites promoting the generation of ·OH (Konecny et al., 2001)."

3. *Furthermore, the PMF factor identification (Figure 10) requires closer scrutiny. In Figure 10(b) LFS, the fourth factor is dominated by Cl^-; although K^+ is elevated, only ~20% of K^+ is apportioned to this factor, and the K^+ concentration is about an order of magnitude lower than Cl^-. This does not support a biomass burning source and is more consistent with chloride-rich combustion (e.g., waste burning). Nevertheless, it is labeled as biomass burning. A similar issue appears for the biomass burning factor at (d) GC, whose profile differs markedly from that at (a) NN. In addition, while (b) "agriculture" shows high NH_4^+ as expected, the "agricultural activities" factor in (c) ZZ has minimal NH_4^+ and low NO_3^- but high K^+, EC, and OC, indicating a contribution from biomass burning rather than fertilizer-related ammonium. Factor identification should therefore be revisited. The apparent cross-site differences in*

toxicity in Figure 11 may partly reflect mislabeling of sources. I recommend that the PMF analysis, factor identification and toxicity interpretation be strengthened before the manuscript can be considered for publication.

Response: We sincerely thank the reviewer for these critical and constructive comments regarding PMF factor identification. Through careful review, we have revised the factor identifications as follows: the biomass burning factors at LFS and GC sites have been relabeled as chloride-rich combustion, and the agricultural activities factor at ZZ has been relabeled as biomass burning. These modifications have been made in **Lines 464-475, Lines 506-509, Lines 527-530, Lines 544-546, Lines 558-568, Figure 11, Lines 584-594, Lines 634-650, and Lines 678-694** of the revised manuscript.

Lines 464-475: "Sources of PM_{10} in LFS may include secondary aerosols, traffic, dust, chloride-rich combustion, and agricultural activities. The ZZ site showed secondary aerosols, biomass burning, traffic, coal combustion, dust, and industry as the main sources. PM_{10} sources in GC are agricultural activities, traffic, biomass burning, secondary aerosols, chloride-rich combustion, dust, and coal combustion.



(a) NN

(b) LFS

(c) ZZ

(d) GC

Figure 9. The contributions of Traffic, Biomass burning, Secondary aerosol, Dust, Coal combustion, Agricultural activities, Chloride-rich combustion, Sea salt, and Industry to the atmospheric concentration of PM_{10} mass (%) as derived by PMF modelling at NN, LFS, ZZ, and GC.

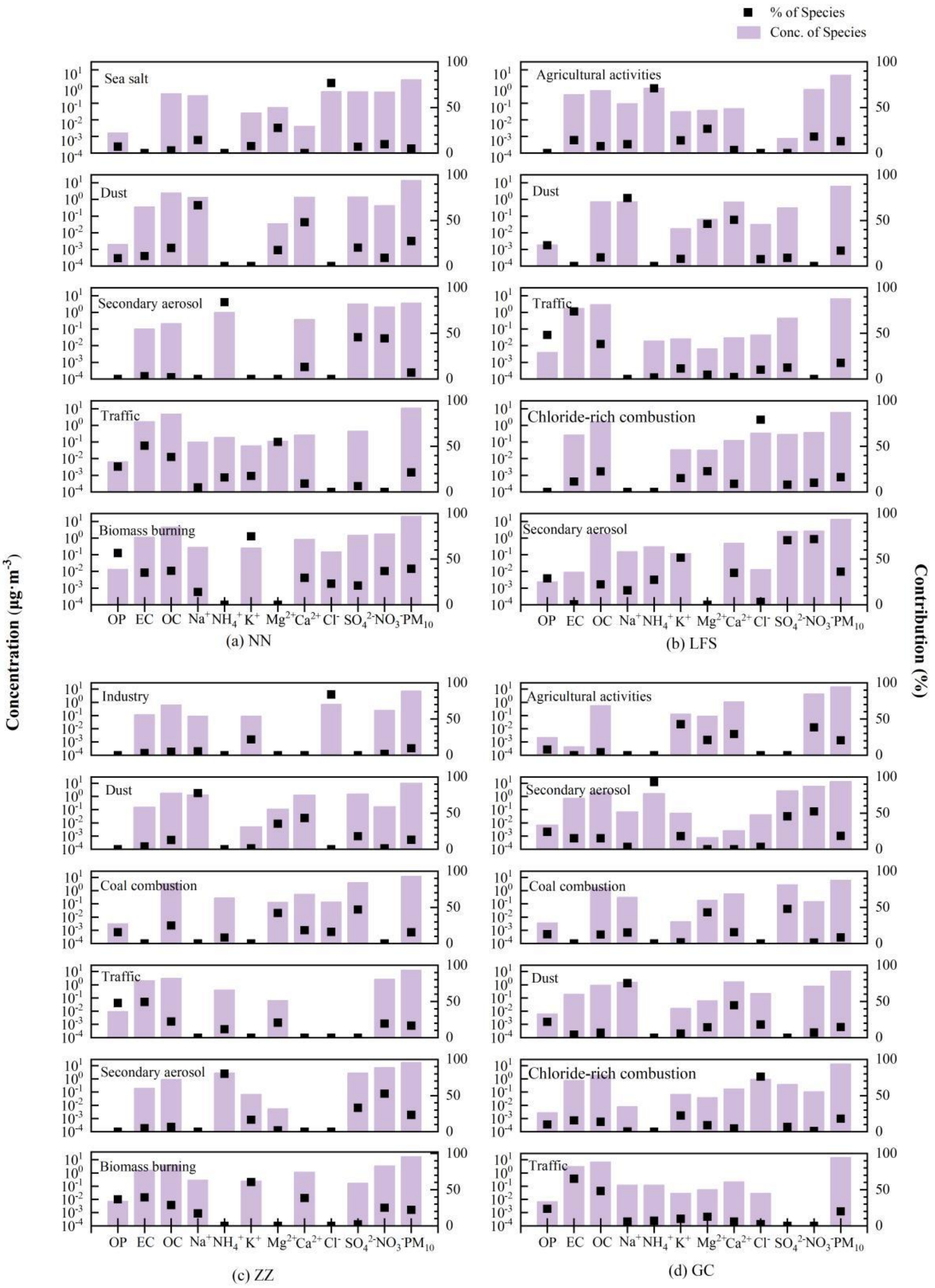


Figure 10. Chemical profiles of the source factors identified at NN, LFS, ZZ and GC. The bars represent the chemical composition profiles (left y-axis) and the dots indicate the contribution values (right y-axis).”

Lines 506-509: “The fourth factor is dominated by Cl^- (79.2%) with concentrations approximately one order of magnitude higher than K^+ (22.7%), indicating a chloride-rich combustion source. This pattern suggests combustion of chloride-containing materials or waste burning, which can produce elevated chloride emissions."

Lines 527-530: "The sixth factor had high levels of K^+ (60.6%), EC (39.3%), Ca^{2+} (38.4%), and OC (28.8%), contributing approximately 22% to PM_{10}. K^+, EC, and OC are tracers for biomass burning emissions (Stracquadanio et al., 2019). Given ZZ's location in a suburban of Zhengzhou, this factor likely represents crop residue burning and residential biomass combustion common in central China's agricultural regions."

Lines 544-546: "The fourth factor characterized by Cl^- (76.2%) represents chloride-rich combustion, accounting for 18.1%. Similar to LFS, this factor is dominated by Cl^- with concentrations, indicating a specific chloride-rich combustion source."

Lines 558-568: "A notable pattern observed among the two sites with agricultural activities (LFS and GC) is the differential contribution of NH_4^+ within agricultural emission factors, with NH_4^+ being exclusively associated with agricultural activities at the LFS site. This spatial variation reflects the complex interplay between regional meteorological conditions, agricultural practices, and atmospheric chemistry processes. At the LFS site in northeastern China, cooler climate conditions favor the stability of particulate NH_4^+, allowing its direct retention within agricultural emission factors (Wang et al., 2020). The concentrated fertilizer application during the spring planting season, combined with lower ambient temperatures that minimize NH_4^+ volatilization, preserves the distinct agricultural source signature at this remote location (Huo et al., 2025). Conversely, at the warmer GC site in central and northern China, NH_4^+ undergoes more extensive atmospheric processing due to higher ambient temperatures. These conditions promote the volatilization of NH_4^+ to gaseous NH_3, which subsequently undergoes secondary reactions with acidic species (SO_4^{2-} and NO_3^-) to form ammonium-containing secondary aerosols (Stelson and Seinfeld, 1982; Wang et al., 2015)."

Figure 11:

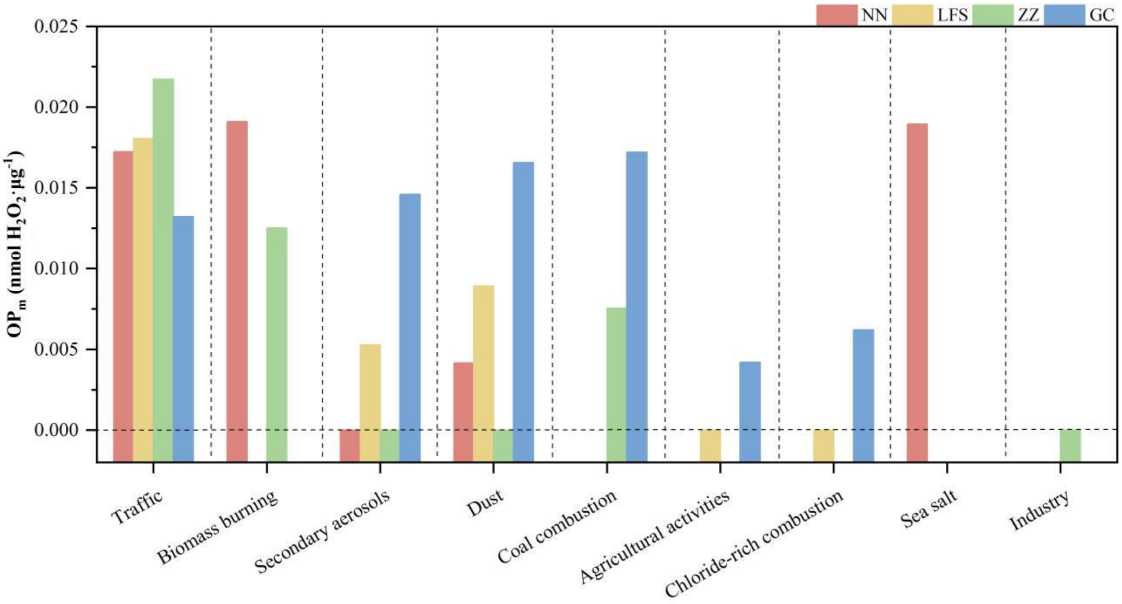


Figure 11. Comparison of OP_m contributions from different emission sources across NN, LFS, ZZ and GC sites.

Lines 584-594: "Biomass burning

Biomass burning sources were highly detected to contribute to OP at NN and ZZ sites, with OP_m of 0.019 and 0.012 nmol H_2O_2·μg^{-1}, respectively. This is closely related to the frequent crop straw burning activities in the region. Particulate matter emitted from biomass burning has complex chemical compositions, and its OP primarily originates from various organic compounds produced during the combustion process. Biomass burning produces substantial amounts of PAHs, which can form quinone intermediates, subsequently participating in ROS generation processes (Zhu et al., 2024; Libalova et al., 2018). The biomass burning process also releases water-soluble organic carbon (WSOC), which contains humic-like substances (HULIS) with significant oxidative activity (Yan et al., 2015; Salma et al., 2010). These macromolecular organic compounds contain abundant functional groups such as hydroxyl, carbonyl, and carboxyl groups, which can generate reactive oxygen species like hydroxyl radicals and hydrogen peroxide through photochemical reactions and metal-catalyzed reactions (Verma et al., 2015; Lin and Yu, 2011)."

Lines 634-650: "**Agricultural activity**

The OP of agricultural activities was only detected at the GC site, with OP_m of 0.004 nmol $H_2O_2 \cdot \mu g^{-1}$, while no OP contribution from agricultural activities was observed at the LFS site. PMF analysis showed contrasting patterns: agricultural factors at LFS were dominated by NH_4^+ while GC showed minimal NH_4^+ contributions. At LFS, the predominance of NH_4^+ in agricultural factors likely forms stable ammonium salts that neutralize particle acidity and reduce redox activity. In contrast, the lower NH_4^+ at GC may allow agricultural particles to maintain higher acidity and preserve the redox activity of trace metal components or organic matter (Tong et al., 2017; Wei et al., 2022). The presence of NH_4^+ can alter particle pH and ionic strength, affecting the solubility and reactivity of redox-active species (Zhang et al., 2021; Zhang et al., 2025).

Chloride-rich combustion

The OP of chloride-rich combustion was only detected at the GC site, with an OP_m of 0.006 nmol $H_2O_2 \cdot \mu g^{-1}$, while no OP was observed at the LFS site. This difference may be mainly attributed to the different pollution source characteristics and geographical locations of the two sites. The GC site is located in the Beijing-Tianjin-Hebei atmospheric pollution transport corridor and, as an industrial agglomeration area, the chlorine-rich combustion processes may be accompanied by co-emission or formation of more transition metals and other catalytically active components (Li et al., 2022). These metal ions can catalyze the generation of ROS through Fenton reactions and other pathways, thereby possibly exhibiting significant OP (Saffari et al., 2014; Guo et al., 2020). In contrast, LFS as a remote site may lack catalytically active metal components, and therefore showed no detectable OP.”

Lines 678-694: “3. PM_{10} Source Apportionment

Source apportionment using PMF indicated that dust (13.2-27.4%), secondary aerosols (6.9-36.2%), traffic (16.6-21.4%), and biomass burning (22-39.3%) were likely the main contributing sources to PM_{10} mass concentrations at the study sites. Understanding the contributions of these sources is crucial for developing more effective PM_{10} reduction strategies.

4. OP Source Analysis

OP_m analysis revealed significant spatial variations in toxic efficiency across different sources. Traffic sources demonstrated consistently high OP_m values (0.013-0.022 nmol $H_2O_2 \cdot \mu g^{-1}$) across all four sampling sites, while biomass burning exhibited elevated OP_m at NN and ZZ sites (0.012 and 0.019 nmol nmol $H_2O_2 \cdot \mu g^{-1}$, respectively). Coal combustion also showed high OP_m values at both ZZ and GC sites (0.008 and 0.017 nmol nmol $H_2O_2 \cdot \mu g^{-1}$, respectively), indicating the substantial toxic efficiency of particulate matter from these three major sources. Other sources displayed notable regional variations. Secondary aerosols contributed zero OP at NN and ZZ sites, likely due to local VOC emission patterns and oxidation processes. Dust sources showed zero contribution at ZZ, potentially related to the inherent composition of local dust particles. Agricultural activities exhibited zero OP at LFS, possibly due to NH_4^+ neutralization effects that reduced redox activity. At GC, chloride-rich combustion demonstrated OP contribution (0.006 nmol $H_2O_2 \cdot \mu g^{-1}$), likely attributed to elevated TMs emissions in Beijing-Tianjin-Hebei region that promote ROS formation. Sea salt aerosols at NN showed an OP_m of 0.016 nmol $H_2O_2 \cdot \mu g^{-1}$, which was attributed to the catalytic effect of halogen compounds on ROS. In contrast, industrial emissions at ZZ exhibited zero OP due to minimal organic carbon content.”