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Rapid Increase in China's Industrial Ammonia Emissions: Evidence from Unit-Based Mapping

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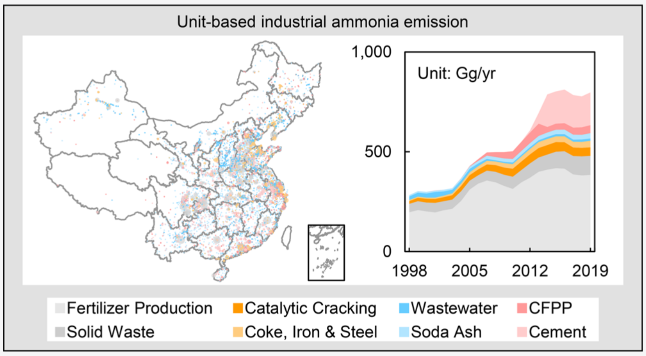
ABSTRACT: Ammonia (NH_3) is an important precursor of secondary inorganic aerosols and greatly impacts nitrogen deposition and acid rain. Previous studies have mainly focused on the agricultural NH_3 emissions, while recent research has noted that industrial sources could be significant in China. However, detailed estimates of NH_3 emitted from industrial sectors in China are lacking. Here, we established an unprecedented high-spatial-resolution data set of China's industrial NH_3 emissions using up-to-date measurements of NH_3 and point source-level information covering eight major industries and 27 subdivided process categories. We found that China emitted 798 (90% confidence interval: 668−933) gigagrams of industrial NH_3 into the atmosphere in 2019, equivalent to 44 ± 20% of the industrial emissions worldwide; this flux is 3-fold larger than that in 1998 and has fluctuated since 2014. Furthermore, although fertilizer production is responsible for approximately half of the emissions in China, the emissions from cement production and coal-fired power plants increased dramatically from near zero to 164 and 41 gigagrams, respectively, in the past two decades, primarily due to the NH_3 escape caused by the large-scale application of the denitration process. Our results reveal that, unlike other major air pollutants, China's industrial NH_3 emission control is still in a critical period, and stricter NH_3 emission standards and innovation in pollution control technologies are highly desirable.



KEYWORDS: *ammonia (NH_3), precursor of secondary inorganic aerosols, point source-level data set, industrial sources, NH_3 escape*

1. INTRODUCTION

As the main alkaline gas in the atmosphere, ammonia (NH_3) can react with acid gas (e.g., nitric and sulfuric vapor) and generate secondary inorganic aerosols, which account for up to 60% of China's $PM_{2.5}$ (particles with a diameter smaller than 2.5 μm).[1−3] Many studies have shown that $PM_{2.5}$ has adverse health effects on the population, such as respiratory diseases, cardiovascular diseases, and even cancer, and causes increased morbidity and mortality worldwide.[4−6] $PM_{2.5}$ can also influence radiative forcing and global warming trends.[7−9] In addition, although NH_3 is essential in balancing the acidity of precipitation,[10] it is an important part of nitrogen deposition and can acidify the soil, affect biodiversity, lead to eutrophication of aquatic environments, and disturb the carbon cycle.[11−14]

Many previous studies have estimated global and regional NH_3 emissions,[11,15,16] and most of these efforts mainly focused on the atmospheric emissions of NH_3 from agricultural sources.[17,18] A new generation of estimates has shown that the combustion of fossil fuels is the main source of atmospheric NH_3 in urban areas during heavily polluted periods, indicating a significant contribution from non-agricultural sectors.[19−21] Though it is commonly believed that agricultural sources contribute the majority of NH_3 emissions worldwide, NH_3 from nonagricultural sources, such as combustion, traffic, and industrial emissions, may have a significant impact on human health in urban areas[5,6] and have gained more and more attention in recent years.[20,22,23]

Comprehensively understanding the spatial and temporal distributions of NH_3 emissions is essential for determining their impact on the environment and human health. China is an interesting testing ground to study nonagricultural NH_3 emissions because (1) China is experiencing rapid industrialization and urbanization, which are greatly related to industrial pollutant emissions; (2) the large population in China, especially in urban areas, leads to potential NH_3-related health

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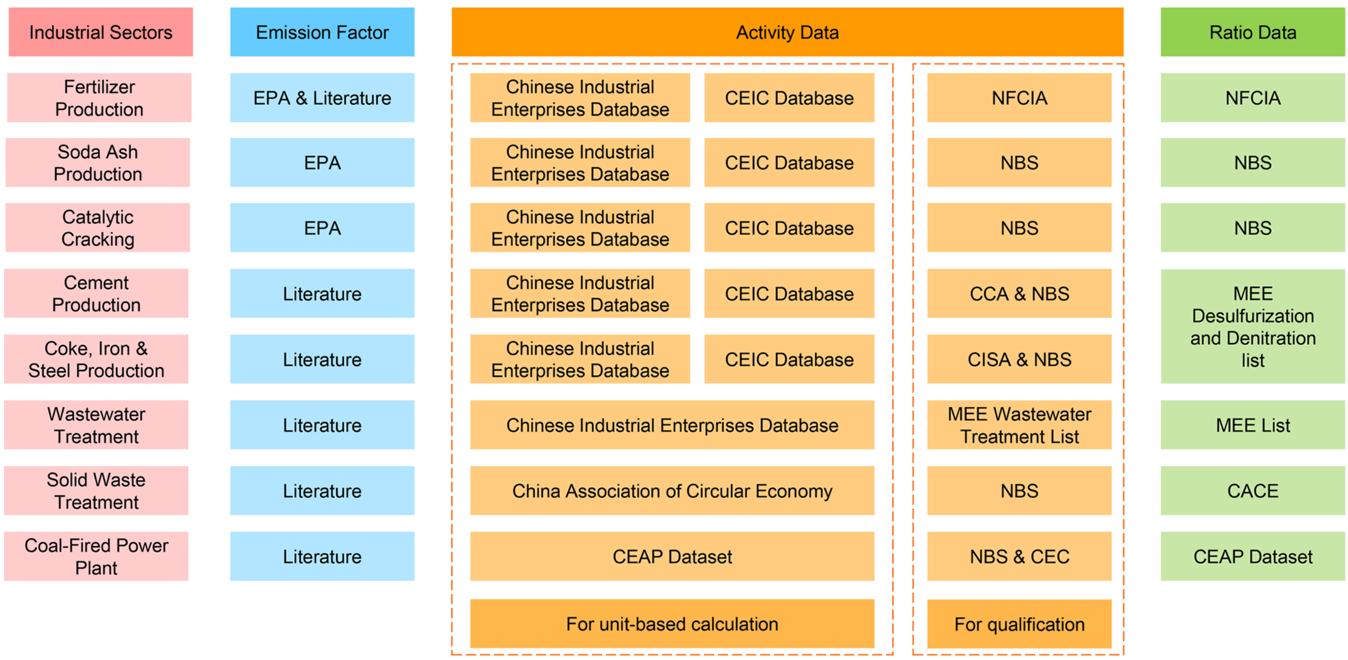

Figure 1. Main framework and databases used in this study. Abbreviation: EPA: Environmental Protection Agency, CEIC: Census and Economic Information Center, CEAP: China Emissions Accounts for Power plants, CNFIA: China Nitrogen Fertilizer Industrial Association, NBS: National Bureau of Statistics, CCA: China Cement Association, CISA: China Iron & Steel Association, MEE: Ministry of Ecologic Environment, CEC: China Electrical Council, CACE: China Association of Circular Economy.

risks; and (3) industrial NH_3 emission standards and relevant policies are still lacking. Previous estimations of NH_3 emissions in China usually focused on improving the accuracy in the agricultural sector.[24−26] However, recent assessments stressing nonagricultural (e.g., combustion and traffic-related) NH_3 emissions are increasing[27,28] and have shown that NH_3 emissions from these sources are underestimated.[20,29−31] For example, Chang et al. stressed the contribution of non-agricultural NH_3 emissions at the municipal level, and Fu et al. initially suggested the significance of industrial NH_3 emissions.[31,32] Furthermore, based on satellite inversion data, Van Damme et al. and Dammers et al. revealed that industrial emissions could be an important but previously under-estimated source across China.[22,23] However, insufficient resolution and the lack of point source data hinder the precise calculation of industrial NH_3 emissions.[25,28] Combined with the lack of updated localized emission factors (EFs), the existing industrial NH_3 estimates present considerable uncertainties (range 179−863 Gg in 2014).[15,25] These studies called for more attention to industrial point sources, highlighting that a high-resolution estimate of atmospheric NH_3 emissions from industrial sources in China is desirable.

In this study, we produced a new unit-based data set regarding the annual atmospheric NH_3 emissions from industrial sources in China. This data set is constructed based on plant-level activity data and updated localized EFs of NH_3 generated from newly gathered investigated and measured data. The composition and contribution of industrial sources at provincial and municipal levels, together with emission time series, were quantified. Furthermore, we discussed the impacts of industrial restructuring and innovation on the emissions in China and proposed relevant policy recommendations. We aim to refine the spatial resolution and narrow the uncertainty range of atmospheric NH_3 emissions from industrial sources and identify emission hotspots across China in the past two decades. Our study could assist in developing better target strategies to reduce the impacts of NH_3 emissions.

2. MATERIALS AND METHODS

We employed a bottom-up method for the estimation of atmospheric NH_3 emissions from point sources following published literature.[11,16,25,28,33] This method has long been applied to various air pollutants and has served as a basic and solid estimation of emissions for further model simulation. Eight main industrial sectors were covered (Figure 1), namely, fertilizer production, soda ash production, catalytic cracking, coke, iron and steel production, cement production, waste-water treatment, solid waste treatment, and coal-fired power plants (CFPPs). Some other industries should also be included in the data set when point source-level activity data and/or EFs of data are available, namely, geothermal industry, beet sugar production, explosive production, nickel processing, mineral wool production, and pulp and paper industry.[34,35] Up-to-date EFs were used.[16,33,35−41] As shown in Figure 1, plant-level activity data were collected from the Chinese Industrial Enterprise Database and other specific industrial associations. Together with the price for each product acquired from the Census and Economic Information Center (CEIC) database, the unit-based production was calculated. For validation, the annual production of each industry derived from our method was compared with reports from government agencies and authorized associations (Figure 1). The detailed application ratios of production methods and pollution control tech-nologies were collected from industrial associations and governments, and were processed (Figure 1).

2.1. Compilation of an Industrial NH_3 Emission Data Set. Hotspots showing exceptionally high density of point sources were identified, and provinces with large contributions to total emissions were stressed. Key time points of the

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emission trend were revealed. The results were compared to previous estimates. The unit-based atmospheric emission of NH_3 was calculated as follows:

$$E^{ijk} = EF^{ijk} \times AD^{ijk} \times \text{ratio type}^{ijk} \times \text{ratio tech}^{ijk}$$

where E is the amount of NH_3 emissions (Gg/yr); i is the index for a given year; j is the index for a given province/point; k is the index for a given industry; EF is the emission factor (kg/t); AD is the activity data (t); ratio type is the ratio of the yield (ton) of one kind of product to the yield (ton) of all products in a given industry (%); ratio tech is the ratio of the yield (ton) of products using a specific Air Pollution Cleaning Device (APCD) to the yield (ton) of all products in a given industry (%). The EFs used in this study are provided in Supporting Information (SI) Table S1. By calculating each year's provincial data for each industry, the composition of NH_3 emissions of each province is obtained, and analyses of their differences and relevant changes of contributions over time are conducted. Different products were considered in industries such as fertilizer production, soda ash production, and catalytic cracking, where NH_3 emissions are more relevant with raw materials themselves. Up-to-date application ratios of APCDs were studied in industries such as cement production and CFPPs, where NH_3 emissions are produced in the cleaning process.[42] Specific techniques and APCDs used together with their ratios are presented in SI Tables S2 and S3. Per unit, per industry-type information in our emission inventory has been uploaded to Dryad (https://doi.org/10.5061/dryad.63xsj3v3w). The point source data for solid waste treatment includes incineration plants, whereas data for landfill and composting plants are not officially reported.

2.2. Activity Data. Specific production and financial information on point sources from 1998 to 2013 were acquired from the annual report of the Chinese Industrial Enterprises Database.[43] In 2013, a total of 9817 plants containing detailed information on APCD and whose sales revenues exceeded five million yuan (766,295 US dollars in 2020) were obtained, covering over 95% of plants in each industry. Detailed location information was acquired by converting the names of the plants to longitudes and latitudes using Baidu API (version 3). To show more complete and up-to-date trends of NH_3 emissions, data from 2014 to 2019 were calculated with provincial activity data obtained from the National Statistical Bureau of China (NSB).[44]

For fertilizer production, soda ash production, catalytic cracking, cement production, coke, and iron and steel production, the output value of each plant was obtained from the Chinese Industrial Enterprises Database (Figure 1). The price for each product was acquired from the Census and CEIC database, in which the yearly price is calculated with the average monthly price given. Then with the output value and the price, the production was calculated for each plant. For wastewater treatment plants, the treatment amount for each plant was obtained from the Chinese Industrial Enterprises Database, and the specific techniques used for each plant were obtained from the Ministry of Ecology and Environment (Figure 1).[45] For solid waste treatment plants, the treatment amount for each plant was acquired from the China Association of Circular Economy. For CFPPs, the electricity production and information about cleaning devices were obtained from the China Emissions Accounts for Power Plants (CEAP) data set (Figure 1).[46]

The temporal trends of production of each subcategory are illustrated in SI Figure S1, together with the gross domestic product in China from 1998−2019. Data regarding fertilizer production, cement production, iron and steel production, wastewater treatment, and CFPPs mainly came from the China Nitrogen Fertilizer Industry Association, the China Cement Association, the China Iron and Steel Association, the Ministry of Ecology and Environment, and the China Electricity Council, respectively. Data of other industries came from NSB. The validation of fertilizer production is shown in SI Figure S2.

2.3. Uncertainty Analysis. Monte Carlo simulation was employed to generate the probabilistic distribution of atmospheric NH_3 emissions in each industrial sector,[28,47−49] and we ran the models for 10 000 iterations in accordance with previous studies. The distribution of industrial NH_3 concentrations was set following published literature.[25] Uncertainties in NH_3 emissions originated from the values of the EFs, AD, and ratios used. In each iteration, the NH_3 emissions of each sector were calculated as the product of three multipliers (EFs, AD, and ratios), and the multipliers were randomly generated based on their distributions, mean values, and coefficients of variations (CVs). The CVs for EFs, AD, and ratios of each industry are listed in SI Table S4, and they were set following published literature.[25] Notably, the CVs for AD in industries with point-source-level information were set as 5% due to the improved accuracy, lower than the CVs (10%) used in previous literature.[50] The distribution of final emissions was generated for each year from 1998 to 2019. A P5−P95 confidence interval was employed as the uncertainty range,[51] where P5 and P95 represent 5% and 95% probabilities of the emissions, respectively.

3. RESULTS AND DISCUSSION

3.1. NH_3 Emissions from Industrial Sectors. Our unit-based estimate indicates that the industrial NH_3 emissions were 798 Gg (range: 668−933 Gg) in China in 2019. This flux is equivalent to 44 ± 20% of the annual industrial NH_3 emissions to the atmosphere worldwide in the 2010s,[15,22,23] although China's land area only accounts for 7% of the globe, demonstrating that China is disproportionately important to industrial NH_3 emission control. The NH_3 emissions from fertilizer production were 384 Gg (323−447 Gg) in 2019, accounting for 48.1% of total industrial emissions. Among the rest of the NH_3 emissions, cement production and solid waste treatment were the leaders, contributing 164 Gg (143−186 Gg) and 96 Gg (79−113 Gg), respectively, with the other sources contributing less than 10%.

NH_3 emissions from different industries are caused by either raw materials or air pollution control technologies, and specific calculations and policy regulations are required for each type. One type of emission is derived from the raw materials in which nitrogen is contained and converted into NH_3 during these processes, such as fertilizer production, soda ash production, and catalytic cracking that have considerable amounts of NH_3 emissions (384, 27, and 43 Gg in 2019, respectively). Then, the amounts of NH_3 emissions from these industries are determined by the number of products manufactured, given that the production technology has not changed significantly over a short period of time.[25] This helps explain why fertilizer production is the dominant emitter of industrial NH_3,[24−26] as substantial amounts of nitrogen are contained in the raw materials, and China produces large

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quantities of nitrogen fertilizer (71 Tg in 2020). For different product types, the production processes differ slightly, resulting in diverse EFs. For fertilizer production, the production processes of five major products were differentiated: urea, ammonium bicarbonate, ammonium chloride, ammonium nitrate, and synthetic ammonia. Similarly, the production of catalytic cracking, including gasoline, diesel, and kerosene, and the respective NH_3-emission-related processes are considered.[25] Though NH_3 emissions from the waste disposal sector are more relevant to the amount and characteristics of the waste they collected, the technology of wastewater treatment has been well upgraded in recent years; thus, the NH_3 emissions from wastewater treatment plants were quite small (i.e., 12 Gg in 2019).[41] In particular, new technologies such as the anaerobic-anoxic-oxic method and the Sequencing Batch Reactor have been widely used in China, resulting in higher removal efficiency of ammonium nitrates.[41] For solid waste treatment, the incineration process has been upgraded, whereas landfill and composting technologies remain undeveloped,[52,53] causing large NH_3 emissions from solid waste treatment (96 Gg in 2019).

For industries with combustion processes involved, such as iron and steel production, CFPPs, and cement production, APCDs are commonly performed to reduce air pollutants, such as NO_x and SO_2.[54,55] In this case, not only the production amount but also the technology and application rate of APCDs influence the NH_3 emissions. On the one hand, the APCDs reduce the NH_3 emissions by physically withholding ammonium particles (e.g., desulfurization devices used in CFPPs). On the other hand, APCDs increase the emissions when NH_3 or nitrogen-related chemicals are used as absorbents and end up emitted during the industrial processes. For example, the large-scale applications of non-selective catalytic reduction (SNCR) and selective catalytic reduction (SCR) technologies in cement production and iron and steel production are aimed at reducing NO_x emissions, where NH_3 is a critical absorbent in the processes, which could cause “slip out” of NH_3.[54,56,57] This issue is discussed in detail in the next section.

Some industry categories are not covered in our unit-based NH_3 emission data set because point source-level activity data and/or EFs of these industries are not available. We estimated the NH_3 emissions from the geothermal industry and beet sugar production in China. For the geothermal industry, the capacity of geothermal power plants in China was 45 MW in 2019, taking up 0.28% of the global capacity.[58] With the EF of NH_3 emissions (6.8 kg NH_3/MWh) based on the measurements from Bravi et al.,[59] we estimated that NH_3 emissions from the geothermal industry in China in 2019 were approximately 1 Gg. According to the China Sugar Association, the national production of beet sugar was 1237 Gg in China in 2019. With the emission factor (0.0026 kg NH_3/Mg) from Battye et al.,[35] we estimated the national NH_3 emissions to be 0.003 Gg, much lower than the emissions from other industries. Other industries, namely, nickel processing, explosive production, froth flotation, mineral wool production, and the pulp and paper industry, have been verified to be sources of NH_3 emissions to the atmosphere.[34,35] For example, NH_3 emissions from nickel processing and explosive production have been observed in other countries using satellite imaging.[23,34] However, measured EFs of these five industries are lacking. Although NH_3 emissions from these industries should be much less than that from fertilizer production,[35] it would be desirable to include these industries when their point source-level activity data and EFs are available.

3.2. Rapid Increase in Industrial NH_3 Emissions. Our new data set further draws attention to the rapid increase in industrial NH_3 emissions in China, which experienced a 3.4-fold increase from 1998 to 2019 (Figure 2a). The emission trend has not yet experienced a turning point, while the emissions of many other major air pollutants have been decreasing since 2010 due to the “clean air actions” in China.[42] The temporal trend of industrial NH_3 emissions could be divided into three periods: a slow increase period (1998–2003), a rapid increase period (2003–2014), and a steady period (2014–2019), with average annual increase rates of 2.3%, 8.6%, and 0.4%, respectively. During the rapid increase period, a slight fluctuation from 2008 to 2010 was identified

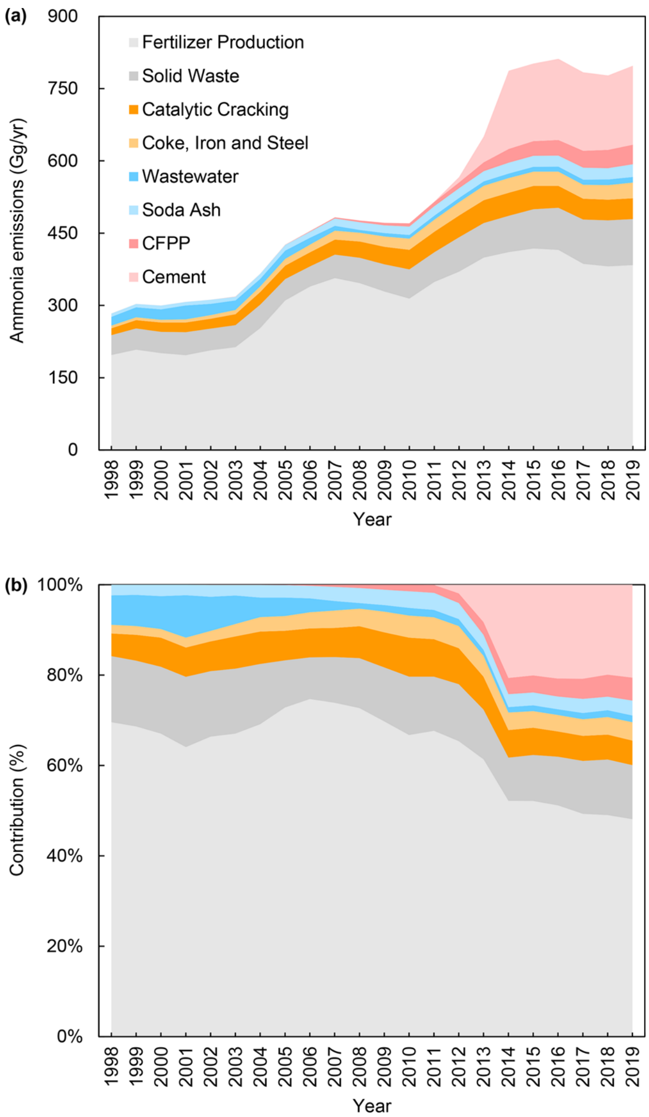

Figure 2. The temporal trends of industrial ammonia emissions in China. (a) Temporal trends of industrial NH_3 emissions from 1998 to 2019. (b) Temporal trends of compositions of industrial NH_3 emissions. The detailed emission curve of each province from 1998–2019 is given in SI Figure S3. Changes of the contribution of each province are shown in SI Figure S4.

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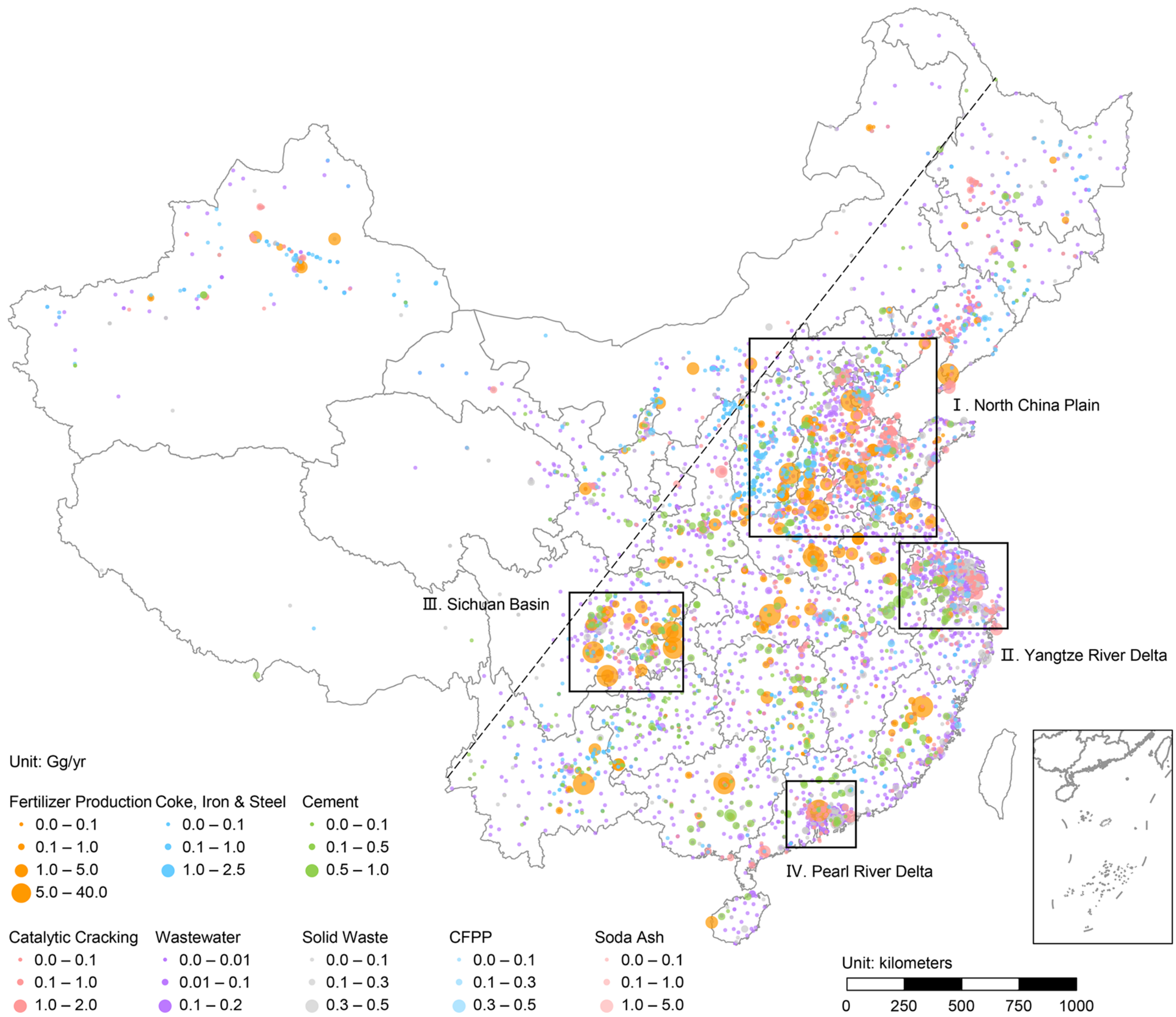

Figure 3. Unit-based mapping of industrial ammonia emissions in China in 2013. The area of points represents the amount of NH_3 emission of each point (unit: Gg/yr). Areas squared out above are emission hotspots. Detailed displays of the four hotspots are shown in SI Figure S5. SI Figures S7–S11 show the detailed data regarding fertilizer production, catalytic cracking, coke, iron and steel production, wastewater treatment, and soda ash production in 1998, 2003, 2008, and 2013. CFFP: coal-fired power plant.

that could be attributed to the decrease in production in the financial crisis.[60]

Significant differences were identified in the temporal trends of emissions among the industrial sectors. Fertilizer production dominated the NH_3 emissions, accounting for 69.6% of the total contribution in 1998. This ratio dropped to 48.1% in 2019, which was still large, while fertilizer production increased from 197 Gg to 384 Gg during this period. This is related to the industrial restructuring in the 10th Five-Year-Plan, when nitrogen fertilizer production was recognized as a key industry to promote, and large numbers of small fertilizer production factories were forced to merge into larger ones (SI Figure S7). The emissions from solid waste processing and CFPPs have been steadily increasing over the past two decades. For solid waste processing, the NH_3 emissions increased from 41 Gg to 96 Gg during this period (Figure 2a). For CFPPs, the NH_3 emissions rapidly increased from 0.01 Gg to 41 Gg during this period (Figure 2a), while the contribution of CFPPs to the national emissions reached 5.1% in 2019, 2,700-fold higher than that in 1998 (Figure 2b), mostly driven by the NH_3 "slip out" from the application of SCR and the increase in coal use.[61,62] Emissions from coke, iron and steel production increased rapidly before 2014 (average increase rate: 10.8%/yr), and recently, the trend has been stabilizing (1.2%/yr), mainly due to the overproduction of iron and steel that began in 2014, when the domestic demand for coke, iron and steel began to decrease.[63] Emissions from catalytic cracking and soda ash production share similar trends with iron and steel production (Figure 2a). Contrary to other industries, emissions from wastewater treatment show a declining trend, that is, from 18 to 12 Gg in the past two decades (Figure 2a), primarily due to the gradual upgrade of new technologies.[41]

Interestingly, our data set identified a sudden increase in the NH_3 emissions from cement production from 2013 (54 Gg) to 2014 (162 Gg, Figure 2a). This unexpected increase is related to the application of SNCR technology, as a result of stricter NO_x emission standards implemented in 2013 (ref[6]). Our results support a recent preliminary identification of the "NH_3 escape" issue, and further help clarify the details of this finding.[32] We found that the application ratios of SNCR technology in cement production were 7.9% and 31.5% in 2012 and 2013, respectively, and the ratio jumped to 92.9% in

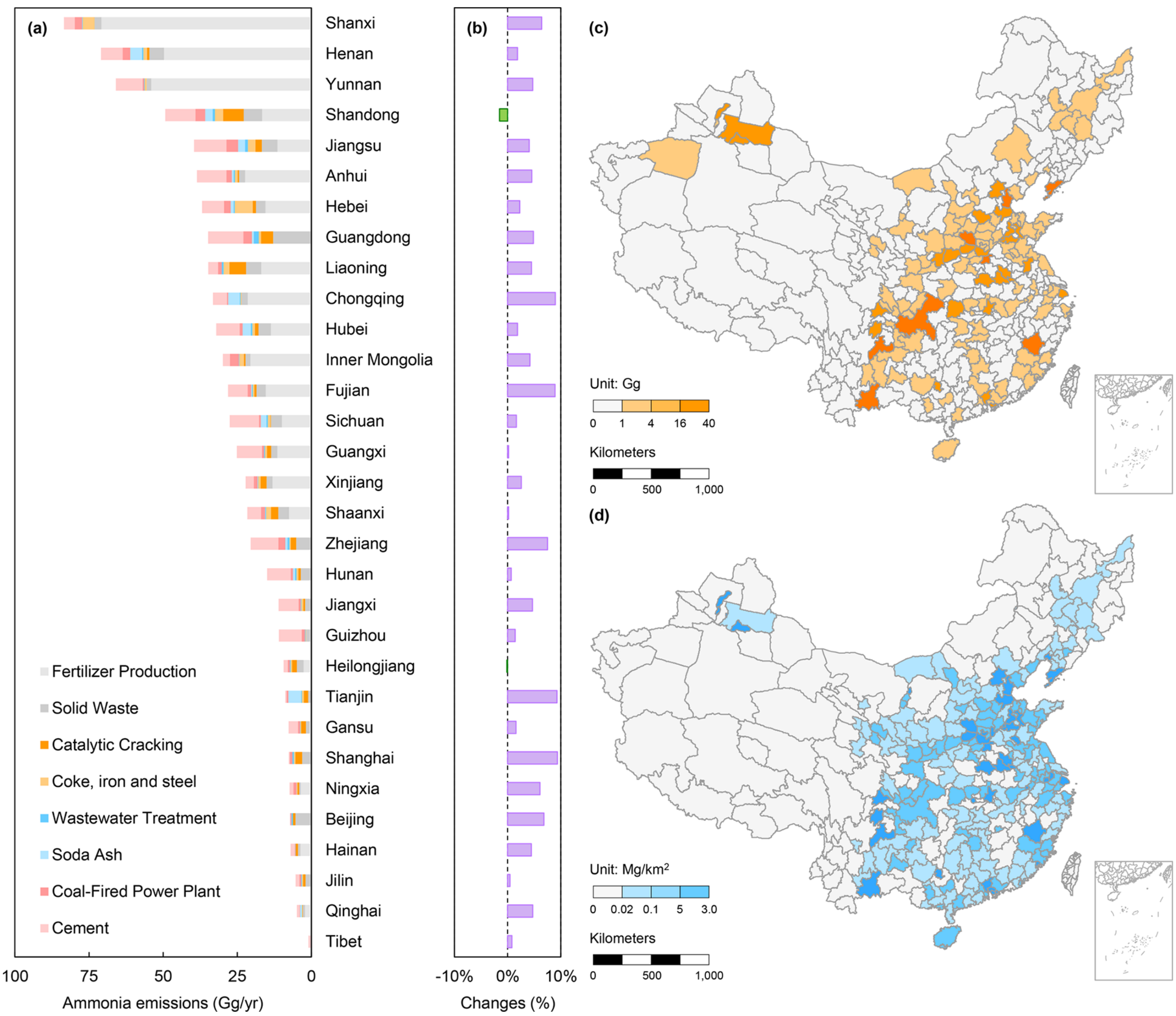

Figure 4. Provincial and city-level industrial ammonia emissions in China. (a) The composition of provincial industrial NH_3 emissions in 2019. (b) The changes of the contribution of industrial NH_3 emissions to the total NH_3 emissions from 2008 to 2017 in each province. (c) The city-level industrial NH_3 emissions in China in 2013. (d) The intensities of city-level industrial NH_3 emissions (flux divided by area) in China in 2013.

2014, showing the dramatic impacts of SNCR application and NO_x emission standards on industrial NH_3 emissions.[42] The application of the denitration process in other industries (e.g., CFPPs and iron and steel production) took place even earlier (e.g., 2006−2007) and increased rapidly from 2010−2014. Although the increases of industrial NH_3 emissions from these industries were not as dramatic as cement production, the changes in the application ratio (e.g., 10.9% and 100% in 2010 and 2014, respectively, for the denitration process in CFPP) as well as the "slip out" of NH_3 need to be given serious consideration. Overall, the large-scale application of SCR and SNCR in combustion-related industries is the result of balancing priorities. That is, the impact of NO_x on air quality is more significant than the NH_3 issue, while the "NH_3 escape" is either neglected or underestimated due to insufficient knowledge of industrial NH_3's impacts.

3.3. Spatial Characteristics of Industrial NH_3 Emissions. Our data set identifies four industrial NH_3 emission hot spots in China, namely, the North China Plain, the Yangtze River Delta (YRD), the Sichuan Basin, and the Pearl River Delta (PRD, Figure 3). These four regions constitute only 10% of the land area of China but account for ∼60% of the total industrial emissions. The North China Plain, renowned for its developed economy, large population, and industrial production, mainly includes Beijing, Tianjin, Hebei, Shanxi, Shandong, and Henan provinces. The industrial NH_3 emissions of the North China Plain were 257 Gg in 2019, accounting for 32% of the national emissions, strongly indicating the significance of air pollution control in this region. YRD and PRD are the two most developed regions in China, with the top-ranking economies, with YRD covering Jiangsu, Zhejiang, Shanghai, Anhui provinces, and PRD being located in Guangdong province. The industrial NH_3 emissions from YRD and PRD were 106 Gg and 35 Gg in 2019, accounting for 13.3% and 4.4% of the national emissions, respectively. The contributions are still considerable when the population density is considered. Similar to many other major air pollutants,[64] the Sichuan Basin is a newly identified hotspot due to its recent industrial development, and its emissions reached 61 Gg in 2019. Despite the high density of point sources in the Sichuan Basin, their contribution to national emissions is rather small, primarily because they are mostly small-scale plants. Overall, the densities of industrial NH_3 emissions (flux divided by area) of these four regions reached

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0.35−0.84 Mg/km^2, higher than the national average by factors of three to nine.

We identified that the top five largest provinces in terms of industrial NH_3 emissions accounted for nearly half (38.8%) of the national emissions in 2019 (Figure 4a). Shanxi province alone accounted for 10.5% (83 Gg, range 70−98 Gg) of the national emissions, followed by Henan (8.9%) and Yunnan (8.3%) provinces (Figure 4a), primarily due to more industries in these provinces. Shanxi and Henan are both located in the North China Plain and share similarities in their large investments in manufacturing industries. High concentrations of fertilizer production and iron and steel industry are identified in Shanxi and Henan, respectively. Point sources with high NH_3 emissions are mostly fertilizer production factories, and their emissions are usually significantly higher than those of other industries (Figure 4a), while coke, iron, and steel production are among the typical energy-intensive industries that produce NH_3 due to the APCDs used for the treatment of waste gas.[63] Like Henan, Yunnan is one of the largest fertilizer production provinces, producing 305.2 tons of nitrogen fertilizer in 2019 (Figure 4a).

Our data set can be used as guidance for controls of city-level industrial NH_3 emissions and health risk assessments. The emission intensities of industrial NH_3 among different provinces show similar trends with their emission amounts (SI Figure S6). However, many densely populated megacities, such as Shenzhen and Shanghai (17.6 and 21.5 million population in 2020, respectively), show markedly higher emission intensities than other cities (Figures 4c,d), further supporting the importance of industrial NH_3 in these megacities.[20,31,65,66] For example, the densities of industrial NH_3 emissions of Shenzhen, Shanghai, and Beijing reached 2.9, 0.8, and 0.7 t/km^2, respectively, 11- to 47-fold higher than the national average (0.06 t/km^2), calling for more attention to the severe health risks related with industrial NH_3 emissions in highly industrialized and developed cities.[28,31]

The contribution percentages of industrial NH_3 emissions in all sectors increased significantly at the provincial level (SI Figure S4). Almost all provinces have experienced increases to some extent from 2008 to 2017, except for Shandong and Heilongjiang (Figure 4b). Changes in the composition of total NH_3 emissions in each province were largely influenced by industrial restructuring and reallocation. In provinces like Fujian and Chongqing, the increases of contribution percentages were remarkable (from 3.4% to 12.4% and from 3.6% to 12.5%, respectively), indicating the possibility of the contribution shares from industry likely exceeding 20% in the next decade, while the contribution shares from the agricultural sector may decrease. Sharp increases in these two provinces were identified in 2010−2013, while the fertilizer production factories were relocated, and the production of urea and ammonia nitrate boomed in these two provinces.[67] Contribution shares in provinces with high industrial NH_3 emissions also increased rapidly (e.g., from 6.3% to 11.0% in Yunnan and from 28.9% to 35.3% in Shanxi). In Tianjin, Shanghai, and Beijing, the increases of contribution shares were 9.3%, 9.4%, and 6.8%, respectively. Considering that these provinces (or megacities) are economically developed with extraordinary population densities, a more severe threat to public health may occur in the future. Significantly different from other provinces, solid waste processing takes the lead in NH_3 emissions in Beijing and Shanghai. With the long-term economic restructuring in the two cities, heavy industries have been gradually moved out, resulting in significant contribution shares of solid waste (74% and 41% in Beijing and Shanghai, respectively).

3.4. Comparison with Previous Work and Uncertainty Analysis. Our estimate for the industrial NH_3 emissions in China is within the range of previous estimates (179−863 Gg/yr, Figure 5a). For example, our estimate is approximately 3-fold higher than that of the Emissions Database for Global Atmospheric Research (EDGAR) because our estimate captures the NH_3 emissions from combustion-related industries and the "slip out" of NH_3. This also explains the higher rate of increase after 2012 in this study. Our results are 10−50% lower than the spatially explicit estimate of PKU-Fuel since PKU-Fuel focuses more on fuels, and large uncertainties might exist in the estimates of industries showing non-negligible significance are considered (e.g., cement production).[15,28] Additionally, our estimate is coincidentally close to

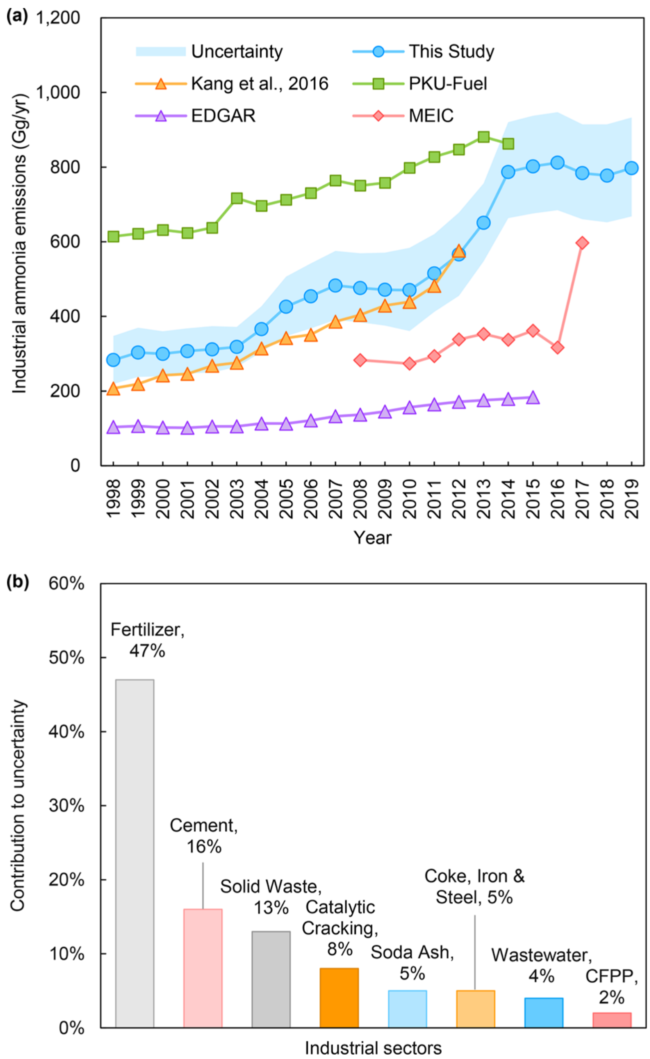

Figure 5. Comparison between different data sets and uncertainty. (a) Temporal comparison between this study and previous estimates. Data from Kang et al.,[25] Multiresolution Emission Inventory for China (MEIC),[42,60] PKUFuel-inventory,[28] EDGAR-inventory,[15] and this study. The shaded area represents the uncertainty range in the present study. (b) Each sector's contribution to the total uncertainty in the present study. The detailed uncertainty range of each year is given in SI Table S5.

that of Kang et al.,[25] as they did not consider NH_3 escape, while emissions from waste treatment are lower in this study due to the use of updated EFs, which are derived from the newest investigations in China.[41]

The overall uncertainty of this study ranges from −17% to 18% for 2019 (Figure 5a). Our point source data can reduce the CVs of ADs and mitigate the uncertainty. Fertilizer production takes up 47% of the total uncertainty, followed by cement production (16%) and solid waste (13%, Figure 5b), mainly due to the uncertainties in EFs of these industries. The contribution of CFPPs to the total uncertainty is relatively small (2%), partly due to the high accuracy in EFs, with a substantial number of studies having already thoroughly investigated its production process.[39] Among different provinces, industrial NH_3 emissions in Shanxi (70−97 Gg) and Yunnan (55−76 Gg) show higher uncertainty, where industries, especially fertilizer production, are densely located. Finally, comparisons show that our unit-based estimate may provide a more solid basis for model simulation, risk assessment, and policy-making.

3.5. Implications for NH_3 Emission Control. Previous studies have greatly improved our understanding regarding industrial NH_3 emission as a significant source to the atmosphere and the impact of emissions on human health.[22,23,29,31,32] Our efforts could advance these understandings through the development of a high-spatial-resolution data set of China's industrial NH_3 emissions with plant-level data. Despite the fact that the amount of industrial NH_3 emissions is relatively small compared to conventionally accepted major contributors (e.g., agriculture sources),[11,25,28] the industrial plants are usually located in urban areas, where ∼60% of China's population lives, while farmlands are generally distant from dense populations. The impact of industrial NH_3 emissions is further stressed with our plant-level emission data. For example, we identified that fertilizer production factories are merging into larger emitters and are more concentrated (SI Figure S7). Similar circumstances exist in other industrial sectors, such as catalytic cracking and CFPPs (SI Figures S8 and S9). Thus, the health risk potential in surrounding urban areas needs thorough attention. Together with previous findings,[5,31,66] we suggest that industrial NH_3 emissions may pose a greater impact on air pollution, such as $PM_{2.5}$ and related health risks.

Currently, industrial NH_3 emissions are in a critical period, and relevant policies are urgently needed. First, the emissions are increasing rapidly, playing a more important role in the increase of the total NH_3 emissions, especially with uncertainties in agricultural NH_3 emissions calculations.[24,26,68] Second, the current emission trend is fluctuating, and the future situation is largely dependent on what policy is adopted. Assuming a more restricted policy is carried out in the next decade, we speculate that the emission flux will decrease to approximately 700 Gg/yr in the next decade, indicating the importance of a strict NH_3 emission regulation. Otherwise, the NH_3 emissions from industrial sectors can be fast-increasing, which might increase to >1000 Gg/yr if a less restricted industrial NH_3 control policy is put into action in the next 5−10 years. Our efforts, together with a recently implemented, more accurate NH_3 monitoring system,[20,66] stress the importance and potential risks of industrial NH_3 emissions and call for more restricted and comprehensive policies. In addition, other developing countries in South Asia, Southeast Asia, South America, and other regions can gain some insights from this study. They could try to control industrial NO_x and NH_3 emissions at the same pace with strict regulations to prevent unexpected air pollution problems and health risks.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c08369.

Information on China's eight industries (fertilizer production, soda ash production, catalytic cracking, iron and steel production, cement production, solid waste treatment, wastewater treatment, and coal-fired power plants) from 1998−2019; data from previous studies on emission factors of industrial ammonia emissions; data on provincial industrial ammonia emissions from 1998−2019 in China; additional figures and tables about ammonia emissions from each industry; comparison with previous studies; and validation and uncertainty analysis of the mapping (PDF)

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Notes

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