Ground Ammonia Concentrations over China Derived from Satellite and Atmospheric Transport Modeling

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Abstract: As a primary basic gas in the atmosphere, atmospheric ammonia (NH₃) plays an important role in determining air quality, environmental degradation, and climate change. However, the limited ground observation currently presents a barrier to estimating ground NH₃ concentrations on a regional scale, thus preventing a full understanding of the atmospheric processes in which this trace gas is involved. This study estimated the ground NH₃ concentrations over China, combining the Infrared Atmospheric Sounding Interferometer (IASI) satellite NH₃ columns and NH₃ profiles from an atmospheric chemistry transport model (CTM). The estimated ground NH₃ concentrations showed agreement with the variability in annual ground NH₃ measurements from the Chinese Nationwide Nitrogen Deposition Monitoring Network (NNDMN). Great spatial heterogeneity of ground NH₃ concentrations was found across China, and high ground NH₃ concentrations were found in Northern China, Southeastern China, and some areas in Xinjiang Province. The maximum ground NH₃ concentrations over China occurred in summer, followed by spring, autumn, and winter seasons, which were in agreement with the seasonal patterns of NH₃ emissions in China. This study suggested that a combination of NH₃ profiles from CTMs and NH₃ columns from satellite obtained reliable ground NH₃ concentrations over China.

Keywords: NH₃; satellite; CTM; spatial; ground

1. Introduction

Ammonia (NH₃) is the primary form of reactive nitrogen (Nr) in the environment and a key component of the ecosystems, representing more than half of atmospheric Nr emissions [1,2]. NH₃ emissions have been increasing in recent years due to the increasing agricultural livestock numbers and the increasing application of Nr fertilization [2,3], resulting in the high NH₃ concentrations in the atmosphere. NH₃ increase has enhanced the acidification and eutrophication of the ecosystems on local and international scales [2,4]. Previous studies have shown that the lifetime of NH₃ is very short from hours to several days [5,6] converting to particulate matter (PM) as well as leading to dry and wet depositions. NH₃ reacts with acid-forming compounds such as sulfur...
dioxide (SO$_2$) and nitrogen oxides (NO$_x$) to form particles containing ammonium sulfate ((NH$_4$)$_2$SO$_4$) and ammonium nitrate (NH$_4$NO$_3$) in the atmosphere [7]. These processes increase the amount of atmospheric particulate matter, particularly for particles smaller than 2.5 micrometers in diameter (PM$_{2.5}$), thereby reducing visibility and negatively affecting environmental and human health [8,9]. Therefore, monitoring the ground NH$_3$ concentrations on a regional scale is vitally important to assist in enacting effective measures to protect the eco-environments and public health, with respect to air, soil, and water quality.

Progress in the understanding of the NH$_3$ cycling process, flux measurements, and instrumentation have allowed advances in estimating NH$_3$ concentrations in the atmosphere on a local or regional scale, based on the simulation of the chemical transport models (CTM). For example, a coupled MM5-CMAQ modeling system was used for computing the ground NH$_3$ concentration based on the NH$_3$ emission developed with a spatial resolution of 27 km $\times$ 27 km in the Beijing–Tianjin–Hebei (BTH) region of China [10]. The simulation error of ground NH$_3$ concentration in different seasons in BTH range from $-24.4\%$ to 7.8\%, indicating the ground NH$_3$ concentrations simulated by MM5-CMAQ are comparable with the observations; A GEOS-Chem model was used to estimate the global and seasonal NH$_3$ with a resolution of 2$^\circ$ latitude $\times$ 2.5$^\circ$ longitude [11], showing that the simulated ground NH$_3$ concentrations are biased low compared to the Tropospheric Emission Spectrometer (TES) with seasonal mean differences of $-0.92$ to 1.58 ppb. Similar reports on estimating ground NH$_3$ concentrations from CMT could also be tracked in several studies [12–14]. Although these CTMs could simulate the profiles of NH$_3$ concentrations in the atmosphere, the ground NH$_3$ concentrations over a large scale, such as on a national scale over the entire area of China, are still poorly understood due to the large pixel sizes and the relatively high uncertainties resulting from errors of the emission data and the simplification of the chemistry schemes. Fortunately, numerous studies have shown that CTMs can produce profiles for aerosol [15–18], NO$_2$ [19–21], NH$_3$ [2,22–24], and SO$_2$ [19,25], denoting that the vertical profiles of the NH$_3$ concentrations from CTM were highly beneficial in calculating the ground NH$_3$ concentrations.

In comparison with CTM simulations, satellite remote sensing is considered as an observational perspective and offers another way to obtain large-scale NH$_3$ columns with high spatial resolutions, based on advanced infrared spectroscopy (IR) sounders, such as the Infrared Atmospheric Sounding Interferometer (IASI), the Tropospheric Emission Spectrometer (TES), and the Cross-track Infrared Sounder (CrIS) [26,27]. Large-scale distributions of IASI NH$_3$ columns could denote the status of NH$_3$ levels in regions not covered by ground measurement networks, expanding insight into new NH$_3$ sources including industry, agriculture, and biomass burning [2,22]. However, satellite NH$_3$ can only provide the columns and has no information of the vertical distributions of the columns (from the ground to the top of the atmosphere), presenting a barrier in obtaining the ground NH$_3$ concentrations. Fortunately, as mentioned in the last paragraph, the detailed NH$_3$ profiles could be obtained from CTMs. Combining the advantages of CTMs (NH$_3$ profiles) and satellite observations (large-scale overages with high spatiotemporal resolutions), the ground NH$_3$ concentrations can be derived.

We aimed to generate spatiotemporal ground NH$_3$ concentrations with the aid of the remotely sensed NH$_3$ columns and vertical NH$_3$ profiles from a CTM. The estimated ground NH$_3$ concentrations were further compared with the national ground monitoring network of the Chinese Nationwide Nitrogen Deposition Monitoring Network (NNDMN). Our purpose is not to replace traditional algorithms, but to combine the advantages of satellite with high spatial and temporal resolutions, and CTMs with detailed NH$_3$ vertical profiles in order to obtain high spatiotemporal ground NH$_3$ concentrations over China, hence providing basic information for the ground status of NH$_3$ concentrations and guiding the monitoring plans in the future over China.
2. Materials and Methods

2.1. Ground NH\textsubscript{3} Concentrations in the Atmosphere

Monitoring ground-based NH\textsubscript{3} concentrations on a regional scale is not straightforward due to the technical limitations and great variability of the concentrations in time and space [28]. While the availability of NH\textsubscript{3} concentration data and the flux measurements on local scales is increasing, the measurements on a regional scale are sparser [1].

We used the monthly ground NH\textsubscript{3} concentrations from the Chinese Nationwide Nitrogen Deposition Monitoring Network (NNDMN, made available on request by Prof. X.J. Liu, China Agricultural University) to evaluate the accuracy of the satellite-derived ground NH\textsubscript{3} concentrations. Monthly NH\textsubscript{3} concentrations (in units of µg N m\textsuperscript{-3}) were measured at 44 sites from 2010 to 2013 (Figure 1). The network mainly covered farmland sites but also included some grassland (two) and forest (four) sites across China [29,30]. The ground NH\textsubscript{3} concentrations in NNDMN were monitored using both DEnuder for Long-Term Atmospheric (DELTA) systems as well as Adapted Low-cost, Passive High Absorption (ALPHA) samplers [30,31]. ALPHA is a passive sampling system, while DELTA is an active sampling system. Monthly ground NH\textsubscript{3} concentrations were mostly monitored by DELTA, and few monitoring sites were measured by ALPHA. Xu et al. [30] showed that these two methods on measuring ground NH\textsubscript{3} concentrations were not significantly different and can be considered consistent.

![Figure 1. Spatial distribution of ground monitoring NH\textsubscript{3} sites in the Chinese Nationwide Nitrogen Deposition Monitoring Network (NNDMN).](image)

2.2. IASI NH\textsubscript{3} Columns

The IASI instrument is on board the polar sun-synchronous MetOp platform, which crosses the equator at a mean local solar time of 9.30 a.m. and p.m. [32]. In this study, we used the measurements from the morning overpass as they are generally more sensitive to NH\textsubscript{3} because of higher thermal contrast at this time of day [1]. IASI has an elliptical footprint of 12 km by 12 km (at nadir) and up to 20 km by 39 km (off nadir), depending on the satellite viewing angle. The availability of measurements is mainly dependent on the cloud coverage.
The current method is based on the calculation of a spectral hyperspectral range index and subsequent conversion to a NH₃ total column using a neural network. Details on the retrieval algorithms can be found in Whitburn et al. [32]. We requested the IASI NH₃ data from Université Libre De Bruxelles, and processed the daily observation data to monthly average data for deriving the ground NH₃. In the present work, the observations with a cloud coverage lower than 25%, and relative error lower than 100% or absolute error less than 5 × 15 molec. cm⁻² were processed [27].

2.3. NH₃ Profiles from MOZART-4

MOZART-4 (Model for Ozone and Related chemical Tracers, version 4) is a three-dimensional (3-D) global chemical transport model simulating the chemical and transport processes, which can be driven by essentially any meteorological dataset and with any emissions inventory [24,33]. The MOZART-4 used in this study includes detailed chemistry, an improved scheme for the determination of albedo, aerosols, online calculations of photolysis rates, dry deposition, H₂O concentration, and biogenic emissions. A comprehensive tropospheric chemistry with 85 gas-phase species, 12 bulk aerosol species, 39 photolyses, and 157 gas-phase reactions has been included in MOZART-4 [24]. The chemical initial and boundary conditions, spatially and temporally varying (6 h), are constrained by global chemical transport simulations from MOZART-4/GEOS-5 (Goddard Earth Observing System-5) with 1.9° latitude × 2.5° longitude horizontal resolution and 56 vertical levels from the surface. Details on the meteorological data and emission inventory used for driving MOZART-4 as well as related configurations can be tracked in Emmons et al. [24]. We requested the MOZART output data from NCAR (National Center for Atmospheric Research, Boulder, CO, USA). The output data are varying 6 h (daily). We calculated the monthly data by averaging the daily data, and then used the monthly data for analysis.

2.4. Satellite Derived Ground NH₃ Measurements

The fundamental thoughts of the methodology in this work were demonstrated in previous studies for aerosol [15–17], NO₂ [19–21] and SO₂ [19,25]. The recent progress in satellite NH₃ measurements also made this methodology applicable in estimating the ground NH₃ concentrations by combining the NH₃ profiles from CTM and NH₃ columns.

We had three major steps to estimate the satellite-derived ground NH₃ concentrations (Figure 2). First, we produced continuous monthly IASI NH₃ columns according to the method in previous studies [27,32]. Second, we simulated the vertical profiles from MOZART-4, and calculated the ratio of ground NH₃ to NH₃ columns. Third, we derived the satellite-derived ground NH₃ concentrations combining the IASI NH₃ columns and the ratio in the second step. Of these three steps, the second step of simulating the vertical profiles was the most important and complex one. We demonstrate here the key algorithms to simulate the vertical profiles from MOZART.

Figure 2. Schematic of the method to estimate the satellite-derived ground NH₃ concentrations.
We retrieved the NH$_3$ profiles from MOZART to convert the IASI NH$_3$ columns to ground NH$_3$ concentrations. The NH$_3$ vertical profile function was simulated by the following equation in the grid cell using the output data from MOZART-4:

$$f(h) = \sum_{i=1}^{n} a_i e^{-\frac{(h-b_i)^2}{2c_i^2}}$$

where $n$ ranges from 2 to 6, representing the number of Gaussian items; $a_i$, $b_i$, and $c_i$ indicate the constants for each Gaussian item; $h$ indicates the vertical height from the ground and $f(h)$ is the NH$_3$ concentration at height $h$. Theoretically, we can use $n$ larger than 6 (with more Gaussian items). However, it is highly dependent on the computational time cost and computer memory limitations.

We simulated the NH$_3$ vertical profile using Equation (1) by each grid cell, based on the 56 vertical layers of NH$_3$ concentrations from MOZART. For each grid cell, we had five models ($n = 2, 3, 4, 5, 6$) and used $R^2$ and root-mean-square error (RMSE) to assess each model performance. We selected the best one with highest $R^2$ and lowest RMSE (i.e., determined the value of $n$).

The MOZART NH$_3$ columns can be gained by integration based on the simulated profile function:

$$F(h_{trop}) = \int_0^{h_{trop}} f(h) dh$$

where $F(h_{trop})$ denotes NH$_3$ columns and $h_{trop}$ indicates the tropospheric height.

The satellite-derived ground NH$_3$ concentration is calculated as:

$$[SNH_3]_G = [SNH_3]_{Trop} \times \frac{f(h_G)}{F(h_{trop})}$$

where $[SNH_3]_{Trop}$ indicates the IASI NH$_3$ columns, $f(h_G)$ denotes the ground NH$_3$ concentration from MOZART, and $F(h_{trop})$ represents the MOZART NH$_3$ columns.

We used the national ground-based NH$_3$ concentrations in NNDMN between 2010–2013 to validate the satellite-derived ground NH$_3$ concentrations. We applied the correlation coefficient ($r$) and relative error ((observation-estimation)/observation) at each monitoring site to assess the accuracy of the satellite-derived ground NH$_3$ concentrations.

3. Results and Discussion

3.1. Accuracy Assessment of the Estimated Ground NH$_3$ Concentrations

To convert the IASI NH$_3$ columns to ground NH$_3$ concentrations, it is essential to obtain the vertical NH$_3$ profiles. We retrieved the vertical NH$_3$ profiles from MOZART in this study (as an example, the vertical NH$_3$ concentrations at five locations in January 2013 from MOZART are shown in Figure A1). The NH$_3$ profiles were simulated by each grid cell in China (Figure A9) with determination of coefficients ($R^2$) larger than 0.95 accounting for 99.81% of all grid cells (Table A1 and Figure A9). Then, we estimated the ground NH$_3$ concentrations based on IASI NH$_3$ columns and the modeling MOZART NH$_3$ profiles.

We used 44 ground-based sites from NNDMN between 2010–2013 to assess the performance of the estimated monthly ground NH$_3$ concentrations. The correlation between the estimated and measured at each site is given in Table A2 in Appendix A, and the relative bias of each site as well as the yearly comparisons between the estimated and measured ground NH$_3$ concentration are given in Figures 3 and 4. We found 90.91% of minoring sites has a relative error within $-30%$–$50\%$, showing an agreement between the estimated and measured. The seasonal absolute error by inverse-distance-weighted (IDW) interpolation is also shown in Figure A2. We found the absolute error in winter (December, January, and February) was higher than in other seasons, which can be explained
by the highest relative error in IASI NH$_3$ columns in the winter season (Figure A3). In addition, Figure 4 demonstrates a comparison between the estimated and measured ground NH$_3$ concentrations before and after applying the IASI NH$_3$ data. We found a relatively higher correlation (R, 0.81 vs. 0.57) and a better consistency (slope, 0.96 vs. 0.50) between the satellite-derived ground NH$_3$ concentrations and the measured ground NH$_3$ concentrations than those from MOZART not applying the IASI NH$_3$ data.

![Figure 3](image1.png)

**Figure 3.** Spatial distribution of the relative error (a), correlation (b) and root-mean-square error (RMSE) (c) of the estimated ground NH$_3$ concentration (µg N m$^{-3}$) at 44 NNDMN sites.

![Figure 4](image2.png)

**Figure 4.** Yearly comparisons between the estimated and measured ground NH$_3$ concentration (µg N m$^{-3}$). (a) indicates the comparison between the measured ground NH$_3$ concentrations and the estimated ground NH$_3$ concentrations from MOZART at the lowest layer before applying the satellite data, while (b) represents the comparison between the measured and estimated ground NH$_3$ concentrations by applying the satellite data using the methods in Section 2.4.
3.2. Spatial Pattern of the Ground NH₃ Concentrations

Spatial distribution of ground NH₃ concentrations in 2012 over China is shown in Figure 5a. High ground NH₃ concentrations greater than 10 µg N m⁻³ were concentrated in North China and South China including Beijing–Tianjin–Hebei (BTH), Shandong, Henan, Hubei, Anhui, Sichuan and Jiangsu provinces, forming the major regions of intensive agriculture over China. Low ground NH₃ concentrations are predominantly located in TP (Tibetan Plateau), where both the synthetic fertilizers and livestock waste were the least among 32 provinces [34,35]. The spatial ground NH₃ concentrations revealed considerable spatial heterogeneity across China and were in agreement with the percent farmland area (Figure 5a,b), reflecting its unique agricultural structure and farming practice.

High ground NH₃ concentrations were also observed in some areas in Xinjiang province (Figure 5a), where our estimation were about −30% to −10% underestimation compared with measurements in NNDMN (Figure 3). Moreover, relatively high NH₃ columns could be observed by satellite IASI instrument (Figure 5c). Synthetic N fertilizers and livestock waste both dominated the spatial distribution of the total emissions [34,35], hence determining the spatial patterns of the ground NH₃ concentrations. Previous studies reported that the NH₃ emissions from livestock exceeded those from the farmland in China, and NH₃ emissions from livestock accounted for about 54% of the total NH₃ emissions over China [35]. The contribution of livestock to the total NH₃ emissions in Xinjiang (where sheep are widely raised) accounted for higher than 60% [10,35]. Thus, due to the combining influence of both synthetic N fertilizers and livestock waste, the spatial distributions of ground NH₃
concentrations and percent farmland differed, especially in regions where the livestock dominated the NH₃ emissions. In addition, most of the ground NH₃ emissions were more concentrated on the ground and relatively hard to transport vertically compared with other regions in China, which can be clearly seen by the ratio of ground NH₃ concentrations to NH₃ columns from MOZART (Figure 5d).

3.3. Seasonal Variations of the Ground NH₃ Concentrations in China

To demonstrate the seasonal variations of the ground NH₃ concentrations in China, we calculated the monthly average values throughout China (Figure 6a). We found the maximum ground NH₃ concentrations over China occurred in summer (June, July, and August), followed by spring (March, April, and May), autumn (September, October, and November) and winter (December, January, and February) seasons. It is interesting that the seasonal ground NH₃ concentrations were in agreement with the seasonal patterns of NH₃ emissions in China conducted by Kang et al. [36], Huang et al. [35], and Xu et al. [37] (Figure 6b–d), indicating that the NH₃ emissions are the key factor influencing seasonal pattern of the ground NH₃ concentrations. The maximum NH₃ emissions in summer is reasonable due to more than 40% of the fertilization and more than 25% of livestock emissions occurring in summer [36,37]. In addition, high temperature in summer in China may also accelerate the NH₃ volatilization (NH₄⁺ → NH₃ + H⁺) from fertilizer, animal waste, city garbage or vehicles [6,38–40], and hence cause high ground NH₃ concentrations. In contrast, in winter, temperature frequently below freezing leads to reduced NH₃ volatilization and lower NH₃ concentrations than in other seasons.

![Figure 6](image_url)

**Figure 6.** Seasonal patterns of ground NH₃ concentrations in China. (a) indicates the monthly variations of ground NH₃ concentrations (µg N m⁻³) in China; (b) represents the monthly variations of the total NH₃ emissions (Tg, 10^{12} g) in China conducted by Kang et al. [36]; (c) shows the the monthly variations of the sum of fertilizer and livestock NH₃ emissions (Tg) in China conducted by Huang et al. [35] and (d) denotes the monthly variations of the fertilizer NH₃ emissions (Tg) in China conducted by Xu et al. [37].

To more accurately quantify the effects of meteorological parameters on the seasonal trends of the ground NH₃ concentrations, we selected the five best-simulated ground sites with n >30
(Table A2) for demonstrating meteorological parameters, such as temperature, wind speed, humidity, and precipitation on the seasonal variations of the ground NH$_3$ concentrations (Figures 7 and A4–A8). The monthly wind speed, temperature, relative humidity, and precipitation for each site were taken from the China Meteorological Administration. A positive correlation ($R = 0.6$, $p = 0.00$) was found between the ground NH$_3$ concentrations and temperature. An inverse relationship between the ground NH$_3$ concentrations and humidity (Figure 7), indicated that higher relative humidity may contribute to more NH$_3$ loss rates (NH$_3$ → NH$_4^+$). In addition, we also conducted a partial correlation analysis [41] regarding ground NH$_3$ concentrations, temperature, and humidity by considering their interactions using the function “partialcorr” in Matlab. We found the partial correlation between ground NH$_3$ concentrations and humidity was $-0.10$ ($p = 0.03$), showing a significant inverse relationship between the ground NH$_3$ concentrations and humidity. Significant effects of air humidity on NH$_3$ loss were also demonstrated previously [42,43]. However, precipitation and wind speed were not significantly correlated with ground NH$_3$ concentrations ($p = 0.632$, precipitation vs. NH$_3$; $p = 0.156$, wind speed vs. NH$_3$) as shown in Figures A4–A8.

![Figure 7](image)

**Figure 7.** The seasonal variations of ground NH$_3$ concentrations ($\mu$g N m$^{-3}$), temperature ($^\circ$C), precipitation (mm), humidity (%), and wind speed (m/s) at five sites with best-simulated ground NH$_3$ concentrations from January 2010 to December 2013 (0–12, 2010; 13–24, 2011; 25–36, 2012; 37–48, 2013). The relationship between the ground NH$_3$ concentrations and precipitation (mm), humidity (%), and wind speed (m/s) at each site is provided in Figures A4–A8.

### 3.4. Comparison with Previous Studies

The first relatively complete work on the national ground measurements of NH$_3$ concentrations in China is NNDMN, and the results of ground measurements were published by Xu et al. [30], which we considered as a truly comprehensive and valuable work on the national status of the ground NH$_3$ concentrations, and which shed some light on the actual status of ground NH$_3$ concentrations. The national measurements in NNDMN provide the best accurate datasets for validating the modeling ground NH$_3$ concentrations. In the previous studies, due to very limited ground measurements (not to mention the national monitoring measurements), it was difficult to validate the accuracy of the modeling ground NH$_3$ concentrations in China. The lack of measurements makes it necessary to assess the modeling ground NH$_3$ concentrations in China [44]. Recently, Zhao et al. [45] presented
a comprehensive work on the national-scale model validation of ground NH$_3$ concentrations with 1/2° longitude by 1/3° latitude horizontal resolution using the GEOS-Chem model, showing the correlation coefficient with NNDMN between 2011–2012 which was about 0.65 on the annual scale [45]. Compared with Zhao et al. [45], we used the same datasets from NNDMN while having a longer time period (2010–2013) to validate our estimated ground NH$_3$ concentrations, and found the correlation coefficient was about 0.81 (slope = 0.96 and intercept = 1.31) on the annual scale as shown in Figure 4, demonstrating better agreement with the ground measurements. The relatively higher accuracy in estimating ground NH$_3$ concentrations may result from different datasets used for estimation, where we used the satellite observation and Zhao et al. [45] used the NH$_3$ emission data used for modeling. Uncertainties existed in the estimation of NH$_3$ emission resulting from the methodology of calculation, which simplified the complexity of the real status of emission process [36]. For example, N-fertilizer NH$_3$ emission in BTH between different studies varied greatly as 256.5 Gg [35], 502.5 Gg [46], 432.7 Gg [10]; livestock NH$_3$ emission in BTH between different studies varied as 556.6 Gg [35], 675.2 Gg [46], and 891.6 Gg [10]. The estimation of NH$_3$ emissions by Zhou et al. [10] even nearly doubled that by Huang et al. [35] and Dong et al. [46]. The actual local emission factors in different regions differed from each other greatly, due to the difference of the local meteorological conditions, fertilizing time, and fertilizer kinds [37]. The NH$_3$ emissions are mainly based on statistical NH$_3$ emissions at a city or county level, and the accuracy is strongly dependent on both the limited spatial and temporal resolutions of the coarse statistical data [35–37,44,47].

The present study derived ground NH$_3$ concentrations from IASI NH$_3$ columns and the profiles from MOZART-4, implying that a combination of CTM modeling and satellite monitoring obtained a reliable ground NH$_3$ estimation over China. More generally, this attempt to generate the ground NH$_3$ measurements with a relative high resolution from IASI and MOZART has highlighted known limitations in the ground NH$_3$ monitoring measurements, which may in some cases not be representative of the estimated NH$_3$ concentrations horizontally and vertically. Here we highlight the need to acquire more comprehensive datasets of ground NH$_3$ concentrations, and dedicated measurement campaigns focusing on the ground NH$_3$ measurement will no doubt allow improvements in the validation of estimated NH$_3$ in the future. In addition, we focused on the spatial pattern of ground NH$_3$ concentrations derived from satellite and a CTM, which is based on the monthly average and may be limited for the specific analysis such as secondary aerosol formation, photochemistry, and consideration of regulation. It is also beneficial and even essential to gain higher temporal resolution of ground NH$_3$ concentrations in the future.

4. Conclusions

We critically estimated the ground NH$_3$ concentrations over China, combining IASI NH$_3$ columns and NH$_3$ profiles from MOZART. We aimed to generate ground NH$_3$ concentrations over China, and hence provide potential to understand both the spatial and temporal variations of ground NH$_3$ concentrations in order to guide future ground NH$_3$ monitoring plans. The intention was not to replace traditional algorithms but to provide new insight on the current status of ground NH$_3$ over China, and to generate more reliable ground NH$_3$ concentrations. The IASI NH$_3$ columns and NH$_3$ profiles from the atmospheric chemistry transport model are encouraged to be combined to generate ground NH$_3$ concentrations at local or regional scales, and the estimated results should be further improved.

This study introduced methods to estimate ground NH$_3$ concentrations over China using IASI NH$_3$ columns and NH$_3$ profiles. The estimated ground NH$_3$ concentrations were validated by 44 sites from NNDMN, showing promising results between the estimated and measured, and then the spatial and temporal variations of ground NH$_3$ concentrations were demonstrated. High ground NH$_3$ concentrations greater than 10 µg N m$^{-3}$ were mainly located in Beijing, Hebei, Shandong, Henan, Jiangsu, eastern Sichuan, and some regions in Xinjiang provinces, while low ground NH$_3$ concentrations were concentrated in the Tibet-Plateau area. The maximum ground NH$_3$ concentrations
over China occurred in summer, followed by spring, autumn, and winter seasons, which are in agreement with the seasonal patterns of NH$_3$ emissions in China.

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**Author Contributions:** L.L. and X.Z conceived the idea; L.L. and S.W. conducted the analyses; L.L. and S.W. processed the data; X.L. and W.X. provided the observation data for validation; X.Z, X.L., L.Z., and W.Z. contributed to the writing and revisions.

**Conflicts of Interest:** The authors declare no competing financial interest.

**Appendix A**

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**Figure A1.** Vertical NH$_3$ concentrations (µg N m$^{-3}$) simulated by Mozart at five locations in January 2013.

**Figure A2.** A quick illustration of the site bias of ground NH$_3$ concentrations across China by interpolating the residuals between the measured and estimated using the inverse-distance-weighted (IDW) interpolation. The figures were generated using ArcGIS 12.0 software (https://www.arcgis.com/).
Figure A3. Relative error (%) of IASI NH₃ columns. (a) indicates the annual IASI NH₃ error (with a cloud coverage lower than 25%) averaged from 2008 to 2015; (b) indicates the averaged monthly relative error from 2008 to 2015 in different regions (every dot indicates the relative error at a month in a region); (c) indicates the temporal variations of relative error over China at a monthly scale.

Figure A4. The seasonal variations of ground NH₃ concentrations (µg N m⁻³), temperature (°C), precipitation (mm), humidity (%), and wind speed (m/s) at GZL from January 2010 to December 2013 (0–12, 2010; 13–24, 2011; 25–36, 2012; 37–48, 2013).
Figure A5. The seasonal variations of ground NH$_3$ concentrations (µg N m$^{-3}$), temperature (°C), precipitation (mm), humidity (%), and wind speed (m/s) at TLF from January 2010 to December 2013 (0–12, 2010; 13–24, 2011; 25–36, 2012; 37–48, 2013).

Figure A6. The seasonal variations of ground NH$_3$ concentrations (µg N m$^{-3}$), temperature (°C), precipitation (mm), humidity (%), and wind speed (m/s) at CL from January 2010 to December 2013 (0–12, 2010; 13–24, 2011; 25–36, 2012; 37–48, 2013).
Figure A7. The seasonal variations of ground NH$_3$ concentrations (µg N m$^{-3}$), temperature (°C), precipitation (mm), humidity (%), and wind speed (m/s) at YPH from January 2010 to December 2013 (0–12, 2010; 13–24, 2011; 25–36, 2012; 37–48, 2013).

Figure A8. The seasonal variations of ground NH$_3$ concentrations (µg N m$^{-3}$), temperature (°C), precipitation (mm), humidity (%), and wind speed (m/s) at FYU from January 2010 to December 2013 (0–12, 2010; 13–24, 2011; 25–36, 2012; 37–48, 2013).
Figure A9. (a,b) $R^2$ and RMSE (molec./cm$^2$) for the Gaussian simulation of the NH$_3$ profiles (68–142°E, 5–55°N) in 2013.

Table A1. Descriptive statistics for results of Gaussian simulation.

| Season (%) | $N = 2$ | $N = 3$ | $N = 4$ | $N = 5$ | $N = 6$ | $R^2 > 0.95$ | $R^2 > 0.99$ |
|------------|---------|---------|---------|---------|---------|-------------|-------------|
| Spring     | 0.70    | 12.02   | 33.33   | 34.61   | 19.31   | 99.86       | 96.94       |
| Summer     | 0.79    | 10.47   | 28.24   | 37.09   | 23.38   | 99.86       | 97.52       |
| Autumn     | 0.48    | 7.60    | 24.58   | 37.93   | 29.39   | 99.86       | 98.89       |
| Winter     | 0.92    | 10.25   | 31.03   | 35.80   | 21.97   | 99.64       | 96.46       |
| All        | 0.72    | 10.09   | 29.29   | 36.36   | 23.51   | 99.81       | 97.45       |

Note: Spring includes March, April, and May; Summer includes June, July, and August; Autumn includes September, October, and November; Winter includes December, January, and February. $N$ indicates the numbers of the Gaussian items. For details, please refer to the methods part.
Table A2. Comparison between monthly IASI satellite-derived ground NH\textsubscript{3} concentrations and the NNDMN monitoring sites from 2010 to 2013.

| Site   | Landuse         | Long (°E) | Lat (°N) | n   | \( R (\pm \text{std}) \) |
|--------|-----------------|-----------|----------|-----|--------------------------|
| BYBLK  | Alpine grassland| 83.71     | 42.88    | 22  | 0.68 (0.05)              |
| FK     | Desert-oasis ecotone | 87.93   | 44.29    | 32  | 0.49 (0.04)              |
| TLF    | Desert in an oasis | 89.19   | 42.85    | 28  | 0.84 (0.07)              |
| SDS    | Urban           | 87.56     | 43.85    | 38  | 0.69 (0.06)              |
| TFS    | Suburban        | 87.47     | 43.94    | 35  | 0.56 (0.05)              |
| CL     | Desert-oasis ecotone | 80.73   | 37.02    | 12  | 0.94 (0.08)              |
| TZ     | Desert          | 83.66     | 38.97    | 12  | 0.89 (0.07)              |
| YPH    | Farmland        | 77.27     | 39       | 12  | 0.83 (0.05)              |
| HT     | Farmland        | 79.89     | 37.15    | 5   | 0.99 (0.08)              |
| AKS    | Farmland        | 80.83     | 40.62    | 17  | 0.72 (0.06)              |
| KRL    | Farmland        | 85.86     | 41.68    | 6   | 0.94 (0.08)              |
| NLT    | Forest          | 84.03     | 43.31    | 4   | 0.33 (0.03)              |
| NSXC   | Forest          | 87.04     | 43.35    | 7   | 0.98 (0.09)              |
| CAU    | Urban           | 116.28    | 40.02    | 45  | 0.57 (0.05)              |
| ZZ     | Urban           | 113.63    | 34.75    | 44  | 0.55 (0.04)              |
| SZ     | Farmland        | 116.2     | 40.11    | 45  | 0.86 (0.07)              |
| BD     | Farmland        | 115.48    | 38.85    | 12  | 0.44 (0.04)              |
| QZ     | Farmland        | 114.94    | 36.78    | 45  | 0.50 (0.04)              |
| YQ     | Farmland        | 112.89    | 38.05    | 45  | 0.57 (0.05)              |
| ZMD    | Farmland        | 114.05    | 33.02    | 45  | 0.27 (0.02)              |
| YL     | Farmland        | 108.01    | 34.31    | 45  | 0.27 (0.02)              |
|YC     | Farmland        | 116.63    | 36.94    | 35  | 0.77 (0.06)              |
| GZL    | Farmland        | 124.83    | 43.53    | 42  | 0.82 (0.06)              |
| LS     | Farmland        | 124.17    | 43.36    | 42  | 0.62 (0.05)              |
| DL     | Coastal         | 121.58    | 38.92    | 40  | 0.73 (0.05)              |
| WY     | Forest          | 129.25    | 48.11    | 12  | 0.31 (0.02)              |
| GH     | Forest          | 121.52    | 50.78    | 12  | 0.38 (0.03)              |
| WW     | Farmland        | 102.6     | 38.07    | 39  | 0.32 (0.02)              |
| DL     | Grassland       | 116.49    | 42.2     | 6   | 0.52 (0.04)              |
| WX     | Farmland        | 115.79    | 30.01    | 29  | 0.56 (0.05)              |
| BY     | Farmland        | 113.27    | 23.16    | 44  | 0.47 (0.04)              |
| TJ     | Farmland        | 111.97    | 28.61    | 39  | 0.42 (0.03)              |
| FYU    | Farmland        | 113.34    | 28.56    | 40  | 0.76 (0.06)              |
| HN     | Farmland        | 113.41    | 28.52    | 40  | 0.36 (0.03)              |
| NJ     | Farmland        | 118.85    | 31.84    | 18  | 0.82 (0.06)              |
| FY     | Farmland        | 117.56    | 32.88    | 11  | 0.79 (0.06)              |
| ZJ     | Coastal         | 110.33    | 21.26    | 41  | 0.63 (0.05)              |
| FZ     | Coastal         | 119.36    | 26.17    | 45  | 0.49 (0.03)              |
| FH     | Coastal         | 121.53    | 29.61    | 41  | 0.57 (0.04)              |
| XS     | Forest          | 113.31    | 28.61    | 40  | 0.67 (0.06)              |
| WJ     | Farmland        | 103.84    | 30.55    | 39  | 0.28 (0.02)              |
| ZY     | Farmland        | 104.63    | 30.13    | 42  | 0.74 (0.06)              |
| YT     | Farmland        | 105.47    | 31.28    | 30  | 0.78 (0.06)              |
| JJ     | Farmland        | 106.18    | 29.06    | 12  | 0.94 (0.08)              |

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