Atmospheric Ammonia in Beijing during the COVID-19 Outbreak: Concentrations, Sources, and Implications
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ABSTRACT: This study investigates the concentrations and $\delta^{15}\text{N}$ values of NH$_3$ in Beijing during and after the 2020 COVID-19 lockdown. Higher NH$_3$ concentrations and lower $\delta^{15}\text{N}$-NH$_3$(measured) were observed at most sites in 2020 compared to 2017. Except for a site inside a tunnel, NH$_3$ concentrations did not increase significantly after the lockdown had ended compared to those during the lockdown, while $\delta^{15}\text{N}$-NH$_3$(measured) increased by 2.1–9.9‰. Nonagricultural sources (fossil fuel and urban waste) overall contributed 81% and 62% of NH$_3$ at on-road (tunnel interior) and nonroad (CAU) sites in 2020, respectively, comparable to those in 2017 (without significant difference). The contribution of nonagricultural sources slightly increased after the lockdown compared to the contribution during the lockdown at the nonroad site and hardly changed at the tunnel interior site. Our results suggest that (1) unfavorable meteorological conditions, especially lower boundary layer heights and changes in regional transport patterns, might play a more important role than reduced anthropogenic emissions in the temporal variations of Beijing NH$_3$ and (2) the effect of reduced anthropogenic emissions, during the COVID-19 outbreak or with the future implementation of emission control strategies, on atmospheric NH$_3$ can be better demonstrated by isotope-based source apportionment of NH$_3$, rather than only by changes in NH$_3$ concentrations.

1. INTRODUCTION
It is well-known that air pollution across the world has derived mainly from rapid industrialization and urbanization since the industrial revolution, along a similar trajectory, offset in time, for both developed and developing countries. As the world’s second largest economy, China has experienced severe haze pollution over recent decades, and the Chinese government has taken strict legislative actions to improve air quality since 2013.1 Consequently, the concentrations of fine particles (PM$_{2.5}$), secondary inorganic aerosols [sulfate (SO$_4^{2-}$), nitrate (NO$_3^{-}$), and ammonium (NH$_4^+$) (SNA)], and gaseous acid precursors [sulfur dioxide (SO$_2$) and nitrogen oxides (NO$_x$)] significantly decreased nationwide.2–4 Ammonia (NH$_3$), as the chief alkaline gas in air, plays a very important role in the formation of SNA;5 however, NH$_3$ emissions remain high in China due to the absence of control measures. Controlling NH$_3$ emissions can be a key to reducing PM$_{2.5}$ pollution in China and other regions.7–8

In the winter of 2019 and 2020, the 2019 novel coronavirus (COVID-19) spread rapidly and attracted worldwide attention.9,10 In the Chinese New Year in January 2020, the Chinese government imposed nationwide restrictions on the movement of its population (a lockdown) to control the spread of COVID-19. Restrictions on commercial activities, energy consumption, and traffic led to a significant reduction in air pollutant emissions (e.g., SO$_2$, NO$_x$, and CO$_2$) and PM$_{2.5}$.11,12 Nevertheless, there were still several severe haze events over eastern China during the COVID-19 lockdown. Huang et al.13 found that the haze events were driven by enhancements of secondary pollution and the expected benefit of reduced emissions might be unexpectedly offset by enhanced secondary formation of PM. There is a more generally increasing importance of NH$_3$ emissions relative to SO$_2$ and NO$_x$ emissions due to the slower reduction of NH$_3$ emissions in China.14–16 However, studies of the variations of NH$_3$ concentration and source emissions during the COVID-19 outbreak have been scarce. One topic of interest is to quantify the impact of a series of control measures during the coronavirus outbreak on the concentrations of NH$_3$ in Beijing and to identify whether these activities are the main driver of short-term changes in atmospheric NH$_3$. Given the important role of NH$_3$ in SNA formation, such knowledge is key to designing effective future PM$_{2.5}$ mitigation strategies.

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In view of the decline in primary air pollutants and potential enhanced SNA formation during the COVID-19 outbreak with its reduced anthropogenic emissions, we hypothesize that the concentration of atmospheric NH₃ should decrease during the lockdown and return to normal levels after the lockdown, with the main reductions coming from the decline in vehicle emissions. Here, we describe a unique citywide experiment to test this hypothesis by monitoring the temporal and spatial variation in NH₃ concentrations and δ¹⁵N-NH₃ in Beijing during and after the 2020 lockdown period. In addition, we compared the concentrations and δ¹⁵N values of NH₃ in 2020 with those measured in 2017 at the same sites and reported studies¹⁷,¹⁸ about the 2014 APEC and 2015 Parade periods in Beijing to determine the impact of short-term emission regulations on atmospheric NH₃.

2. MATERIALS AND METHODS

2.1. Site Description. Five sampling sites were established in urban Beijing, China (Figure S1a and Table S1). Sites 1 and 2 are located at the edges of the third and fifth ring roads, respectively. Sites 3 and 4 are located inside and outside the Badaling Highway Tunnel, respectively. Site 5 is located at China Agricultural University’s west campus. Sites 1–4 represent on-road sites that are strongly and directly influenced by vehicle emissions, and site 5 represents a nonroad site that is mainly surrounded by campus and residential areas.

2.2. NH₃ Sampling and Measurements. Weekly atmospheric NH₃ samples were collected using ALPHA passive samplers (Figure S1b)¹⁹ before (December 27 to January 24, 2017) and during (January 25–31, 2017) Chinese New Year Holiday in 2017 and during (February 7–13, 2020) and after (March 25 to April 1, 2020) the COVID-19 lockdown in 2020. Collected samples were extracted with ultrapure water. The concentrations of NH₄⁺ were measured in sampler extracts using a continuous-flow analyzer (Seal AA3). More details about the passive samplers and quality control for NH₃ measurements have been described by Xu et al.²⁰ and Zhang et al.²¹ respectively. NH₃ samples were selected to determine δ¹⁵N-NH₃ with the hydroxylamine (NH₂OH) method, which has been described previously in detail by Li et al.²²

2.3. Calculation of Initial δ¹⁵N-NH₃. The δ¹⁵N values of the initial NH₃ (denoted as δ¹⁵N-NH₃ values) were calculated using an isotope mass-balance model (eq 1),²³ the measured δ¹⁵N-NH₄⁺ and the δ¹⁵N difference between gaseous NH₃ and particulate NH₄⁺ (eq 2),²⁴ as follows:

\[ \delta^{15}N-NH_3 = \delta^{15}N-NH_4^+ (\text{final}) - \epsilon_{(NH_4^+-NH_3)} (1 - f) \]  

\[ \delta^{15}N-NH_4^+ (\text{final}) = \delta^{15}N-NH_3 (\text{measured}) + \epsilon_{(NH_4^+-NH_3)} \]  

where \( \epsilon_{(NH_4^+-NH_3)} \) is the theoretical N isotopic fractionation factor between the gas and particle and equals 12.5₂₂ × 10₀₀₀ / \( T - 11.31 \),²₅ where \( T \) is the temperature in kelvin. \( f \) is the fraction of the initial gaseous NH₃ converted to particulate NH₄⁺ (NH₃⁺/NH₄⁺ in molar concentrations). We measured \( f \) values (the concurrent measurement of the aerosol NH₄⁺ and NH₃ concentrations) at site 5 (CAU site) (Table S2). However, particulate NH₄⁺ concentrations were not measured at the four on-road sites. For site 3 (tunnel interior), the \( f \) value was set at 0.044 on the basis of Walter et al.,²⁶ who reported \( f \) was extremely low in the tunnel. For the other three on-road sites (sites 1, 2, and 4), we performed a sensitivity analysis in which we modeled \( f \) values ranging from 0.0 to 1.0 to estimate the expected δ¹⁵N-NH₃.

2.4. Bayesian Mixing Model. The “SIAR” model, a widely used Bayesian stable isotope mixing model in R,²⁷–³⁰ and isotopic source signatures of NH₃ for Chinese sources²⁸ were used to quantify the contributions of specific NH₃ emission sources to NH₃ in air. Detailed information about used isotopic source signatures is presented in Text S1.

2.5. Backward Trajectory Analysis. Seventy-two hour backward trajectories arriving at Beijing were calculated every 3 h for each observation period 2 m above ground level using meteorological data (GDAS1) and the NOAA HYSPLIT4 model.³¹

2.6. Air Pollutant and Meteorological Data. Daily concentrations of PM₁₀, SO₂, NO₂, O₃, and CO were collected from the National Urban Air Quality Real-time Publishing Platform (http://106.37.208.233:20035/).

Meteorological factors, including relative humidity (RH), temperature (T), wind speed (WS), and wind direction (WD), were obtained from Weather Underground (http://www.wunderground.com).

3. RESULTS AND DISCUSSION

3.1. Spatial Features of Ambient NH₃ Concentrations. Many previous studies have reported air pollution levels in 2020 were much lower than in previous years due to COVID-19 restrictions limiting anthropogenic emissions.¹¹,³² However, in our study, higher NH₃ concentrations (19.9 ± 3.8 μg m⁻³) were observed in 2020 compared to 2017 (13.1 ± 1.6 μg m⁻³). The average NH₃ concentrations at on-road and nonroad sites in 2020 were 7.9 and 2.2 μg m⁻³ higher than those in 2017, respectively. Although this result at first is surprising, it is important to keep in mind that many factors could affect NH₃ concentrations. The considerable reductions in SO₂ and NOₓ in 2020 due to COVID-19 (Figure S2) likely reduced atmospheric HNO₃ and H₂SO₄ levels that would in turn reduce particulate NH₄⁺ levels and leave more NH₃ in the gas phase.³³ However, the fraction of NH₃ observed in the particle phase (FNH₄⁺) slightly increased to 0.57 in 2020 from 0.48 in 2017 at the CAU site (Figure S3). This was likely due to the enhanced oxidation of SO₂ and NOₓ, contributed by the increased photochemical oxidants, e.g., O₃ (Figure S2) and also OH, which increase the efficiencies of formation of SO₄²⁻ and NO₃⁻.³⁴,³⁵ In addition, meteorological conditions strongly regulate near-surface air pollutant concentrations.³⁶ In this study, lower boundary layer heights (Figure S4), higher temperatures, and comparable RHs (Figure S5) were observed during the sampling period in 2020 compared to 2017. Boundary layer height plays a vital role in the vertical dispersion of air pollutants emitted from the Earth’s surface.³⁷,³⁸ Low boundary layer heights can concentrate NH₃ through the boundary layer by holding surface-emitted air pollutants within the shallow and less well-ventilated surface layer, suppressing the vertical atmospheric dispersion and dilution.³⁸ A high T enhances the emissions of NH₃ from soil and urban wastes and promotes the volatilization of NH₃ from aerosol NH₄⁺ pools (e.g., NH₄NO₃).³⁹,⁴⁰ WS and WD can affect air pollution transport. Although WS values in 2020 were slightly lower than in 2017, the frequencies of south wind were much higher in 2020 (Figures S6 and S7). The southern area of Beijing, as a main part of the North China Plain (NCP), is a “hot spot” for NH₃ emissions due to the rapid development of

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industrialization, urbanization, and intensive agriculture. Meanwhile, the northern boundary of Beijing is surrounded by mountains, impeding the dispersion of air pollutants under south winds. With few significant NH3 emission sources in the mountains north of Beijing, the dominant wintertime north winds contribute little to NH3 concentrations in Beijing. Previous case studies suggested that the improvement in air quality and the effective decrease in the levels of air pollutants during the Parade Blue period in 2015 were due to a mixed effect of anthropogenic emission reduction measures and favorable natural meteorological conditions. However, our results indicate that increased unfavorable meteorological conditions might play a more important role than the emission reduction in the observed temporal variations of NH3 concentrations.

Short-term pollutant emission control measures have successfully improved air quality in Beijing in the past, e.g., decreased atmospheric NH3 levels in the 2014 APEC period and aerosol NH4+ levels in the 2015 Parade Blue period (Figure 1b,c). Generally, there was a rapid recovery in NH3 after the pollutant emission control measures had been removed. Although the level of vehicle transportation significantly decreased during the COVID-19 period in 2020, NH3 concentrations fluctuated during and after the lockdown in this study (Figure 1). In detail, NH3 concentrations did not significantly change at three of five sampling sites but decreased at site 1 and increased at site 3. An increased NH3 level at the tunnel site 3 (tunnel interior) after the lockdown suggests increased emissions from an increased number of vehicle trips with the return of people commuting from outside Beijing (https://report.amap.com/download.do). Stable and decreased NH3 concentrations at open sites might be contributed by the dilution effect of increased WS of north winds after the lockdown when emissions presumably increased (Figure S6); this hypothesis is supported by an observed decline in other air pollutants, including PM2.5, SO2, and NO2 (Figure S2). This observation differs from two previous case studies, i.e., 2014 APEC and 2015 Parade, where obvious recovery of SO2 and NOx emissions was observed (Figure S2).

3.2. 15N Stable Isotope. δ15N values of atmospheric NH3 reflect a mixture of contributions from different NH3 emission sources with distinct δ15N-NH3 values. In this study, the δ15N values of NH3 (δ15N-NH3(measured)) across all sites exhibited a slight decline in 2020 compared to 2017, averaging -32.5 ± 2.8 %e and -26.8 ± 2.3 %e, respectively (Figure 1d). A significant decline in δ15N-NH3 was found only at site 2. The values of δ15N-NH3(measured) on-road sites were higher than those of other NH3 emission sources. In this study, the δ15N-NH3 values at the interior tunnel site hardly changed, consistent with NH3 at that site being dominated by traffic sources both during and after the lockdown. These findings are in line with a previous study that reported δ15N values of NH3 averaging -34.8 %e, -41.1 %e, and -30.7 %e before, during, and after the 2014 APEC summit, respectively (Figure 1e), with its own restrictions on vehicle traffic, while there was a similar trend in NH3 concentrations (Figure 1b). In addition to the reduced local anthropogenic emissions in response to the emission controls, regional air mass transport also largely affects the δ15N values of NH3. NH4+ concentrations in PM2.5 decreased during the 2015 Parade period; however, the δ15N values of NH4+ were clearly higher than those before and after...
the measured values of (active and passive samplers) also play a considerable role in source contributions in the apportionment of NH\textsubscript{3}. To samplers were used in both the NH\textsubscript{3} measurements from this (Figure S5). It should be noticed that sampling methods partitioning), while RH was comparable between the two years possible that the large di... that reduced the in... fertilizer applications. As the key parameter in the nitrogen... 0.0 to 0.7, being 7–21% lower than those in 2017 when the f value was assumed to be the same in 2017 and 2020 (Figure S9). According to the positive impact of the f value on the estimated contribution of nonagricultural sources, the difference in the results of source apportionment between 2017 and 2020 could be masked by an increase in f of 0.20. Actually, the measured f value, based on site 5, increased by only ~0.1 in 2020 compared to 2017. Those results suggested a comparable (for on-road sites 1, 2, and 4) or more significant (for sites 3 and 5) contribution of the changed emission pattern than the partitioning effect (slightly increased f) to the decreased contribution of nonagricultural sources as well as δ¹⁵N-NH\textsubscript{3(measured)}. In 2020, to better control the spread of COVID-19, the government required most people to stay at home, shut down commercial activities, and restricted travel beginning in late January 2020.\textsuperscript{9,10,49} These measures dramatically affected the vehicular fleets and domestic flights (reduced by >70%).\textsuperscript{11} Also, energy demand and industrial output fell far below normal levels, such as coal-fired power plants and steel industries.\textsuperscript{53} Hence, decreased fossil fuel combustion emission contributions to NH\textsubscript{3} in 2020 were in line with the simultaneous expected reduction in NH\textsubscript{3} emissions from fossil fuel-related sources during the COVID-19 outbreak. In addition, the relative contributions of fossil fuel-dominated nonagricultural sources at the CAU nonroad site after the lockdown increased to 65% from 60% during the

![Figure 2](https://dx.doi.org/10.1021/acs.estlett.0c00756)

**Figure 2.** Contributions of four main NH\textsubscript{3} emission sources to atmospheric NH\textsubscript{3} at the on-road site (tunnel interior) and a nonroad site (CAU campus) in Beijing during the observation periods in 2017 and 2020.

3.3. Source Apportionment of NH\textsubscript{3}. The results of the “SIAR” model indicated that nonagricultural sources (fossil fuel and urban wastes) contributed approximately 81% and 60–65% of atmospheric NH\textsubscript{3} at on-road (tunnel interior) and nonroad (CAU campus) sites in 2020, respectively, being 6% and 11% lower than in 2017, respectively, but no significant differences were found (Figure 2). For the rest, on-road sites 1, 2, and 4, nonagricultural sources contributed 43–91% when f was varied from 0.0 to 0.7, being 7–21% lower than those in 2017 when the f value was assumed to be the same in 2017 and 2020 (Figure S9). According to the positive impact of the f value on the estimated contribution of nonagricultural sources, the difference in the results of source apportionment between 2017 and 2020 could be masked by an increase in f of 0.20. Actually, the measured f value, based on site 5, increased by only ~0.1 in 2020 compared to 2017. Those results suggested a comparable (for on-road sites 1, 2, and 4) or more significant (for sites 3 and 5) contribution of the changed emission pattern than the partitioning effect (slightly increased f) to the decreased contribution of nonagricultural sources as well as δ¹⁵N-NH\textsubscript{3(measured)}.
lockdown and slightly changed at most on-road sites, except at the exterior tunnel site that saw an increase of 21−37% when f was varied from 0 to 0.6. A similar result was found in Chang’s study,27 where the contribution of traffic initially decreased by 56.7% during the 2014 APEC summit and then doubled after the 2014 APEC summit. This was due to the strict controls of atmospheric pollutant emissions implemented during the summit.28,29 Wu et al.18 also reported fossil fuel sources and NH3 slip from industrial sources and power plants contributed less during the 2015 Parade Blue Period, the sources most sensitive to the emission control measures.

In addition to local NH3 emissions, regional transport plays an important role in the concentrations and source contributions of NH3.28 Local NH3 emissions are strongly associated with industrial and traffic sources, while large agricultural sources to the south in the NCP strongly influence urban Beijing NH3 via regional transport.18,29 Compared to agricultural areas to Beijing. NH3 emissions from those sources likely also increased due to higher T in 2020, as discussed above.

Hence, reduced fossil fuel consumption and air transport from the south of Beijing were two main drivers of decreased contributions of nonagricultural sources to NH3 in Beijing during the COVID-19 outbreak. In addition, higher T (Figure S5) in 2020 may lead to an uncertainty in this study by increasing NH3 emissions of T-dependent agricultural and nonagricultural sources and changes in NH3 partitioning to fine particles.

3.4. Implications for the NH3 Emission Control in the Future. Stable nitrogen isotope techniques were widely used to quantify the contributions of different sources to NH3 in previous studies.33,37−39 Our results indicated that δ15N-NH3 was a more sensitive indicator than NH3 concentrations to the changes in NH3 emissions, suggesting isotope-based source apportionment of NH3 could provide accurate and reliable evidence of the success of NH3 emission controls, consequent air quality improvement, and further mitigation strategy design. However, further work is needed to improve δ15N-NH3 collection techniques.26,47

■ ASSOCIATED CONTENT

* Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/10.1021/acs.estlett.0c00756.

Isotopic source signatures of NH3 used in this study (Text S1); locations of sampling sites, concentrations of air pollutants, NH3 partitioning, boundary layer height, T and RH, WD and WS, 72 h backward trajectories, expected δ15N-NH3 source apportionment in 2017 and 2020, and source apportionment during and after the lockdown in 2020 (Figures S1−S10, respectively); and detailed information about all sampling sites and used f values for sites 3 and S (Tables S1 and S2, respectively) (PDF)

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Notes

The authors declare no competing financial interest.

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