

Article

Incorporating ^{15}N into the Multi-Resolution Emission Inventory to Simulate the Spatiotemporal Variations of $\delta^{15}\text{N}$ in Emitted NO_x over the Pearl River Delta Region, China

Fan Wang ¹ , Yiming Liu ² , Greg Michalski ³, Wendell Walters ⁴ and Huan Fang ^{4,*} 

¹ School of Ecology, Sun Yat-sen University, Shenzhen 518107, China; wangfan25@mail.sysu.edu.cn

² School of Atmospheric Sciences, Sun Yat-sen University, Zhuhai 519082, China; liuym88@mail.sysu.edu.cn

³ Department of Earth, Atmospheric, and Planetary Sciences, Purdue University, 550 Stadium Mall Drive, West Lafayette, IN 47907, USA; gmichals@purdue.edu

⁴ Department of Chemistry and Biochemistry, University of South Carolina, Columbia, SC 29208, USA; wendellw@mailbox.sc.edu

* Correspondence: fangh@mailbox.sc.edu

Abstract

Nitrogen oxides (NO_x), comprising nitric oxide (NO) and nitrogen dioxide (NO_2), are key precursors of atmospheric nitrate, a major component of fine particulate matter ($\text{PM}_{2.5}$) that critically affects air quality, human health, and ecosystems. Emission inventories provide detailed spatial and temporal information on NO_x sources, while stable isotope techniques offer an additional constraint for source apportionment. Here, we incorporated stable nitrogen isotopes (^{14}N , ^{15}N) into the widely used Multi-resolution Emission Inventory for China (MEIC) over South China, with a focus on the Pearl River Delta (PRD) region, one of the most highly urbanized and industrialized regions in China, using an isotopic mass–balance model. The 2008 MEIC inventory indicated that NO_x emissions across South China were spatially heterogeneous, dominated by transportation sources, and concentrated mainly in the PRD and other urban clusters. We then compared the simulated isotopic composition of emitted NO_x with atmospheric measurements to assess the role of emission sources in controlling atmospheric nitrate (NO_3^-). The simulated $\delta^{15}\text{N}(\text{NO}_x)$ values were found to generally underestimate the observed $\delta^{15}\text{N}(\text{NO}_3^-)$ values. This discrepancy highlights the need for future ^{15}N -enabled air quality modeling to better represent both source contributions and atmospheric processing, thereby improving source apportionment, emission inventory evaluation, and our understanding of reactive nitrogen cycling.

Keywords: NO_x emission; stable nitrogen isotope; MEIC; the Pearl River Delta



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1. Introduction

Nitrogen oxides (NO_x), comprising nitric oxide (NO) and nitrogen dioxide (NO_2), can be oxidized in the atmosphere to form atmospheric nitrate (NO_3^-), a major component of fine particulate matter ($\text{PM}_{2.5}$) that critically affects air quality and human health [1]. The subsequent deposition of NO_3^- and other reactive nitrogen (N) species contributes to acid rain, soil acidification, aquatic eutrophication, and biodiversity loss, thereby threatening ecosystem functions and environmental sustainability [2–4]. The primary anthropogenic sources of NO_x emissions include power plants, transportation, industry, and residential combustion [5,6].

Coastal bay areas are strongly influenced by human activities because they represent zones of intense interaction among marine, terrestrial, and atmospheric systems [7]. The

Pearl River Delta (PRD) region, where the Pearl River discharges into the South China Sea, is one of the most highly urbanized and industrialized regions globally. Rapid urbanization in the PRD region has been accompanied by large emissions of NO_x , making it a hotspot of inorganic N deposition in China [8–12]. Accurate characterization of NO_x emission sources in the PRD region is therefore crucial for developing effective emission control strategies.

Emission inventories provide detailed spatial and temporal information on source distributions and serve as fundamental inputs for air quality modeling, source attribution, and pollution control strategy design. Over the past decade, continuous improvements in emission inventory development have greatly enhanced the characterization of regional air pollutant emissions, yet substantial uncertainties remain for individual source sectors and regions [13–15]. Meanwhile, stable isotopes have emerged as another promising tool in atmospheric pollution research, especially for source apportionment [16–19]. Different NO_x sources tend to exhibit distinct $\delta^{15}\text{N}$ signatures, expressed as the per mil deviation of the $^{15}\text{N}/^{14}\text{N}$ ratio in NO_x from that of atmospheric N_2 . For example, coal-fired power plants generally emit ^{15}N -enriched NO_x , with $\delta^{15}\text{N}$ values ranging from +2.1‰ to +25.6‰; industrial sources show $\delta^{15}\text{N}$ values between −10.9‰ and +8.1‰; whereas transportation (e.g., on-road gasoline/diesel vehicles) and residential sources (e.g., natural gas furnace, off-road gasoline/diesel equipment) show lower or more variable $\delta^{15}\text{N}$ values, ranging from −28.1‰ to +17‰ and from −21.1‰ to +8.5‰, respectively [20–26]. These differences suggest that $\delta^{15}\text{N}$ can serve as a useful tracer for distinguishing NO_x sources. Nevertheless, what remains lacking is the incorporation of $\delta^{15}\text{N}$ fingerprints into emission inventories to enable large-scale, source-resolved air quality modeling.

To address this gap, this study incorporated ^{15}N into the Multi-resolution Emission Inventory for China (MEIC) across South China, including the PRD region, and simulated the spatiotemporal variations in $\delta^{15}\text{N}$ of emitted NO_x using an isotopic mass-balance framework based on source mixing. The simulated $\delta^{15}\text{N}(\text{NO}_x)$ values captured the inherent emission-derived isotopic variability and were compared with available atmospheric $\delta^{15}\text{N}(\text{NO}_3^-)$ measurements to evaluate the role of emission sources in controlling atmospheric NO_3^- in the PRD region. The resulting ^{15}N -enabled emission dataset will provide input for future air quality modeling to better resolve N emission, transport, and transformation.

2. Methodology

2.1. The Spatial Domains

Two spatial domains were employed: an outer domain covering South China and an inner domain focusing on the Pearl River Delta (Figure 1). The outer domain spans 18–30° N and 106–120° E, encompassing the entirety of Guangdong Province, Guangxi Zhuang Autonomous Region, Hainan Province, Jiangxi Province, and Hunan Province, and part of Guizhou Province. The inner domain, representing the Pearl River Delta, includes the cities of Guangzhou, Shenzhen, Zhuhai, Foshan, Dongguan, Zhongshan, Jiangmen, Huizhou, Zhaoqing, Hong Kong and Macau (Figure 1; light purple). Both domains were simulated at a horizontal resolution of 36 km × 36 km.

2.2. MEIC Inventory

The Multi-resolution Emission Inventory for China (MEIC), developed by Tsinghua University, is a bottom-up anthropogenic emission inventory for mainland China that includes major source sectors such as power, industry, residential, transportation, solvent use, and agriculture (<http://meicmodel.org.cn>). It provides gridded emissions for multiple pollutants, such as SO_2 , NO_x , CO, NMVOCs, NH_3 , PM_{10} , $\text{PM}_{2.5}$, BC, and OC, and has been widely used in air quality modeling and emission assessment studies in China and abroad.

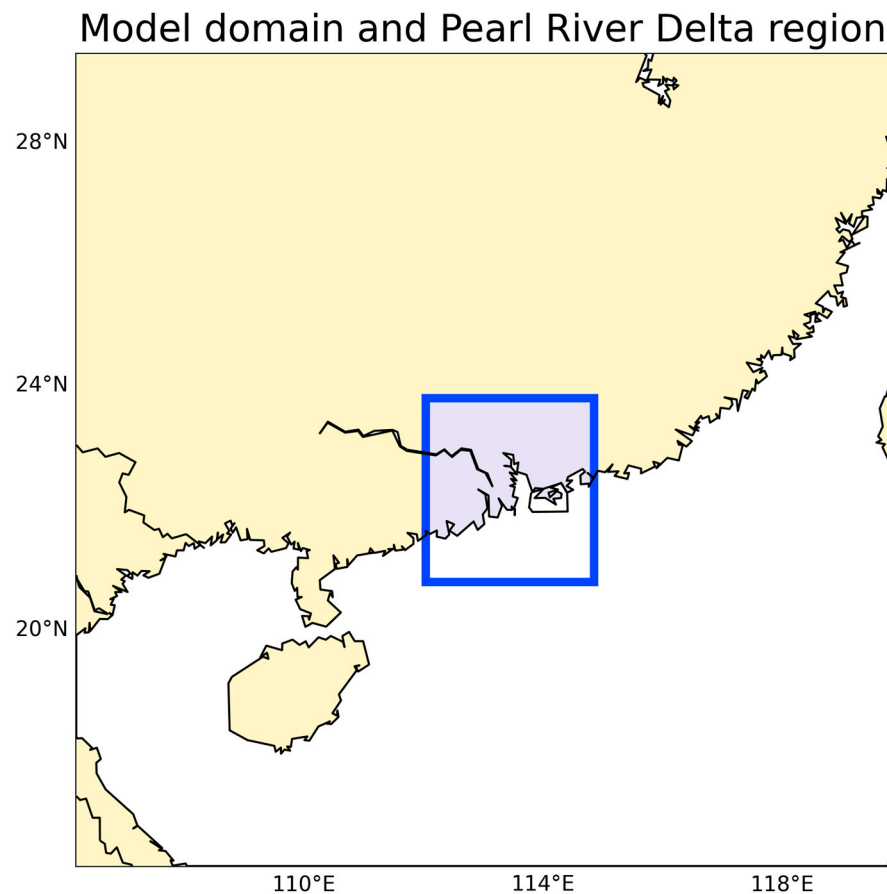


Figure 1. The outer modeling domain over South China and the inner domain focusing on the Pearl River Delta region, indicated by the blue box.

In this study, the 2008 MEIC inventory was adopted as the input emission dataset, consistent with the period of historical $\text{NO}_3^- \delta^{15}\text{N}$ observations available for the PRD region. Within the MEIC framework [13,15], NO_x emissions from the power sector, dominated by coal-fired power plants, were estimated at the level of individual units or plants using detailed information on activity levels, fuel consumption, boiler size, combustion technology, coal type, and the penetration of end-of-pipe control measures. Industrial NO_x emissions were estimated using activity-based approaches linked to industrial fuel consumption, product output, production technology, and control measures, with large point sources such as cement plants and iron and steel plants increasingly represented using factory- or plant-level information. Transportation NO_x emissions primarily represented mobile sources, including on-road vehicles, shipping, and other off-road engines (e.g., agricultural equipment, industrial equipment, locomotives, and commercial aircraft). Residential NO_x emissions were derived from bottom-up estimates that link household energy consumption for cooking and heating to source-specific emission factors associated with the combustion of fossil fuels and biofuels. Agricultural NO_x emissions were estimated using a bottom-up approach based on multi-source activity data, including fertilizer application rates, livestock numbers, and crop residue burning, together with localized emission factors. However, agricultural NO_x emissions were found to be relatively minor in the study domain and therefore not considered further in this study. Solvent use was not treated as a separate sector in this study because it is primarily associated with emissions of non-methane volatile organic compounds rather than NO_x .

The 2008 MEIC inventory was subsequently processed to generate hourly NO_x emissions over South China. Similar to the work of Wang et al. [27], we allocated monthly

sectoral emissions to hourly emissions using sector-specific weekly and hourly temporal profiles, with concurrent species splitting and unit conversion. For spatial allocation, we applied different approaches by sector to allocate emissions from coarse grids to fine grids. Power plant, industrial and residential emissions were allocated using source- or activity-related spatial proxies. Transportation emissions were spatially allocated using line-based surrogates (e.g., roads and flight paths).

2.3. Compilation of $\delta^{15}\text{N}$ Values of NO_x

Based on an extensive review of relevant literature [28], the source-specific $\delta^{15}\text{N}(\text{NO}_x)$ values were compiled and summarized in Table 1. The adopted $\delta^{15}\text{N}$ values (Table 1) were selected through literature synthesis rather than simply taking the midpoint of each reported range, as in our previous study [28]. This selection considered the most frequently reported values, source characteristics, and the relative consistency of observations within each emission sector. Given the relatively wide source-specific $\delta^{15}\text{N}$ ranges reported in the literature, these adopted values should be interpreted as simplified sector-level estimates for regional-scale simulations rather than precise source-specific endmembers. Future work should incorporate sensitivity analyses and region-specific endmember constraints to better quantify and reduce this uncertainty associated with endmember selection.

Table 1. $\delta^{15}\text{N}(\text{NO}_x)$ values (‰) by MEIC emission sector.

MEIC Sector	$\delta^{15}\text{N}$ Range (‰)	Adopted $\delta^{15}\text{N}$ (‰)	Main References
Power plant (coal-fired)	+2.1~+25.6	+15	[20]
Industry	−10.9~+8.1	−0.75	[24,26]
Transportation	−28.1~+17	−2.6	[23]
Residential sources	−21.1~+8.5	−11	[24,26]

2.4. Incorporating ^{15}N into NO_x Emission Datasets

The N isotope ratio $\delta^{15}\text{N}$ is defined as:

$$\delta^{15}\text{N}(\text{NO}_x) = \left[\frac{(^{15}\text{NO}_x / ^{14}\text{NO}_x)}{(^{15}\text{N} / ^{14}\text{N})_{\text{air}}} - 1 \right] \times 1000, \quad (1)$$

where 0.0036 is the natural $^{15}\text{N}/^{14}\text{N}$ ratio in air N_2 (thus $\delta^{15}\text{N}(\text{N}_2) = 0$), which is the reference point for $\delta^{15}\text{N}$ measurements.

Within each grid cell, by combining the known $^{14}\text{NO}_x$ emission flux, treated as the NO_x flux, from each source i with its corresponding $\delta^{15}\text{N}(\text{NO}_x, i)$ value, $\delta^{15}\text{N}(\text{NO}_x)$ can be calculated based on source mixing using an isotopic mass-balance framework:

$$\delta^{15}\text{N}(\text{NO}_x) = \sum [f(i) \times \delta^{15}\text{N}(\text{NO}_x, i)] \quad (2)$$

where $f(i)$ is the fraction of $^{14}\text{NO}_x$ emissions contributed by source i , derived from the emission dataset as:

$$f(i) = \frac{^{14}\text{NO}_x(i)}{\sum ^{14}\text{NO}_x(i)} \quad (3)$$

3. Results and Discussion

3.1. Spatiotemporal Variability of NO_x Emission Rates and Source Fractions

Figure 2 shows that NO_x emissions in 2008 displayed pronounced and seasonally persistent spatial heterogeneity across South China (including the PRD region). Total NO_x emissions varied substantially, ranging from near zero in offshore and remote inland areas to more than 30 t N d^{-1} in the strongest hotspot grids (Figure 2). However, most

of the mapped domain was characterized by relatively low emissions ($<5 \text{ t N d}^{-1}$), while only a small fraction of grids had high emissions ($>20 \text{ t N d}^{-1}$). These high-emission grids were concentrated mainly in the PRD region and other urban clusters around the cities of Xiamen, Changsha, Nanchang, Guiyang, and Chongqing. By contrast, the low-emission grids were located mainly in offshore areas and in many inland rural regions. Moderate-emission areas were mainly distributed in the peripheral zones surrounding the major urban clusters, resulting in distinct spatial gradients from the cluster cores to the adjacent areas. These spatial distributions and approximate orders of magnitudes of NO_x emissions are broadly consistent with satellite-derived NO_2 vertical column densities in 2018 [29], after accounting for NO_x lifetime, NO-NO_2 partitioning, transport, chemical transformation, dilution, and deposition.

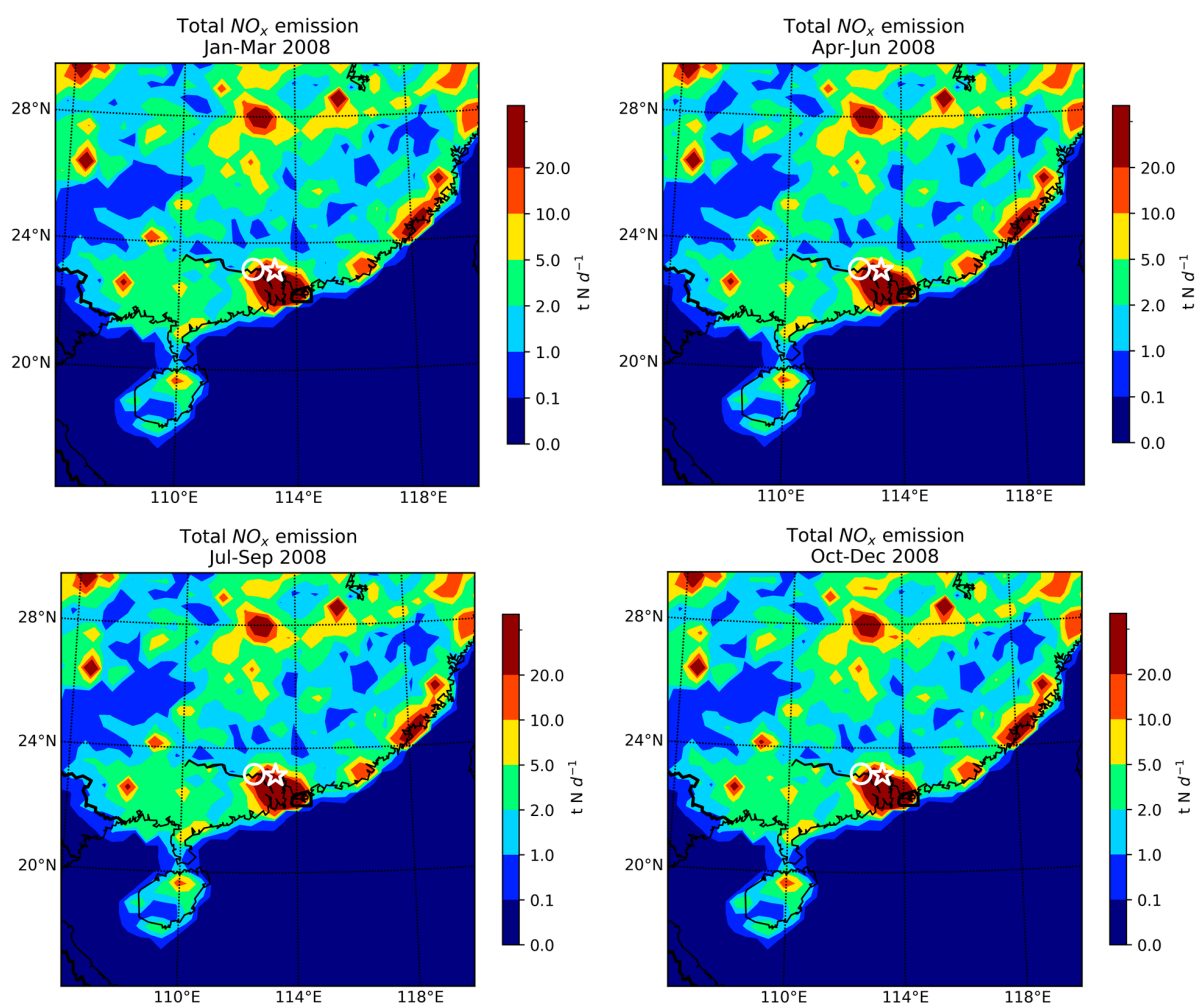


Figure 2. Variability in total NO_x emissions across South China, including the PRD region, during four quarterly periods in 2008. The star indicates the urban Guangzhou site, and the circle indicates the rural Dinghushan site.

Figure 2 further shows modest seasonal changes in emission intensity, but the hotspot pattern remained largely unchanged throughout 2008. Among the four seasons, the overall distribution in October–December appeared slightly stronger in several hotspot regions, whereas the distribution during July–September was comparatively weaker. However, these seasonal differences were much smaller than the persistent spatial contrast between coastal urban hotspots and low-emission offshore or rural areas. This was due to the fact that the spatial distribution of NO_x emissions across South China (including the PRD

region) was largely controlled by the locations of major anthropogenic sources, which change little seasonally [30,31].

Figure 3 indicates that power plant sources contributed a limited fraction of the total NO_x emissions over South China, although their contribution was locally very high in a small number of grids. High grid-level fractions of power plants (f_{pow}) were confined mainly to grids containing major power plants, often in proximity to metropolitan regions, consistent with the typical siting of large thermal power plants in South China [32]. At the regional scale, f_{tra} in South China appeared to be lower than the fraction of 0.343 for annual anthropogenic NO_x emissions from power plant/point sources over the continental United States [14].

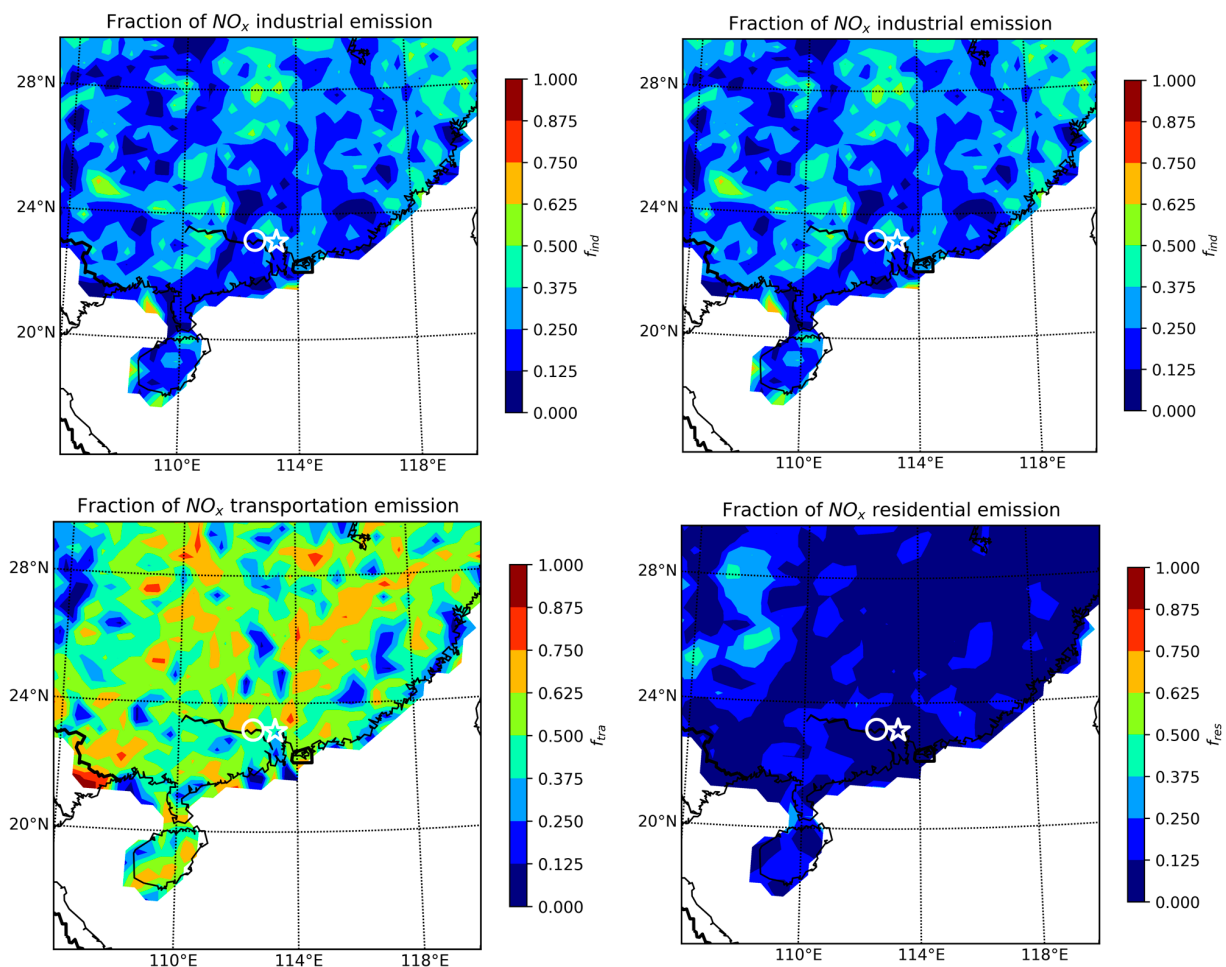


Figure 3. Spatial distribution of the fractional contributions of four major source sectors (power plants, industry, transportation, and residential sources) to grid-level NO_x emissions across South China in 2008. The star and circle indicate the Guangzhou and Dinghushan sites, respectively.

Industrial sources made a substantial contribution to NO_x emissions across South China and were more spatially widespread than power plant sources. The grid-level fraction of industrial NO_x emissions (f_{ind}) was generally moderate over large parts of both inland and coastal areas, with elevated f_{ind} values mainly occurred in industrial belts and peri-urban manufacturing areas rather than in urban cores dominated by mixed sources. This spatial pattern is consistent with previous reports [33–35].

Transportation sources were the most important contributors to NO_x emissions across South China and displayed the most spatially extensive influence among the four major source sectors. High grid-level transportation fractions (f_{tra}) were observed mainly in parts of the coastal zone, including the PRD region and the eastern Guangdong coast, as well

as in portions of the Beibu Gulf region, Hainan Island, and several inland areas. In the PRD region and other major urban clusters, high f_{tra} likely reflected dense road networks, port-related activities and intense traffic flows [36,37]. In non-urban core regions, however, elevated f_{tra} may reflect the weaker influence of other NO_x sources, making transportation relatively more important in the local emission mix.

Residential sources contributed only a minor fraction of NO_x emissions over most of South China, with only a few scattered areas showing moderate contributions. Relatively higher grid-level residential fractions (f_{res}) were observed mainly in a few inland areas where strong competing emissions from transportation and large industrial or point sources were lacking. Even in these areas, however, residential emissions generally did not emerge as the primary NO_x source at the grid scale.

Taken together, transportation emissions were the most spatially pervasive and dominated much of the domain; industrial emissions were also regionally important and spatially widespread, but generally less significant than that of transportation; power plant emissions were highly localized and dominant only in a limited number of grids; residential emissions contributed only a minor fraction over most of the domain and played a comparatively limited role in shaping the regional NO_x distribution.

3.2. Simulated Spatiotemporal Variability of $\delta^{15}\text{N}(\text{NO}_x)$ Values

Using the gridded NO_x emission source fractions of four major sectors, the $\delta^{15}\text{N}$ values of NO_x were simulated across South China in 2008. The predicted $\delta^{15}\text{N}(\text{NO}_x)$ ranged from approximately -8‰ to $+2\text{‰}$ and displayed pronounced spatial heterogeneity (Figure 4). Despite some seasonal variability in magnitude, the overall spatial pattern of $\delta^{15}\text{N}(\text{NO}_x)$ remained broadly consistent among the four seasons.

Widespread $\delta^{15}\text{N}(\text{NO}_x)$ values fell between -4‰ and -2‰ , with a few localized grids approaching positive values. This pattern is consistent with the substantial contributions of transportation sources (Figure 3; $\delta^{15}\text{N}$: -28.1‰ – $+17\text{‰}$; median: -2.6‰). In contrast, more depleted $\delta^{15}\text{N}(\text{NO}_x)$ values ranging from -8‰ to -4‰ occurred mainly in inland and northwestern parts of the domain, suggesting significant contributions of residential sources, whose $\delta^{15}\text{N}$ endmember is approximately -11‰ (Table 1). Seasonally, April–June and July–September appeared slightly more enriched than the other periods in several hotspot regions, whereas January–March was the most depleted period. However, these seasonal differences were small relative to the persistent spatial contrast across the domain.

3.3. Model–Observation Comparisons

The simulated $\delta^{15}\text{N}(\text{NO}_x)$ values in the PRD region were compared with measured $\delta^{15}\text{N}$ values of atmospheric NO_3^- , the most accessible form for isotopic analysis, at two sites: Guangzhou (urban; indicated by the star in Figure 5) and Dinghushan (rural; indicated by the circle in Figure 5). The measured $\delta^{15}\text{N}(\text{NO}_3^-)$ values ranged from -3.9‰ to $+7.9\text{‰}$ ($n = 59$) at the urban Guangzhou site [9] and from $+5.1\text{‰}$ to $+11.1\text{‰}$ ($n = 15$) at the rural Dinghushan site [38]. These observed ranges overlap with recently published $\delta^{15}\text{N}$ measurements of particulate NO_3^- in Guangzhou, which ranged from $+2.59\text{‰}$ to $+9.3\text{‰}$ in July 2013 [39] and from -14.2‰ to $+13.0\text{‰}$ during September 2017–August 2018 [40].

At both urban Guangzhou and rural Dinghushan sites, the simulated $\delta^{15}\text{N}(\text{NO}_x)$ values were generally lower than the observations, but the underestimation was relatively milder at the urban Guangzhou site (Figure 5). The large underestimation at the rural Dinghushan site could be partly due to its proximity to a power plant to the west, which contributed significantly to local NO_x emissions, whereas the mixing caused by atmospheric transport was not considered in the current “emission-only” simulations.

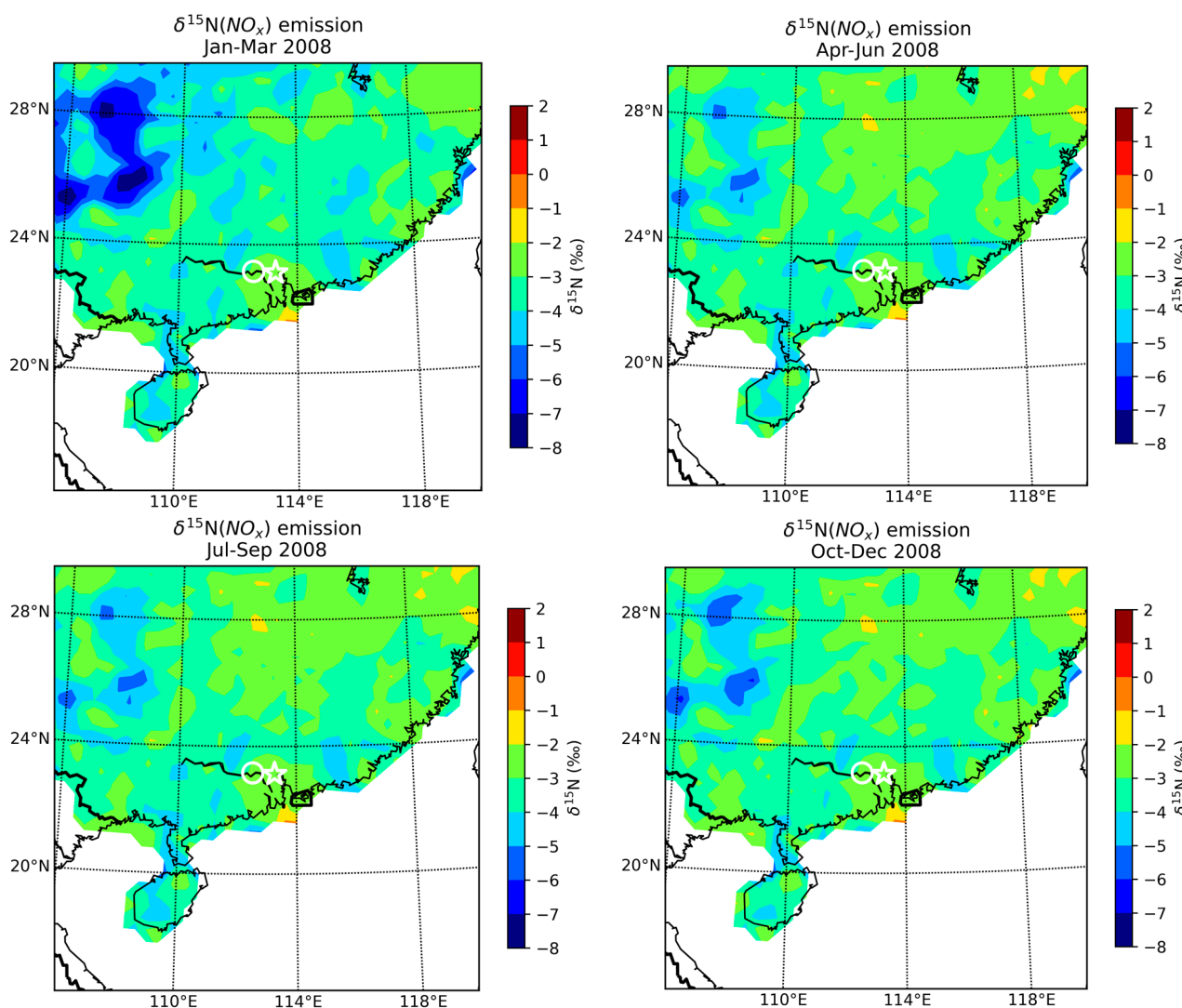


Figure 4. Geographical distribution of the simulated $\delta^{15}\text{N}(\text{NO}_x)$ values (‰) of total NO_x emissions across South China, including the PRD region, for four quarterly periods in 2008. The star and circle indicate the Guangzhou and Dinghushan sites, respectively.

Indeed, the uncertainty in our simulated $\delta^{15}\text{N}(\text{NO}_x)$ values, as well as the approximate 2–6‰ offset between the modeled $\delta^{15}\text{N}(\text{NO}_x)$ and the observed $\delta^{15}\text{N}(\text{NO}_3^-)$, may arise from three main factors. First, uncertainties in source-specific $\delta^{15}\text{N}$ endmembers, possible biases in the emission inventory, and the simplified representation of broad emission sectors may contribute to the model–measurement mismatch. Second, atmospheric transport, mixing and deposition can blur the regional $\delta^{15}\text{N}$ signal of emissions, depending on emission strength, mixing intensity, and deposition schemes [28]. Third, photolytic [41], kinetic [42], and equilibrium [43] isotope effects may occur during the transformation of NO_x into NO_3^- . Therefore, the present “emission-only” model is not expected to directly predict atmospheric $\delta^{15}\text{N}(\text{NO}_3^-)$. Rather, this study serves as a proof of concept by demonstrating the feasibility of incorporating ^{15}N into the NO_x emission inventory to simulate emission-derived $\delta^{15}\text{N}(\text{NO}_x)$ patterns and to evaluate the role of emission sources in controlling atmospheric NO_3^- in the PRD region. The remaining model–observation offset indicates that atmospheric nitrate is not determined solely by local NO_x emissions but is also influenced by subsequent atmospheric transport, mixing, deposition, and photochemical transformation.

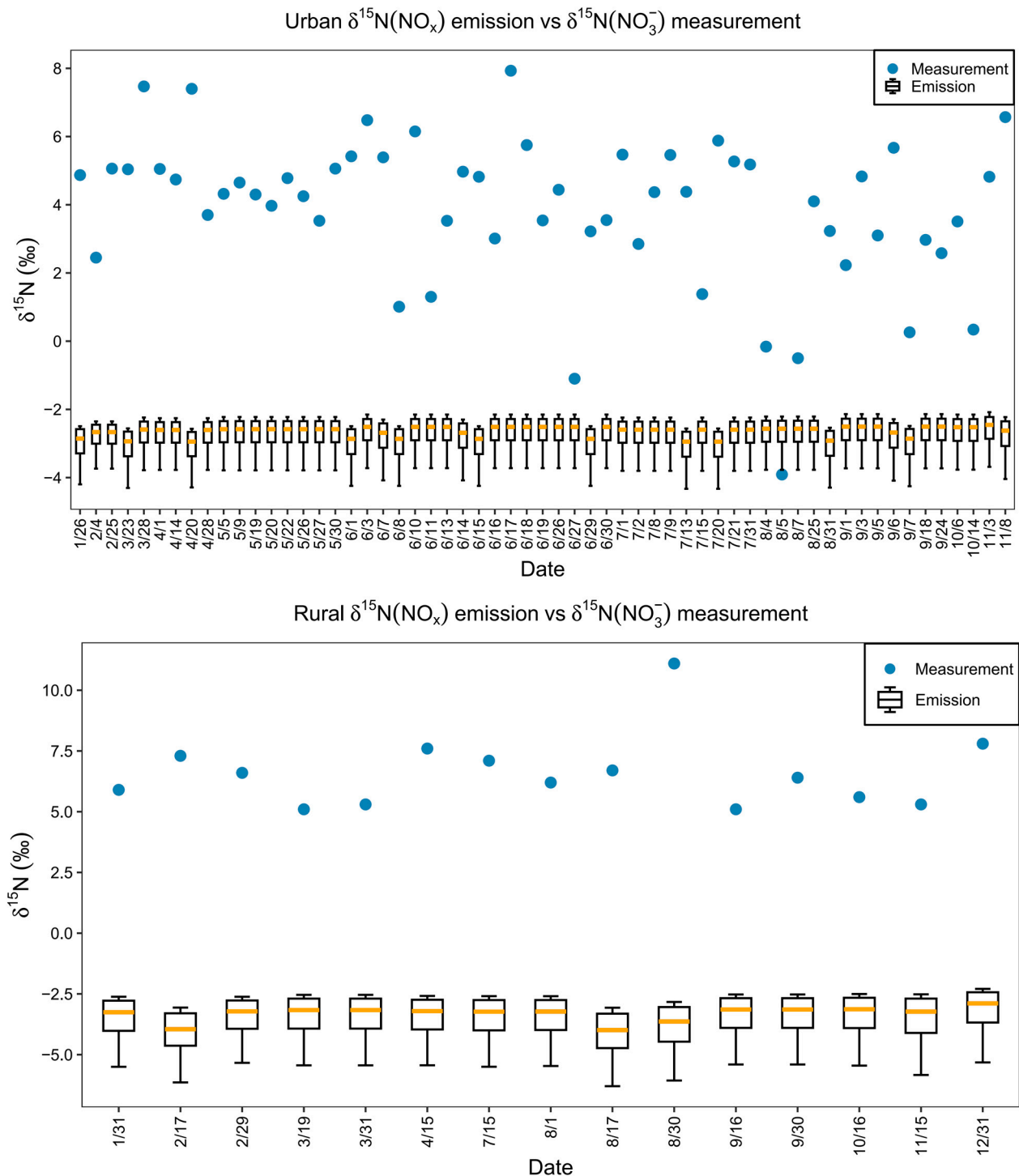


Figure 5. Simulated $\delta^{15}\text{N}(\text{NO}_x)$ values and measured $\delta^{15}\text{N}(\text{NO}_3^-)$ in the 36 km $\times$ 36 km grids centered on the urban Guangzhou (**top**) and rural Dinghushan (**bottom**) sites in 2008.

4. Conclusions

The 2008 MEIC inventory showed strong but seasonally persistent spatial heterogeneity in NO_x emissions across South China, with hotspots concentrated in the PRD and other urban clusters. Transportation was the dominant and most spatially pervasive source, while industrial emissions were regionally important, power plant emissions were localized, and residential sources contributed only minor fractions. The $\delta^{15}\text{N}(\text{NO}_x)$ values were then simulated using sectoral NO_x emissions from MEIC together with source-specific $\delta^{15}\text{N}$ values compiled from previous studies. The widespread $\delta^{15}\text{N}(\text{NO}_x)$ values between -4‰ and

−2‰ across South China, especially in the PRD region, were consistent with the substantial contribution of transportation sources. Compared with the measured $\delta^{15}\text{N}(\text{NO}_3^-)$ values at the urban Guangzhou and rural Dinghushan sites within the PRD region, the simulated $\delta^{15}\text{N}(\text{NO}_x)$ values were significantly lower, suggesting that an emission-only framework is insufficient to reproduce atmospheric NO_3^- isotopic composition. This discrepancy likely arises from the combined effects of inventory uncertainty, atmospheric transport and deposition, and isotope fractionation during NO_x oxidation. Future work will explore the impacts of atmospheric processes and tropospheric photochemistry by incorporating ^{15}N into the CMAQ model to enable process-resolved simulation of the emission, transport, chemical transformation, and deposition of atmospheric reactive N.

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Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

NO_x	Nitrogen oxides
MEIC	Multi-resolution Emission Inventory for China
PRD	Pearl River Delta

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