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Short Communication

Increasing importance of ammonia emission abatement in $PM_{2.5}$ pollution control

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A R T I C L E I N F O

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The coronavirus 2019 (COVID-19) pandemic has severely affected human health and economic activity in countries around the world [1,2]. To slow the spread of the COVID-19 outbreak, most countries have implemented a number of epidemic control interventions, including travel restrictions, business and industry closures, and requests for people to stay at home [2]. In China, the lockdown started in Wuhan City on 23 January 2020 and vehicle movement was restricted there on 26 January 2020. These measures quickly expanded to the entire nation and lasted for >3 weeks. Due to the abrupt and unprecedented restrictions on human activities, emissions of air pollutants were much reduced at local and national scales in China and other regions in the world during the lockdown [3–6]. Recent studies found overall decreases in primary pollutants, but severe haze pollution still occurred [7,8].

The response of atmospheric ammonia (NH_3) over this period, which has a crucial role in secondary aerosol formation contributing to $PM_{2.5}$ (particles smaller than 2.5 μm) air pollution [9,10], is still unknown. Agriculture is conventionally viewed as the dominant source of NH_3 [11]. However, this has been challenged by several recent studies that suggested fuel combustion might exceed agriculture as a source of ambient NH_3 in Chinese urban atmospheres [12,13]. The unprecedented emission controls on fossil fuel-based sources during the COVID-19 pandemic provide a unique opportunity to identify NH_3 sources and their potential contribution to $PM_{2.5}$. Here we analyze surface NH_3 measurements from a Nationwide Nitrogen Deposition Monitoring Network (NNDMN) from 2015 to 2020 (Table S1 online), combined with real-time *in situ* measurements, satellite observations, and atmospheric chemistry model simulations for the pre-COVID period (1–26 January 2020) and COVID-lockdown period (27 January–26 February 2020). We investigated changes in atmospheric NH_3 concentrations as caused by the lockdown measures in China, and the potential need for agricultural emission mitigation in $PM_{2.5}$ abatement when large reductions in non-agricultural pollutant emissions are expected in the future. The detailed information on surface NH_3 measurements, satellite NH_3 observations, GEOS-Chem simulations, as

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well as statistical analyses is described in the Supplementary materials (online).

Ambient mean NH_3 concentrations at the 36 NNDMN monitoring sites significantly ($P < 0.01$) increased (on average by 17%) during the COVID-lockdown period (average 9.0 ± 6.1 μg m^{-3}) compared to those during the pre-COVID period (average 7.7 ± 5.7 μg m^{-3}) (Fig. 1a and Table S1 online), but with considerable variation. Separating sites by land-use type, mean NH_3 concentrations showed significant ($P < 0.01$) increases during the COVID-lockdown period at rural (8.1 ± 6.2 vs. 9.9 ± 6.6 μg m^{-3}) sites while small increase and non-significant decrease were found at background (3.1 ± 1.8 vs. 3.4 ± 1.6 μg m^{-3}) and urban (10.1 ± 3.0 vs. 9.8 ± 3.0 μg m^{-3}) sites (Fig. 1b). During equivalent dates for 2015–2019, mean NH_3 concentrations ranged from 5.2 ± 3.5 to 7.7 ± 5.7 μg m^{-3} in the "pre-COVID" period, and from 5.7 ± 4.3 to 9.0 ± 6.1 μg m^{-3} in the "COVID-lockdown" period (Fig. S1a online). Compared to these levels, NH_3 concentrations in 2020 were 38%–65% higher during the pre-COVID period and 53%–62% higher during the COVID-lockdown (Fig. S1a online). The increases in NH_3 concentrations during the COVID-lockdown were larger in 2020 (17%) than during the same periods in 2015–2019 (9%) (Fig. S1b online).

Based on analysis of real-time measurements, a small increase in daily mean NH_3 concentrations was observed at the urban Beijing site (8%, $P > 0.05$) during the COVID-lockdown period compared to the pre-COVID period. Similarly, daily mean NH_3 concentrations increased at urban (Pudong: 13%) and rural (Chongming: 35%, $P < 0.01$) sites in Shanghai (Fig. 1c, d). Mean concentrations of secondary inorganic aerosols (NH_4^+, NO_3^-, SO_4^{2-}, and Cl^-, abbreviated as SIA) at the urban Beijing site increased from the pre-COVID period (21.6 μg m^{-3}) to the COVID-lockdown period (41.4 μg m^{-3}) (Fig. S2a online). Meanwhile, the ratio of aerosol NH_4^+ to total NH_x (NH_3 + NH_4^+) concentrations denote as $\varepsilon(NH_4^+)$ showed increases of about 28% ($P > 0.05$) during the COVID-lockdown relative to pre-COVID (Fig. 1e, f). By contrast, in Shanghai, the average SIA concentrations decreased from 33.3 (rural) and 30.6 (urban) μg m^{-3} during the pre-COVID period to 22.2 (rural) and 21.6 μg m^{-3} (urban) during the COVID-lockdown (Fig. S2b, c online), and $\varepsilon(NH_4^+)$ reduced by 9% and 7% at rural and urban sites, respectively, during the COVID-lockdown (Fig. 1e, f). The similar increases in gaseous NH_3, but different changes in $\varepsilon(NH_4^+)$ between Beijing and Shanghai cities reflect different driving factors in northern and southern China as discussed below.

Increases of NH_3 levels during the COVID-lockdown period were also seen in satellite observations. IASI (Infrared Atmospheric Sounding Interferometer) NH_3 columns increased by 7% across China from the pre-COVID period to the COVID-lockdown period in 2020, with the largest increase (25%) observed in the North

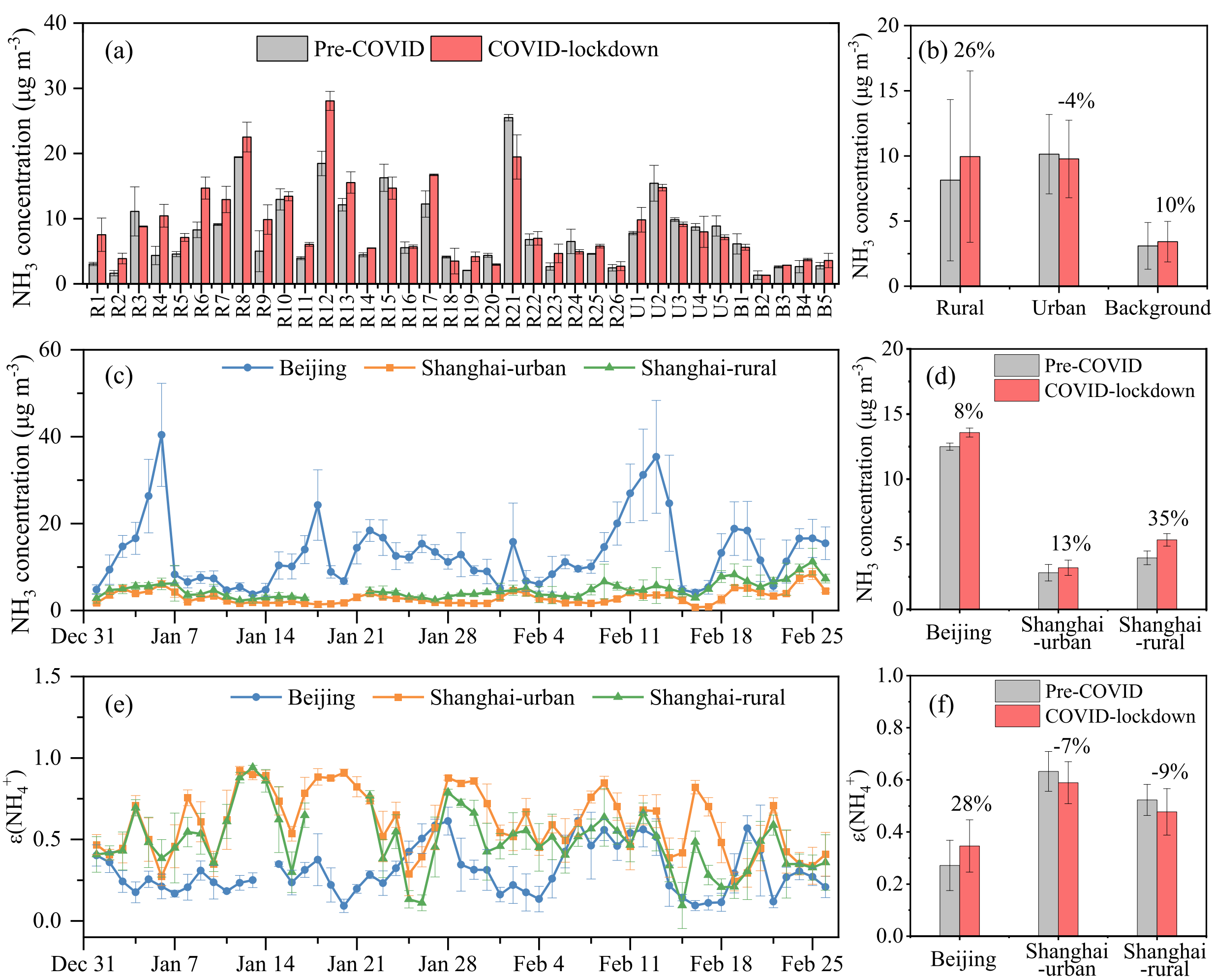

Fig. 1. Ambient NH_3 concentrations at 36 sites during the pre-COVID (1–26 January 2020) and COVID-lockdown (27 January–26 February 2020) periods (a), and average concentrations for rural, urban, and background sites (b). Daily mean NH_3 concentrations at urban site in Beijing and urban and rural sites in Shanghai city (c), and average concentrations during the pre-COVID and COVID-lockdown period (d). Daily mean $\varepsilon(NH_4^+)$ at urban site in Beijing and urban and rural sites in Shanghai city (e), and average concentrations during the pre-COVID and COVID-lockdown period (f). The letters R, U, and B denote, respectively, rural, urban and background sites.

China Plain (Fig. 2). Extracting IASI column values above NNDMN monitoring sites, mean NH_3 columns increased by 33.7% from the 2020 pre-COVID to the COVID-lockdown periods (Fig. S3 online).

We designed a series of GEOS-Chem atmospheric chemistry model simulations as shown in Table S2 (online) to investigate drivers of changes in observed NH_3 concentrations between the pre-COVID and COVID-lockdown periods. The impact of meteorological conditions was assessed by analyzing the GEOS-FP assimilated meteorological fields. The surface variables of the GEOS-FP data, analyzed during the pre-COVID and COVID-lockdown periods, show good agreements with observations (Fig. S4 online). In the GEOS-Chem standard simulation, we applied the latest estimates of anthropogenic emissions in China. The national mean anthropogenic NO_x and SO_2 emissions decreased by 44% (8 Gg N d^{-1}) and 31% (4 Gg S d^{-1}), respectively, between the pre-COVID and COVID-lockdown period (Fig. S5 online) [7,14]. The GEOS-Chem simulations are able to capture the changes in concentrations of secondary inorganic ions (NH_4^+, SO_4^{2-}, and NO_3^-) and major air pollutants (NO_2, SO_2, $PM_{2.5}$, O_3, and CO) between the pre-COVID and COVID-lockdown periods (Figs. S6 and S7 online). The non-agricultural NH_3 emissions were assumed to have the same percentage changes as anthropogenic NO_x emissions and decreased by 1 Gg N d^{-1} between the two periods. In the standard simulation we assumed that agricultural NH_3 emissions were unchanged between the pre-COVID and the COVID lockdown period. The influence of meteorological-driven NH_3 emission changes (e.g., warmer during COVID lockdown than pre-COVID) was also analyzed in a sensitivity simulation (EF_metf in Table S2 online).

As estimated by the standard simulation (with agricultural NH_3 emissions unchanged), the model results showed near zero changes (light purple, Fig. S8a online) in the mean NH_3 concentration averaged over the 36 NNDMN monitoring sites between the pre-COVID and COVID-lockdown periods. Using sensitivity simulations with fixed non-agricultural NH_3 emissions (i.e., emissions fixed to the pre-COVID condition; Pre-COVID_NH_3 in Table S2 online), fixed anthropogenic emissions of other species (mainly SO_2 and NO_x; Pre-COVID_other in Table S2 online), and fixed meteorology (Pre-COVID_metf in Table S2 online), we could separate their contributions to the NH_3 concentration changes (Methods; Fig. S9 online). Decreased anthropogenic emissions of air pollutants other than NH_3 during the lockdown period increased the mean NH_3 concentration by 0.8 μg m^{-3} (yellow, Fig. S8a online) of which 85% (0.6 μg m^{-3}) were shown to be caused by reduced NO_x and VOCs emissions. The reduction in anthropogenic emissions largely suppressed conversion of NH_3 to NH_4^+ aerosol (Figs. S9 and S10 online), but this increase in NH_3 concentrations was largely offset by a 0.7 μg m^{-3} reduction due to the decreases in non-agricultural NH_3 source emissions (mainly from vehicle emissions in the model, yellow, Fig. S8a online).

The standard model simulation showed different changes in surface NH_3 concentrations during the COVID lockdown in northern China vs. southern China (Fig. S8c, d online). The model captured the observed NH_3 increase in Southeast China (1.5 μg m^{-3} in the model versus 1.6 μg m^{-3} in observations), but underestimated changes over the North China Plain (1–2 μg m^{-3} decreases in the model vs. up to 6 μg m^{-3} increases in the observations) (Fig. S8b online). The differences between the two regions were largely attributed to the different changes associated with reductions in anthropogenic emissions during the COVID lockdown (Fig. S9a, b online). Reductions of SO_2 and NO_x emissions tended to reduce the formation of sulfate and nitrate aerosols, which allow more gaseous NH_3 to stay in the atmosphere. Such effects were distinct in central and Southeast China (Fig. S9b online), while insignificant or even led to slight NH_3 decreases over northern China, including Beijing and the northern part of North China Plain (Fig. S9a online). This model simulated spatial features were consistent with the synchronous measurements of NH_3 and NH_4^+ as reported above: decreases in $\varepsilon(NH_4^+)$ in Shanghai (Southeast China) and increases in $\varepsilon(NH_4^+)$ in Beijing (Northern China) during the COVID lockdown.

Changes in non-agricultural NH_3 emissions and meteorological conditions led to additional decreases in NH_3 concentrations over North China Plain (Fig. S9a online). This analysis provided a strong hint that the agricultural NH_3 emissions should have increased during the COVID-lockdown period. Here we tested two possible factors driving the predicted increases in agricultural NH_3 emissions. First, we found that accounting for meteorological influences on NH_3 volatilization following Paulot et al. [15] could result in 1 Gg d^{-1} increases in agricultural emissions during the COVID lockdown. Second, official reports from the Ministry of Agriculture and Rural Affairs of the People's Republic of China (http://www.moa.-

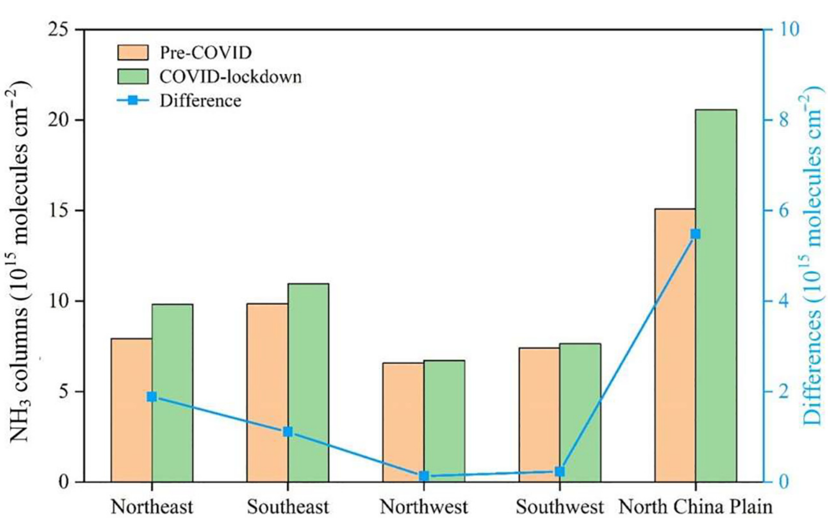

Fig. 2. IASI (Infrared Atmospheric Sounding Interferometer) satellite observed NH_3 columns (in unit of molecule cm^{-2}) over the five regions of China during the pre-COVID period, the COVID-lockdown period, and the COVID-lockdown minus pre-COVID differences in 2020. The five regions included Northeast (Heilongjiang, Jilin, and Liaoning), Southeast (Jiangsu, Anhui, Hubei, Shanghai, Zhejiang, Hunan, Jiangxi, Fujian, Guangxi, Hainan, and Guangdong), Northwest (Inner Mongolia, Xinjiang, Gansu, Shaanxi, Ningxia, Qinghai, and Tibet), Southwest (Sichuan, Chongqing, Yunnan, and Guizhou), North China Plain (Hebei, Henan, Beijing, Shanxi, Shandong, and Tianjin).

gov.cn/) showed that the COVID lockdown partly inhibited the movement and sale of agricultural products, with the breeding stock of hogs and chickens increased by 2.8% and 3.6%, respectively. The model sensitivity simulations considering these NH_3 emission changes (EF_metf and +5%_manure in Table S2 online) estimated increases of ∼20%–50% in surface NH_3 concentrations (light blue, Fig. S8a online) allowing the model to better capture the observed increases. The analyses above concluded that increased NH_3 concentrations during the COVID-lockdown period could be explained by the reduced conversion of gaseous NH_3 to NH_4^+ aerosols in the southern China and increases in agricultural NH_3 emissions in northern China. We estimated that a 20% reduction in agricultural NH_3 emissions would be needed to offset the increases in national mean surface NH_3 concentrations during the COVID-lockdown period (dark blue, Fig. S8a online).

The large reductions of NO_x emissions during the COVID lockdown have led to increases in surface ozone and atmospheric oxidizing capacity facilitating secondary aerosol formation [7]. This was also shown in the GEOS-Chem standard simulation using the SO_4^{2-}/SO_2 and NO_3^-/NO_x ratios as proxies for secondary inorganic aerosol formation efficiency that showed higher values during the COVID lockdown than the pre-COVID period over both the North China Plain and the Yangtze River Delta (in the Southeast China) (Fig. S8c, d online). The sensitivity simulations that applied emissions fixed to the pre-COVID conditions (pre-COVID_all, yellow) confirmed that decreases in other anthropogenic emissions (e.g., NO_x emissions) resulted in the enhancement of SO_4^{2-} and NO_3^- formation in both regions (Fig. S8c, d online), consistent with Huang et al. [7]. We found that a 50% reduction of this source would fully offset the enhanced secondary inorganic aerosol formation during the COVID lockdown (blue bars in Fig. S8c, d online). This 50% reduction was larger than the 31% reduction in NO_x and 27% reduction in SO_2 emissions over this period [7], suggesting that strict agricultural NH_3 emission control strategies are needed to suppress winter haze formation in addition to NO_x and SO_2 emission controls.

In summary, we reported significant and large-scale increases in atmospheric NH_3 concentrations over China during the COVID-19 lockdown. The increases in NH_3 concentrations were most distinct at rural sites (22% enhancement), less notable at urban and background sites, and were stronger during COVID-19 in 2020 than the equivalent periods in earlier years. In northern (southern) China the NH_3 enhancements were largely driven by increased agricultural NH_3 emissions (lowered aerosol partitioning). Such adverse effects on inorganic aerosol formation can be offset by a 50% reduction of agricultural NH_3 emissions.

Conflict of interest

The authors declare that they have no conflict of interest.

Acknowledgments

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Author contributions

Wen Xu, Xuejun Liu, Lin Zhang, and Kebin He designed the study. Wen Xu, Yuanhong Zhao, Zhang Wen, Yunhua Chang, Yele Sun, Yuepeng Pan, Yixin Guo, Lin Zhang, Xin Ma, Zhipeng Sha, Ziyue Li, Jiahui Kang, Lei Liu, Xiuming Zhang, Baojing Gu, Martin Van Damme, Lieven Clarisse, and Pierre-François Coheur conducted the research (collected the data, performed the measurements, and prepared the figures and tables). Wen Xu, Yuanhong Zhao, Lin Zhang, Xuejun Liu, Jeffrey L. Collett Jr, and Keith Goulding wrote the manuscript. All authors were involved in the discussion and interpretation of the data.

Appendix A. Supplementary materials

Supplementary materials to this short communication can be found online at https://doi.org/10.1016/j.scib.2022.07.021.

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