Quantification of Atmospheric Ammonia Concentrations: A Review of Its Measurement and Modeling

Arshad Arjunan Nair * and Fangqun Yu

Atmospheric Sciences Research Center, State University of New York at Albany, 251 Fuller Road, Albany, NY 12203, USA; fyu@albany.edu
* Correspondence: aanair@albany.edu

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Abstract: Ammonia (NH₃), the most prevalent alkaline gas in the atmosphere, plays a significant role in PM₂.₅ formation, atmospheric chemistry, and new particle formation. This paper reviews quantification of [NH₃] through measurements, satellite-remote-sensing, and modeling reported in over 500 publications towards synthesizing the current knowledge of [NH₃], focusing on spatiotemporal variations, controlling processes, and quantification issues. Most measurements are through regional passive sampler networks. [NH₃] hotspots are typically over agricultural regions, such as the Midwest US and the North China Plain, with elevated concentrations reaching monthly averages of 20 and 74 ppbv, respectively. Topographical effects dramatically increase [NH₃] over the Indo-Gangetic Plains, North India and San Joaquin Valley, US. Measurements are sparse over oceans, where [NH₃] ≈ a few tens of pptv, variations of which can affect aerosol formation. Satellite remote-sensing (AIRS, CrIS, IASI, TANSO-FTS, TES) provides global [NH₃] quantification in the column and at the surface since 2002. Modeling is crucial for improving understanding of NH₃ chemistry and transport, its spatiotemporal variations, source apportionment, exploring physicochemical mechanisms, and predicting future scenarios. GEOS-Chem (global) and FRAME (UK) models are commonly applied for this. A synergistic approach of measurements↔satellite-inference↔modeling is needed towards improved understanding of atmospheric ammonia, which is of concern from the standpoint of human health and the ecosystem.

Keywords: review; ammonia; NH₃; modeling; measurement; quantification; atmospheric chemistry; particle formation; PM₂.₅

1. Introduction

Atmospheric ammonia (NH₃), mainly from agriculture with additional sources in industrial and vehicular emissions, plays a key role in many aspects of our environment by virtue of its alkalinity, reactivity, solubility, and abundance. In recent years, there has been an increase in atmospheric ammonia concentrations ([NH₃]) mainly due to increased land use for agriculture [1–3] to support our burgeoning population. This has generated concern about its negative impacts on the climate and human health. This is especially due to the role of NH₃ in the formation of PM₂.₅ through its neutralization of acidic species in the atmosphere. NH₃ has been receiving increasing attention recently due to its potential enhancement of atmospheric new particle formation [4,5]. The impacts of NH₃ and the resultant particulate matter on human health [6], ecosystem, and climate deem understanding its concentration in the atmosphere important. NH₃ has the following major impacts on the environment. Firstly, NH₃ is the greatest contributor to reactive nitrogen deposition [7–10] in several regions across the globe. Such deposition can contribute to eutrophication of aquatic and
This section also examines the validation of modeling studies with observations and some insights from publications. Here we review these previous studies, with the following aims: (1) aggregating current findings on the processes that affect the concentration of ambient atmospheric ammonia (NH₃), and its effects. Figure 1 outlines this review; Section 2 details the various measurement techniques of [NH₃]. Some key studies and their results are discussed here. Section 3 discusses the modeling of NH₃ and the processes that affect its concentration in the atmosphere. This section also examines the validation of modeling studies with observations and some insights into this. Section 4 presents a picture of the spatial distribution on the global and regional scales. Section 5 examines the temporal trends in [NH₃] on varying scales from several studies. Issues with the various approaches of quantifying [NH₃] are discussed in each corresponding section and summarized in Section 6, which also concludes with the future steps and challenges in the quantification of atmospheric NH₃.
2. Measurement of $[\text{NH}_3]$  

2.1. In Situ Measurements

It is natural that the first measurements of $\text{NH}_3$ in the gas-phase were made through ground-based instrumentation. The earliest detailed measurements of atmospheric $\text{NH}_3$ were made by Egner and Eriksson [49] over Scandinavia. “Ammonia and nitrate are determined in one aliquot of the sample by successive distillations in all-glass stills with excess of sodium hydroxide, and Devarda’s alloy. The final estimate of ammonia is made with a special Nessler technique, using a photoelectric colorimeter.” However, their approach did not distinguish between the gas-phase $\text{NH}_3$ and the aerosol $\text{NH}_4^+$. The first measurements of $[\text{NH}_3]$ solely in the gas-phase were performed by Junge [50] over locations in Florida and Hawaii, with an important realization that the $\text{NH}_4^+$ aerosol is formed from atmospheric $\text{NH}_3$. Over the next six decades, there have been advancements in measurement methods, initially aiming at distinguishing gas and aerosol phases and the later goal of realizing continuous measurement of $[\text{NH}_3]$.

Towards the first goal, the first major development was the filter pack method [51], which filtered out aerosol particles on a Teflon pre-filter before collection of $\text{NH}_3$ on an acid-coated filter. However, there were issues with volatility of the aerosol $\text{NH}_4^+$ causing positive error, and humidity causing $\text{NH}_3$ deposition in the pre-filter and consequently a negative error in $\text{NH}_3$ measurement [52]. In 1979, Martin Ferm discussed a “method for determination of atmospheric ammonia” based on the differential diffusion rates to a surface for gas molecules and aerosols [53]. Using an oxalic acid-coated tube, separation of $\text{NH}_3$ (trapped onto the tube walls) and $\text{NH}_4^+$ aerosols can be achieved. Many of the later refinements into $\text{NH}_3$ detection instruments have been based on this simple denuder technique. Continuous measurement of $\text{NH}_3$ was achieved by Wyers et al. [54], where a fully automated continuous flow rotating wet denuder was developed.

Numerous ground-based measurements of atmospheric $\text{NH}_3$ are listed in Table S1. The observations made in the reviewed literature are synthesized into Figure 2, with a global map of the average surface $[\text{NH}_3]$ measurements ranging from <1 ppbv in remote continental and oceanic areas to >24 ppbv, typically over regions of intensive agriculture. Longer detailed measurements have been made over the United States, Europe, and China. Some of the important results and inferences from these studies are discussed in detail in subsequent sections. From Table S1 and Figure 2 we note that most in situ measurements of $[\text{NH}_3]$ are over land. Most surface-based measurements over land are made in North America and Europe. Systematic measurements have been made through $\text{NH}_3$ monitoring networks (see Table 1), which have been established after identifying increasing $\text{NH}_4^+$ and $\text{NH}_3$ trends and their effects on $\text{PM}_{2.5}$ formation. In the US, the Ammonia Monitoring Network
(AMoN; Puchalski et al. [55], Butler et al. [56]) provides biweekly integrated surface measurements of [NH$_3$] from a network of 123 sites, with the longest measurement period from October 2007 to the present and an average measurement duration of 7 years. NH$_3$ is collected using a passive diffusion sampler and subsequently its laboratory measurement is conducted by sonic dislodgment of NH$_4^+$ ions from the phosphoric acid sorbent and subsequent flow injection analysis (FIA). In the UK, the National Ammonia Monitoring Network (NAMN) was established in September 1996 and provides monthly integrated surface [NH$_3$] data from a network of 72 active sites. The NAMN uses a combination of an active diffusion denuder method (DELTA samplers: DEnuder for Long Term Atmospheric, Sutton et al. [57]) and passive samplers (ALPHA: Adapted Low-cost Passive High-Absorption sampler, Tang et al. [58]). In the Netherlands, the Dutch National Air Quality Monitoring Network (LML: Landelijk Meetnet Luchtkwaliteit) and the Measuring Ammonia in Nature Network (MAN) have been making measurements of [NH$_3$] across the Netherlands since 1993 and 2005, respectively. The LML reports hourly [NH$_3$] since 1993 at eight sites and since 2014 at six sites using continuous-flow denuders (AMORs: Amanda for MOnitoring RIVM; Wyers et al. [54]) and currently miniDOAS [59], and in two sites, triplets of passive samplers [60] for monthly mean values. While the LML provides a high temporal resolution, its low spatial distribution is compensated by over 300 sites of MAN that provide monthly passive sampling measurements of [NH$_3$]. Apart from these NH$_3$ monitoring networks, observations of [NH$_3$] have been made over the ocean in certain campaigns. However, such in situ measurements are significantly fewer compared to those over land. While this has been justified by their distance from human settlements and there being orders of magnitude less NH$_3$ over oceans compared to land, recent evidence from Yu et al. [5] suggests that NH$_3$ even in pptv levels can have a significant effect on atmospheric new particle formation.

![Figure 2. Global in situ measurements reviewed in this paper aggregated to $1^\circ \times 1^\circ$ resolution.](image-url)
Table 1. Details of long-term [NH$_3$] measurement networks and their data access links.

| Network      | Location | Start Year | Sites | Data Access                                      |
|--------------|----------|------------|-------|--------------------------------------------------|
| AMoN         | US       | 2007       | 123   | http://nadp.slh.wisc.edu/data/AMoN/              |
| AMoN-China   | China    | 2015       | 53    | https://doi.org/10.1021/acs.est.7b05235          |
| LML          | Netherlands | 1993      | 8     | https://www.luchtmeetnet.nl/meetpunten?component=NH3 |
| MAN          | Netherlands | 2005      | 300+  | https://man.rivm.nl/                             |
| NAMN         | UK       | 1996       | 85    | https://uk-air.defra.gov.uk/networks/network-info?view=nh3 |
| NAPS         | Canada   | 2004       | 7     | http://maps-cartes.ec.gc.ca/rnspa-naps/data.aspx* |

*At the time of this review, this link did not contain the [NH$_3$] data, which should be procurable from Yao and Zhang [61].

In recent years, novel methods of instrumentation have been developed for the measurement of [NH$_3$] with a finer temporal resolution. As seen above, most instrumentation deployed in NH$_3$ monitoring networks have poor temporal resolution, to the extent of biweekly sampling for reduced costs. Additionally, offline analysis may introduce errors due to revolatilization of NH$_3$ and human errors. Some recent advances that are being implemented or have the scope of implementation include: chemical ionization mass spectrometers (CIMS), The Quantum Cascade Tunable Infrared Laser Differential Absorption Spectrometer (QC-TILDAS), differential optical absorption spectroscopy (DOAS), and Monitor for AeRosols and GAses in ambient air (MARGA). Chemical ionization mass spectrometers (CIMS) have been developed for fast time resolution measurement of NH$_3$ [62–68]. Compared to other emerging instrumentation, the CIMS technique is highly advantageous due to its fast time response (<1 min). However, Nowak et al. [64] note that the absolute level and the variability in the instrumental background are a limitation. QC-TILDAS determines the mixing ratio of NH$_3$ by monitoring the molecule’s absorption of radiation at 967 cm$^{-1}$ [69]. Differential optical absorption spectroscopy (DOAS) works on the linearization of the Lambert–Beer law. With open-path arrangement, DOAS provides a contact-free technique for in situ measurement of [NH$_3$], within the 203.7–227.8 nm UV wavelength range over path lengths up to 100 m [70–73]. To overcome the issues with instrument performance [74,75], significant improvements have been made over recent years, mainly by Volten et al. [76] and Sinterrmann et al. [77], which improve the reliability as well as reduce the cost of this instrument. The Monitor for AeRosols and GAses in ambient air (MARGA; ten Brink et al. [78], Rumsey et al. [79]) is an online instrument that provides hourly time-resolved measurement of water-soluble gases and aerosols using a dual-channel ion chromatograph. These new developments of in situ [NH$_3$] measurement techniques are exciting for the insights that can come from continuous and high-resolution data, especially from model–observation comparisons.

For further reading, the reader is directed to some literature on the intercomparison of these varied techniques [67,68,80–82].

2.2. Satellite Remote Sensing

NH$_3$ has a remarkable microwave spectrum due to its characteristic “ammonia inversion” [83]. The molecule has a trigonal pyramidal shape with protons on the three points on the base and nitrogen on the top/bottom. This and the fortuitous distance (crossable tunneling barrier) between the protons on the base, allow for nitrogen to pass through the base and change the orientation of the molecule. This causes a strong absorption when microwave/IR photons cause the rapid flipping between the upward pyramidal and downward pyramidal states. Another outcome of the inversion is an inversion doubling, where the infrared spectrum undergoes doubling due to the two possible positions of the nitrogen atoms. Now this opens a whole new avenue for satellite remote sensing of NH$_3$ in the gas-phase. The unique IR spectrum of NH$_3$ can make it distinguishable from other chemical species and background noise. With the plethora of infrared spectrometers available in orbit around Earth, providing the possibility of continuous, real-time, global measurement, it was inevitable that this feature of the NH$_3$ molecule would be tapped into.

The first attempt was when Beer et al. [84] used infrared radiances measured by the Tropospheric Emission Spectrometer (TES) onboard the EOS Aura satellite to infer [NH$_3$] from the spectral residual
differences (calculated as per Rodgers and Connor [85]) in the region of 960–972 cm\(^{-1}\). This was a demonstration of the possibility of detecting NH\(_3\) from nadir viewing remote sensing instruments. Due to the limitations of the TES—it’s small geographic coverage and consequent inability to provide daily coverage—attention was shifted to the Infrared Atmospheric Sounding Interferometer (IASI) onboard the MetOp-A satellite. Although it had a poorer spectral resolution than the TES, it provided a broader spatial coverage. Clarisse et al. [86] made the first annual “global NH\(_3\) integrated concentrations retrieved from satellite measurements.” Expectedly, there has been a subsequent flurry of activity (see Table 2) in using satellite remote sensing to measure ambient NH\(_3\) in the atmosphere.

Satellite remote sensing is capable of capturing spatiotemporal variations in columns as well as surface NH\(_3\) concentrations [86–90]. For instance, Heald et al. [91] found underestimation of NH\(_3\) emissions in the Midwest during spring and in California using IASI data. This was followed by demonstration of potentially constraining NH\(_3\) emissions using TES [NH\(_3\)] data [92], with improved modeling of [NH\(_3\)] over the US. Thus satellite remote sensing of [NH\(_3\)] can also provide constraints towards improvement of NH\(_3\) emission inventory used in chemical transport models.

Table 2. Some satellite remote sensing studies of atmospheric NH\(_3\) and instrumentation details.

| Reference | Region | Instrument | Period | Typical Concentration |
|-----------|--------|------------|--------|-----------------------|
| [84]      | Beijing| TES        | 10 Jul 2007 | 5 to 25 ppbv           |
| [86]      | Global | IASI       | 2008    |                       |
| [93]      | Mediterranean basin | IASI | 25 Aug 2007 | 5.7 ± 0.1 × 10\(^{16}\) molecules cm\(^{-2}\) |
| [94]      | Global | IASI       | 2006 to 2009 (Various) |                       |
| [95]      | California | IASI | 2008 | 3 to 10 ppbv           |
| [97]      | North Carolina | TES | 2002 | 1 to 6 ppbv           |
| [96]      | Global | IASI       | Apr 2009 to Mar 2010 | < 8 × 10\(^{17}\) molecules cm\(^{-2}\) |
| [97]      | Central Russia | IASI | Jul and Sept 2010 | < 8 × 10\(^{17}\) molecules cm\(^{-2}\) |
| [92]      | Global | IASI       | Apr, Jul, and Oct 2006 to 2009 | < 10\(^{17}\) molecules cm\(^{-2}\) |
| [88]      | Global | IASI       | May 2016 | 2 to 10 ppbv           |
| [90]      | Global | TANSO-FTS  | 2009 to 2014 | ≈ 10\(^{18}\) molecules cm\(^{-2}\) |

| Instrument | Satellite  | Agency | Period | Overpass | Field-of-View | Data Access |
|------------|------------|--------|--------|-----------|---------------|-------------|
| AIRS       | Aqua       | NASA   | 2002–2016 | 13:30 | 13.5 km | https://dx.doi.org/10.5067/AIRA/TES/TL2NH3N.008 |
| CrIS       | Suomi-NPP  | NOAA   | 2012–2017 | 13:30 | 14 km | https://pvc.colab.science.gc.ca/~max01/satellite_ext/cris/isspp/n3/v1_5/ |
| IASI       | MetOp-A/B/C | ESA    | 2006–2020 | 09:30 | 12-39 km | https://iasi.aeri-data.fr/kt3/ |
| IASI       | MIPAS      | ESA    | 2002–2012 | N/A * | 420 km | http://share.lsf.kit.edu/mk/asi/sat/mipas-export/ |
| TANSO-FTS  | GOSAT      | JAXA   | 2009–2020 | 13:00 | 10.5 km | https://doi.org/10.5194/amt-13-309-2020 |
| TES        | Aura       | NASA   | 2004–2018 | 13:30 | 5–8 km | https://dx.doi.org/10.5067/AURA/TES/TL2NH3N.008 |

Abbreviations: FTIR: Fourier-Transform InfraRed spectroscopy, AIRS: Atmospheric Infrared Sounder onboard NASA’s (National Aeronautics and Space Administration, US) Aqua satellite, CrIS: Cross-track Infrared Sounder onboard NOAA’s (National Oceanic and Atmospheric Administration, US) Suomi NPP satellite, IASI: Infrared Atmospheric Sounding Interferometer onboard the ESA’s (European Space Agency) MetOp satellites, MIPAS: Michelson Interferometer for Passive Atmospheric Sounding onboard ESA’s Envisat satellite, TANSO-FTS: Thermal And Near infrared Sensor for carbon Observations—Fourier Transform Spectrometer onboard JAXA’s (Japan Aerospace eXploration Agency) GOSAT-1, TES: Tropospheric Emission Spectrometer onboard NASA’s Aura satellite. Overpass refers to the daytime (when thermal contrast is optimal for NH\(_3\) retrieval) crossing of the equator. * Overpass not defined for MIPAS as it makes edge of the atmosphere (limb) NH\(_3\) retrievals.
While satellite-based inference can fill the gaps in observations of $\text{NH}_3$ and additionally provide its vertical profile, there are limitations associated with this technique as noted in several studies listed in Table 2. Currently deployed instrumentation is not onboard geostationary satellites, which results in discontinuous temporal measurements. The requirement of a strong thermal contrast reduces reliability of nighttime measurements. The presence of clouds also affects the retrievals. Furthermore, there are issues in the inference of $\text{NH}_3$ from the radiances measured by the satellite instrument, with a priori assumptions on the $\text{NH}_3$ profile and shape for conversion of radiances into a concentration. This is further compounded by the small signal of $\text{NH}_3$ in comparison to the background.

To overcome and understand the above issues, validation studies have been carried out. For the TES, $\text{NH}_3$ retrievals were able to capture spatiotemporal patterns observed on the surface by an ammonia monitoring network (CAMNet) in North Carolina [87] and with aircraft measurements [99]. Damme et al. [102] demonstrated the consistency of IASI $\text{NH}_3$ retrievals with surface $\text{NH}_3$ monitoring networks and aircraft campaigns. These studies show that despite the aforementioned limitations, satellite-based remote-sensing inference of $\text{NH}_3$ is an invaluable method of quantification, a tropospheric vertical profiler, and a source apportioner of atmospheric ammonia.

### 3. Modeling $\text{NH}_3$

Atmospheric modeling provides another approach to quantify $\text{NH}_3$ in the atmosphere. Considering $\text{NH}_3$, models can (1) evaluate the environmental impacts of their emissions and depositions, (2) help project the impact of future scenarios, (3) aid in developing our understanding of the processes controlling atmospheric $\text{NH}_3$, (4) examine the role of $\text{NH}_3$ in affecting atmospheric chemical and physical processes, and (5) study aerosol formation (both mass and number concentrations) dependent on $\text{NH}_3$.

The first modeled global distributions of $\text{NH}_3$ were derived by Dentener and Crutzen [113] with their development of an $\text{NH}_3$ emission inventory incorporated into a climatological three-dimensional global tropospheric transport model (MOGUNTIA; Zimmermann [114]). This was a first attempt at understanding the fate of reduced nitrogen species in the atmosphere. A $1^\circ \times 1^\circ$ $\text{NH}_3$ emission inventory was used in this initial modeling study. Subsequently, there has been vigorous development in the modeling of $\text{NH}_3$, with two approaches: Eulerian (Table 3) and Lagrangian (Table 4). The more typical implementation is the Eulerian approach (in GEOS-Chem, CMAQ, and EMEP, among others), where the properties of reference grid cells are monitored. The Lagrangian approach, which “follows” the air parcel, is applied in models such as FRAME, TREND, and STILT-CHEM.

GEOS-Chem (www.geos-chem.org) is one of the most widely used (see Table 3) three-dimensional chemical transport models (3-D CTM) in the study of $\text{NH}_3$ in the atmosphere. Due to the environmental policies in Europe, there are several implementations for the region—TM5 [115], the EMEP model [116–118], the DAMOS/Danish Eulerian Hemispheric Model (DEHM) [119–121], CHIMERE [122], MATCH [123], and LOTUS-EUROS [124]. The Community Multiscale Air Quality Modeling System (CMAQ) [125], while commonly used for air quality studies, especially over North America, is not widely [126] used to understand $\text{NH}_3$. The Particulate Matter Comprehensive Air Quality Model with Extensions (PMCAMx) [127–129] has also been utilized for modeling $\text{NH}_3$ over the US [130] and Europe [131]. Unlike these Eulerian CTMs, the Lagrangian approach in modeling atmospheric $\text{NH}_3$ is used in the following: TERN [132], TREND [133,134], ACDEP [119,135–140], FRAME [141–143], NAME [144], AURAMS [145], OPS [146–148], and STILT-Chem [149] models. Some of the significant modeling studies of atmospheric $\text{NH}_3$ using these various models are enumerated in Tables 3 and 4. The following three subsections detail the three most important processes to be considered in the modeling of $\text{NH}_3$, viz., emission, gas–particle partitioning, and deposition.
### Table 3. Some modeling studies of atmospheric [NH₃] using Eulerian chemical transport models.

| CTM          | Equilibrium Model | Reference                        | Region                  | Period                |
|--------------|-------------------|----------------------------------|-------------------------|-----------------------|
| CHIMERE      | ISORROPIA         | De Meij et al. [122]             | Northern Italy          | 2005                  |
| CMAQ         | MARS-A            | Gilliland et al. [150]           | Eastern US              | 2001                  |
| CMAQ         | MARS-A            | Gilliland et al. [126]           | US                      | 2001                  |
| DAMOS        | Froh [151]        | Brandt et al. [128]              | Europe, North America   | 2007                  |
| DAMOS        | Froh [151]        | Geels et al. [122]               | Denmark                 | 2007                  |
| EMEP         | EQSAM             | Furgeri and Aos [116]            | Europe                  | 1990-2003             |
| EMEP         | EQSAM             | Horvath et al. [117]             | Hungary                 | 1990 and 1995-2004    |
| GEOS-Chem    | ISORROPIA II      | Heald et al. [91]                | US                      | May 2009 to April 2010|
|              | ISORROPIA II      | Walker et al. [152]              | California              | 2009                  |
|              | RPMARES           | Zhang et al. [153]               | US                      | 2006 to 2008          |
|              | ISORROPIA II      | Schiferl et al. [34]             | California              | May and Jun 2010      |
|              | ISORROPIA II      | Paulot et al. [154]              | Global oceanic           | 1995-2004             |
|              | RPMARES           | Luo et al. [103]                 | Global                  | 2007                  |
|              | MARS-A            | Zhu et al. [135]                 | US                      | 2008                  |
|              | ISORROPIA II      | Schiferl et al. [156]            | US                      | 2001-2017             |
|              | ISORROPIA II      | Nair et al. [138]                | US                      | 2001-2017             |
|              | RPMARES           | Paulot et al. [159]              | US, E.U., China         | 2005-2008             |
| LOTOS-EUROS  | Modified* CBM-IV  | van Damme et al. [160]           | Europe                  | 2008-2011             |
| MATCH        | ISORROPIA         | Langner et al. [123]             | Baltic Sea Basin        | 1996-2001             |
| PMCAM        | ISORROPIA         | Pinder et al. [130]              | US                      | Jul and Oct 2001, Jan and Apr 2002 |
|              | RPMARES           | Murphy and Pandis [127]          | Eastern US              | 2003                  |
|              |                    | Tsipidis et al. [126]            | Mexico City             | Apr 2003              |
|              |                    | Karydis et al. [125]             | Mexico City             | Apr 2003              |
|              |                    | Fourtounakis et al. [133]        | Europe                  | 2008                  |
| TMS          | EQSAM             | de Meij et al. [115]             | Europe                  | 2000                  |

See Table S2 for details of Equilibrium Models. Abbreviations: CTM: Chemical Transport Model, CHIMERE, CMAQ: Community Multiscale Air Quality, DAMOS: Danish Ammonia MOdeling System, EMEP: European Monitoring and Evaluation Programme, GEOS-Chem: Goddard Earth Observing System - Chemistry, LOTOS-EUROS: LOngTerm Ozone Simulation - EURopean Operational Smog, MATCH: Multi-scale Atmospheric Transport and Chemistry, PMCAM:x: , TMS: Tracer Model 5. * Modification of the Carbon Bond Mechanism IV reaction mechanism with a scheme for NH₄NO₃ and (NH₄)₂SO₄ by Zlatev [161].

### Table 4. Some modeling studies of atmospheric [NH₃] using Lagrangian chemical transport models.

| CTM          | Equilibrium Model | Reference                        | Region                  | Period                |
|--------------|-------------------|----------------------------------|-------------------------|-----------------------|
| ACDEP        | Modified* CBM-IV  | Hertel et al. [135]              | Denmark                 | 1990                  |
|              |                   | Hertel et al. [119]              | Baltic Sea              | 1999-2001             |
|              |                   | Gyldenkaerne et al. [136]        | Northwestern Europe     | 1999-2001             |
|              |                   | de Leeuw et al. [137]            | North Sea               | 1999                  |
|              |                   | Skjøth et al. [138]              | Denmark                 | 1998 and 1999         |
|              |                   | Skjøth et al. [139]              | Denmark                 | 1999 to 2001          |
|              |                   | Skjøth et al. [140]              | Europe                  | 2007                  |
|              |                   | Makar et al. [162]               | North America           | 2003                  |
| AURAMS       | FRAME             | Makar et al. [163]               | British Isles           | 1996                  |
|              |                   | Fournier et al. [164]            | British Isles           | 1996                  |
|              |                   | Fournier et al. [165]            | British Isles           | 1996                  |
|              |                   | Fourrier et al. [166]            | UK                      | 1996                  |
|              |                   | Dore et al. [167]                | UK                      | 2002                  |
|              |                   | Sutton et al. [168]              | UK                      | 2002-2004             |
|              |                   | Tang et al. [169]                | UK                      | 2012                  |
|              |                   | Verno [170]                      | UK                      | 1999                  |
|              |                   | Sutton et al. [171]              | UK                      | Sep 1996 to Nov 2000  |
|              |                   | Sutton et al. [172]              | UK                      | 1996                  |
|              |                   | Heilsten et al. [173]            | UK                      | 2000                  |
|              |                   | Single et al. [174]              | Great Britain           | 1987 to 1990          |
|              |                   | Hallsworth et al. [175]          | UK                      | 2002-2004             |
|              |                   | Krzyz et al. [176]               | Poland                  | 2002-2005             |
|              |                   | Zhang et al. [177]               | North China Plain       | 2003                  |
| NAME         | OPS               | Meng and Seinfeld [178]          | UK                      | 1996                  |
|              |                   | Redington and Derwent [144]      | Netherlands             | Sep 2000 to 2003      |
|              |                   | van Jaarsveld [179]              | Netherlands             | Jan 1998 to Dec 2000  |
|              |                   | van Pal et al. [146]             | Eastern Netherlands     | 2002-2003             |
|              |                   | Stolk et al. [147]               | Netherlands             | 2005-2007             |
|              |                   | van Pal et al. [148]             | Netherlands             | Jul 2002-Aug 2003     |
|              |                   | Krut et al. [149]                | Netherlands             | 1990-2014             |
|              |                   | van der Swaluw et al. [180]      | Netherlands             | 1998-2002             |
| STILT-Chem   | Wen et al. [149]  | Wen et al. [149]                 | Canada                  | Jun to Nov 2006       |
| TERN         | TREND             | ApSimon et al. [132]             | UK                      | 1989                  |
|              |                   | Asman and van Jaarsveld [133]    | Europe                  | 1990                  |
|              |                   | Asman [134]                      | Denmark                 | 1996                  |

Abbreviations: CTM: Chemical Transport Model, ACDEP: Atmospheric Chemistry and DEPosition, AURAMS: A Unified Regional Air quality Modeling System, FRAME: Fine Resolution AMmionia Exchange, NAME: Nuclear Accident ModEl, OPS: Operational Priority Substances, STILT-Chem: Stochastic Time-Inverted Lagrangian Transport and Chemistry, TERN: Transport over Europe of Reduced Nitrogen. * Modification of the Carbon Bond Mechanism IV reaction mechanism with a scheme for NH₄NO₃ and (NH₄)₂SO₄ by Zlatev [161].
3.1. Emission

Emission inventories are required for representation in chemical transport models (CTMs), in addition to utility for assessment of air quality policies. They have been developed on scales ranging from global to regional to local for use in CTMs. Global NH$_3$ emission inventories include the Community Emissions Data System (CEDS; Hoesly et al. [179]), the Emissions Database for Global Atmospheric Research (EDGAR; Crippa et al. [180]), the Magnitude And Seasonality of AGricultural Emissions for NH$_3$ (MASAGE_NH$_3$; Paulot et al. [159]), and the Global Emissions Inventory Activity (GEIA; Bouwman et al. [18]). Examples of regional NH$_3$ emission inventories are the US EPA/NEI (National Emission Inventory), Canada’s APEI (Air Pollutant Emissions Inventory; St-Pierre et al. [181]), EMEP (European Monitoring and Evaluation Programme) WebDab, NAEI (National Atmospheric Emission Inventory) for the UK [182], NEMA (National Emission Model for Ammonia; Velthof et al. [183]) for Netherlands, Asian MIX inventory [184], Regional Emission inventory in ASia (REAS) [185–190], and DICE-Africa (Diffuse and Inefficient Combustion Emissions in Africa; Marais and Wiedinmyer [191]). These emission inventories are detailed in Table 5 and their long-term trends are shown in Figure 3. There are inventories developed with higher spatial resolution for the Eastern United States [150], North Carolina and the San Joaquin Valley [192], and the Pearl River Delta [193] and Yangtze River Delta [194] in China.

NH$_3$ emissions are primarily anthropogenic, coming mainly from agriculture through excreta from domestic animals and the use of synthetic fertilizers [195–201]. Approximately 60% of total NH$_3$ emissions are from anthropogenic sources [18,202], of which 80–90% are from agricultural activity (fertilizers and livestock wastes) [18,192,195,203]. Other sources of NH$_3$ include fuel combustion from vehicles and industries, biomass burning, and human wastes. Emission inventories are, therefore, generally based on emission factors from a particular source type and associated activity rate. Emission factors are based on measurement of NH$_3$ fluxes from individual source types, such as animal houses and storage, fertilizer application, fuel combustion, biomass burning, human wastes, industries, transportation, vegetation, and soil. Activity factors indicate the potential emission from each source for specific locations based on parameters like the amount of livestock, the amount of fertilizer applied, and the amount of fuel combusted, based on the source type. The multiplicative product of the source-specific emission factor with the activity factor is the estimated spatial distribution of NH$_3$ emissions, i.e., the emission inventory.

![Figure 3](image-url)
Table 5. Global and regional ammonia emission inventories.

| Inventory | Location | Period      | Reference                                      | Data Access                                      |
|-----------|----------|-------------|------------------------------------------------|-------------------------------------------------|
| CEDS      | Global   | 1750–2014   | Hoesly et al. [179]                            | https://github.com/JGCRI/CEDS/                   |
| EDGAR     | Global   | 1970–2015   | Crippa et al. [180]                            | https://edgar.jrc.ec.europa.eu/overview.php?c=50_AP |
| MASAGE_NH3 | Global   | 2006        | Paulot et al. [159]                            | http://geoschemdata.computecanada.ca/ExtData/HEMCO/MASAGE_NH3/v2015-02/* |
| APEI      | Canada   | 1990–2018   | St-Pierre et al. [181]                         | https://open.canada.ca/data/en/dataset/face688b-bf78-4eb9-9c1e-2a5534b92131 |
| DICE-Africa | Africa | 2006, 2013  | Marais and Wiedimmyer [191]                    | https://doi.org/10.1021/acs.est.6b02602         |
| EMEP      | EU       | 1990–2018   |                                  | https://www.ceip.at/webdb-emission-database/     |
|         |          |             | reported-emissiondata                         | reported-emissiondata                           |
| EPA/NEI   | US       | 1990–2019   |                                  | https://www.epa.gov/air-emissions-inventories/rational-emissions-inventory-nei/ |
| MIX       | Asia     | 2006, 2008  | Li et al. [184]                               | http://meicmodel.org/dataset-mix.html           |
| NAI      | UK       | 1980–2018   | Ress et al. [182]                             | https://naei.beis.gov.uk/data/                   |
| NEMA      | Netherlands | 1990–2018 | Veldhof et al. [183]                          | https://www.wur.nl/nl/Onderzoek-Resultaten/Projects/Commissie-van-Deskundigen-Meststoffenwet/CDM/Documenten/Gasvormige-emissies-NEMA.htm |
| REAS      | Asia     | 1950–2015   | Kurokawa and Ohara [185]                      | http://www.nies.go.jp/REAS/                      |

Abbreviations are noted in-text. * The original dataset at https://fpaulot.bitbucket.org/MASAGE/ was unavailable at the time of publication of this review article.

The main issues with the development of reliable NH₃ emissions inventories are the dearth of emission measurements, incomplete identification of all known sources of emissions, the lack of validation with [NH₃] measurements, the variability and uncertainty of emissions estimates dependent on free ammonia concentration, differences in inventory compilation approaches due to assumptions regarding underlying emission factors and activity rates, and source significance influenced by spatial scales [130,182,192]. While the bottom–up approach of emissions inventory is expected to be accurate due to the detailed consideration of each source type and their emissions, it requires comprehensive information for activity factors to apply emission factors. This would require detailed measurements to capture the complex spatial variability of emissions across source types as well as temporal variability (as discussed in Section 5). Gilliland et al. [126,150] and Pinder et al. [130] observed erroneous seasonal variations in model-simulated ammonium and reduced nitrogen. Zhang et al. [153] determined that NH₃ emissions (constrained and scaled by observations) are lower by a factor of 3 in winter relative to summer and are in better agreement with US network measurements of NH₄⁺ and NH₄⁺ wet deposition fluxes. Applying modifications seemed to correct the previous observations. The underestimation of NH₃ emissions in models was confirmed further by Walker et al. [152], with the additional under-prediction of HNO₃ contributing to under-prediction of nitrate over California. Recently, top–down approaches have been shown to reduce the identified issues with the accuracy of NH₃ emissions inventories by using inverse modeling to relate observed [NH₃] to model emissions [91,92,126,130,150,152,159,204]. Although these studies have to grapple with the complexities of other model processes, comparison with bottom–up emission inventories can identify improvements in NH₃ source identification and contribution. Another approach towards improving emissions inventories is using a combination of bottom–up and top–down approaches in a “hybrid” approach [139,205]. Regardless, current emission inventories have enabled high-resolution chemical transport models to generally capture the spatial variability of atmospheric NH₃ and NH₄⁺ concentrations and provide a wealth of information that is currently unobtainable from direct observations, while also improving our understanding of the effects of various processes and parameters in determining [NH₃].

3.2. Gas–Particle Partitioning

Another significant aspect of modeling atmospheric [NH₃] is the accurate capture of phase partitioning. This is achieved through either thermodynamic equilibrium schemes or explicit mass transfer dynamical schemes. Thermodynamic equilibrium schemes generally assume aerosols to be internally mixed, i.e., all aerosol particles of a certain size range have the same composition. They
also generally assume thermodynamic equilibrium in the gas–particle partitioning of volatile chemical species. These approaches trade off accuracy (the former) versus computational resources (the latter). Some balance can be achieved by assuming the volatile constituents being in equilibrium and shifting to mass transfer schemes when this assumption is not valid in conditions resulting in a longer chemical equilibration time as compared to the gas–aerosol diffusion timescale, typically in cooler conditions.

Over the last four decades, there has been steady development in this aspect. EQUIL was developed by Bassett and Seinfeld [206] to calculate the aerosol composition of the \( \text{NH}_4^+ - \text{SO}_4^{2-} - \text{NO}_3^- - \text{H}_2\text{O} \) aerosol system. KEQUIL was their [207] improvement of EQUIL with the incorporation of the Kelvin effect. Saxena et al. [29] developed the MARS scheme for the \( \text{SO}_4^{2-} - \text{NO}_3^- - \text{NH}_4^+ - \text{H}_2\text{O} \) system with reduced computational time through sub-domains based on RH and \( \text{NH}_3\text{SO}_4^{2-} \) for aerosol species to reduce the number of equations. Further improvements were made by Binkowski and Shankar [208] and Binkowski and Roselle [209]. SEQUILIB by Pilinis and Seinfeld [210] considered Na and CI, which were heretofore ignored, but are important for marine aerosols. Kim et al. [211] developed the computationally efficient gas–aerosol equilibrium model SCAPE (Simulating Composition of Atmospheric Particles at Equilibrium) without limiting assumptions. AIM [212] and AIM2 [213] approached the problem by direct minimization of the Gibbs free energy. The absence of approximations on the equilibrium concentration made AIM computationally intensive. GFEMN [214] was another iterative Gibbs free-energy minimization method.

A significant step forward towards improved computational efficiency was achieved by Nenes et al. [215] with the development of ISORROPIA. ISORROPIA examines the \( \text{Na}^+ - \text{NH}_4^+ - \text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^- - \text{H}_2\text{O} \) aerosol system using pre-calculated lookup tables and a single level of iteration to make this scheme computationally efficient. This was further developed by Fountoukis and Nenes [216] into ISORROPIA II, which treats the \( \text{K}^+ - \text{Ca}^{2+} - \text{Mg}^{2+} - \text{NH}_4^+ - \text{Na}^+ - \text{SO}_4^{2-} - \text{NO}_3^- - \text{Cl}^- - \text{H}_2\text{O} \) aerosol system, i.e., added crustals. ISORROPIA II is currently the most used thermodynamic equilibrium module within GEOS-Chem. A contrasting effort aimed at accuracy over speed is EQUISOLV [217], where direct numerical computation without simplifying assumptions was used to obtain equilibrium concentration. The three levels of nested iteration loops for higher order reactions made this computationally expensive. This was further developed into EQUISOLV II [218] with the replacement of the mass-flux iteration (MFI) method with the analytical equilibrium iteration (AEI) method for solving the set of equilibrium equations. Furthermore, the scheme was expanded to consider the potassium, calcium, magnesium, and carbonate systems. Development of thermodynamic equilibrium schemes has not been stagnant, with recent examples including HETV (HETerogeneous Vectorized) [162], MESA (Multicomponent Equilibrium Solver for Aerosols) [219], ADDEM (Aerosol Diameter DEpendent Model) [220], and UHAEREO (University of Houston inorganic atmospheric AEROsol phase equilibrium model) [221]. The multicomponent equilibrium solver for aerosols (MESA) appears most interesting, with the use of a computationally efficient modified pseudo-transient continuation technique for solving the set of equilibrium reactions while maintaining overall accuracy. For a detailed review and comparison of thermodynamic equilibrium schemes, we direct you to the works of Kim et al. [211], Amundson et al. [221], Pilinis [222], Zhang et al. [223].

The assumption of equilibrium may not be justified under certain conditions, which then require explicit mass transfer dynamical schemes for gas–particle partitioning. While the equilibrium assumptions are not there to affect the accuracy of the solution, the computational cost hinders the use of these mass transfer schemes within CTMs. Development in this aspect has been limited compared to thermodynamic equilibrium schemes, with work by Meng and Seinfeld [175], Jacobson et al. [217], Jacobson [224,225], Meng et al. [226], Sun and Wexler [227], Pilinis et al. [228]. Hybrid methods have recently been developed, where the condensation or evaporation of aerosol particles with diameters of less than a threshold (\( \approx 1 \mu\text{m} \)) are simulated using thermodynamic equilibrium schemes, and the dynamic mass-transfer approach is used for larger particles [229–232].
3.3. Deposition and Bi-Directional Exchange

Atmospheric ammonia is deposited to the surface in either the gaseous (NH₃) or aerosol (NH₄⁺) form. Unionized NH₃ is mainly transferred out of the atmosphere to the surface through dry deposition [153,233]. Dry deposition of NH₃ is faster than that of NH₄⁺ by a factor of 10–100, depending on [NH₃], plant/terrain, and diurnal variations with meteorological conditions [234]. This leads to a short atmospheric lifetime for NH₃, leading to significant deposition near its sources due to its fast deposition and higher concentration [235,236].

Representation of deposition of NH₃ in models is difficult due to the dearth of its measurements and because this is a complex process. Dry deposition can be typically separated into that onto soil and that onto vegetation. This process is dependent on [NH₃] above the particular surface, the equilibrium of [NH₃] over the surface determining whether emission or deposition occurs, turbulence over the surface, and extent of moisture. Studies, such as Langford and Fehsenfeld [237], show that bi-directional exchange of NH₃ over vegetation is an important process, which still does not find adequate representation in models. Even this process is further complicated in that the mechanism for vegetation as a sink of NH₃ could be either through cuticular or stomatal uptake [234,238]. Sutton et al. [239] identify that an accurate model representation of bi-directional exchange of NH₃ is important for models to accurately quantify [NH₃], with implications for its deposition, emission, re-volatilization, and lifetime.

Although Sutton et al. [240] and Nemitz et al. [241] developed the first models for bi-directional surface–atmosphere exchanges of NH₃, chemical transport models neglected to incorporate these until over a decade later. Studies [242–245] have shown better model–observation agreements resulting in more accurate quantification of [NH₃]. However, some more recent studies highlight that the consideration of bi-directional exchange alone may not be sufficient. While Zhu et al. [155] implemented bidirectional exchange of NH₃ into their GEOS-Chem simulations and observed improved agreement of model simulated [NH₃] with network (AMoN) measurements, the general large underestimates (2–5 times) were not corrected, reiterating the need for an accurate representation of emissions. In contrast to the results of Heald et al. [91] for the San Joaquin valley, Zhu et al. [155] also demonstrated that adjustment to HNO₃ does not significantly affect simulated [NH₃] over AMoN sites. This is consistent with the work of Park [246], which suggested that nitrate formation is NH₃-limited over most of the United States. Incorporation of bi-directional exchange of NH₃ did not resolve the model overestimation (3–5 times) of nitrate. Recent work by Luo et al. [247,248] shows that applying an improved wet scavenging parameterization in GEOS-Chem reduces the overestimation of nitrate and nitric acid in the model and corrects some of the deviations in atmospheric NH₃ concentrations.

3.4. Model–Observation Comparisons

Validation of model-simulated [NH₃] with real measurements is the most important exercise not merely for the justification of model output, but for further model refinement and development. It also aids in improving our understanding of the processes controlling the concentration of ammonia in the atmosphere. A review of the literature indicates that such model–observation comparisons have been limited in spatiotemporal coverage. While there have been some comparisons of modeled [NH₃] with observations over other regions, such as Hungary [117], Denmark [121,139], and Canada [149], a majority of the more comprehensive studies have been made over the United States using the GEOS-Chem model.

Over the US, GEOS-Chem underestimation of [NH₃] was attributed to excessive HNO₃ formation from N₂O₅ hydrolysis in the model by Zhang et al. [153]. The authors of Heald et al. [91] examined GEOS-Chem simulated [NH₃] with ground-based (IMPROVE) and satellite (IASI) measurements. The study demonstrated that NH₃ emissions were underestimated in California and in the Midwest, which was the likely reason for the underestimation of NO₃⁻ formation in GEOS-Chem. This was further confirmed by Walker et al. [152] with the added possibility of HNO₃ underestimation and
topography effects on mixed-layer depths. Both studies showed that GEOS-Chem underestimated \([\text{NH}_3]\) in comparison with satellite and in situ measurements.

Schiferl et al. [156] showed that the mean modeled \([\text{NH}_3]\) was underestimated (2.5 ppbv vs. observed 3.4 ppbv) over the US during summer through a comparison with measurements from 2008 to 2012 at 11 AMoN sites. During summer, the range in model-simulated \([\text{NH}_3]\) was smaller, and its mean was lower (by more than 25%) when compared to AMoN measurement data. Schiferl et al. [156] suggests that this may not be model deficiency, but that the AMoN sites’ locations near high \(\text{NH}_3\) source regions causes a sampling bias due to inadequate representation of the range of \([\text{NH}_3]\) across the US. Although modeled \([\text{NH}_3]\) was underestimated during summer, especially near source regions (including both agricultural and fire emissions), it showed consistency in the spatiotemporal variability of \([\text{NH}_3]\) in the column and at the surface. Inter-annual variability of modeled surface \([\text{NH}_3]\) was lower than measurements, but the trends and variability were significant considering the fixed \(\text{NH}_3\) emissions in the model.

Paulot et al. [159] optimized simulated seasonality and magnitude of \([\text{NH}_3]\) in the Northeast and Southwest US using the adjoint of GEOS-Chem by inversion of \(\text{NH}_4^+\) wet deposition fluxes from NADP network data for the period 2005–2008. They developed a novel bottom–up emission inventory (MASAGE\_\text{NH}_3), which simulated the magnitude and seasonality of \([\text{NH}_3]\) in better agreement with observations in the Northeast and Southeast US, which is consistent with overestimated \(\text{NH}_3\) emissions in the US National Emissions Inventory. In the Midwest and upper Midwest, spring enhancement in \(\text{NH}_3\) was captured but not the elevated summer concentrations. Underestimation is still significant in the Atlantic and Central regions, especially in winter in the Central US.

US \(\text{NH}_3\) sources were constrained using TES satellite observations with the GEOS-Chem model and its adjoint by Zhu et al. [92]. There was an improvement in the underestimation of \(\text{NH}_3\). The range and variability improved in April and October with reduced model underestimates; however there was overestimation in July due to the constraints applied. Zhu et al. [155] tried further improvements by incorporating a bi-directional exchange (BIDI) scheme for \(\text{NH}_3\), which improves the normalized mean bias. Large underestimations (especially in October and April) still existed, which is likely due to significant errors in \(\text{NH}_3\) emission inventories.

Yu et al. [157] and Nair et al. [158] comprehensively assessed long-term (last two decades) GEOS-Chem-simulated \([\text{NH}_3]\) over the US with empirical measurements. The strong dependence on emissions for seasonality and on acid precursor gases for long-term trends were demonstrated. Potential improvements in the representation of emissions, especially over the US Great Plains region, were identified. Furthermore, their results indicate that modeled \([\text{NH}_3]\) is more strongly dependent on \(\text{NH}_3\) emissions than observations are. Additionally, especially over the Southeast US, considerations of changing acid precursor gas concentrations or particle acidity may need to be made in modeling \(\text{NH}_3\).

Such studies provide an examination of the role of acid precursor gases in the \(\text{NH}_3\) budget, improvement of emission parameterization, land-use and topographical considerations, evaluation of thermodynamic partitioning, and identification of other avenues for better process representation towards improved modeling of \([\text{NH}_3]\). This is important considering the expected increasing trend of \([\text{NH}_3]\) and its role in various atmospheric chemical processes, especially those resulting in increased aerosol loading. It should be noted that during model–observation comparisons, it is vital to also understand the limitations of observational techniques, consider nonlinearities in these comparisons (which can reveal rich information), and additionally facilitate inter-model comparisons.

4. Spatial Distributions

\([\text{NH}_3]\) is predominantly controlled by emissions due to the short atmospheric lifetime of \(\text{NH}_3\). Transport, obviously, plays a role; especially that of particulate ammonium that re-volatilizes into the gas-phase \(\text{NH}_3\). The main source of \(\text{NH}_3\) emissions is agricultural land, especially in the period after chemical fertilizer application. Consequently, most \(\text{NH}_3\) hotspots are over these regions of agriculture.
Putting together the vast literature of surface measurements of \( \text{NH}_3 \) shows their spatial distribution and hotspots (Figure 2). Over the US, the highest surface \( \text{NH}_3 \) are observed in the Midwest and California. This is primarily due to agriculture (including concentrated animal feeding operations) and biomass burning, respectively. Similar reasons explain the observed hotspots of \( \text{NH}_3 \) over Europe and the North China Plain. The recent use of satellite remote sensing to quantify \( \text{NH}_3 \) has the advantage of directly providing a global picture. Clarisse et al. [86] provided the first such global \( \text{NH}_3 \) map using data from the IASI/MetOp satellite for the year 2008. Their results confirm global \( \text{NH}_3 \) hotspots (total column \( \text{NH}_3 > 0.5 \mu g \text{ m}^{-2} \)) over agricultural valleys and regions of biomass burning. Using 5 years of IASI measurements, Damme et al. [102] provided a more detailed global map. Agricultural hotspots were identified over the Indo-Gangetic plain, the North China Plain, and other highly irrigated regions in Asia. Over Indonesia, there was the combined effect of intensive fertilizer application in Java and wildfires in Borneo and Sumatra. Over South America, hotspots were mainly due to biomass burning, but new agricultural hotspots over Chile and Colombia-Venezuela were revealed. Over North America, \( \text{NH}_3 \) was elevated over the US Midwest and the San Joaquin Valley. Anthropogenic \( \text{NH}_3 \) effects were seen in parts of Canada as well. Over Europe, hotspots were over the Netherlands and the Po Valley, Italy. The effect of industrial emissions was captured over South Africa. Over these hotspots, measurements would exceed total column \( \text{NH}_3 \) of \( 3 \times 10^{16} \) molecules \text{cm}^{-2}. Using TES data for the year 2007, Luo et al. [100] showed the enhancement of \( \text{NH}_3 \) over Northern India (up to 14.45 ppbv \( \text{NH}_3 \) in the summer) and North-Central China and spring biomass burning effects in parts of Africa and Asia. Warner et al. [89] provided a 13-year record of global \( \text{NH}_3 \) distribution that confirmed the importance of agriculture and biomass burning in determining the hotspots over these regions.

Agricultural land is often mixed-use, with livestock rearing, which further contributes to \( \text{NH}_3 \) emissions. Xing et al. [249] indicated that \( \text{NH}_3 \) emissions from livestock activities have increased by 11% from 1990 to 2011 in the US. This could have effects on the spatial distribution of \( \text{NH}_3 \). Li et al. [250] found large spatial differences in \( \text{NH}_3 \) over the northeastern plains of Colorado, a region of concentrated agricultural activities and animal feeding operations, with mean \( \text{NH}_3 \) concentrations ranging between 4 and 60 ppbv from grasslands to feedlots with almost one hundred thousand cattle. Over the US, data from measurements and modeling [157,158] indicate spatial heterogeneity based on land use. The Midwest shows the highest concentrations of \( \text{NH}_3 \) due to intensive agricultural activities (including livestock rearing) as well as the energy sector. In the eastern part of the US, \( \text{NH}_3 \) in the gas-phase is limited due to elevated \( \text{SO}_2 \) and \( \text{NO}_x \) emissions from the coal-fired power plants as well as manufacturing in the Ohio River Valley region. In the western part of the US, \( \text{NH}_3 \) is elevated over the San Joaquin Valley, possibly due to biomass burning and agriculture. Their study indicates that hotspots are highly \( \text{NH}_3 \) emission-dependent and cold spots are highly \( \text{SO}_2 \) and \( \text{NO}_x \) emission-dependent.

China is a large agricultural nation contributing approximately 20% of global \( \text{NH}_3 \) emission [186,188,189]. Here, roughly 20% of the world population is fed by 10% of Earth’s arable land, requiring intensive agriculture and livestock rearing. The concentrated use of fertilizers (30% of global usage) as well as livestock wastes contribute significantly to \( \text{NH}_3 \) emissions [251,252]. Meanwhile, chemical fertilizer application in China is less efficient, resulting in a high degree of nitrogen loss (\( \text{NO}^- \), \( \text{NH}_3 \), \( \text{N}_2\text{O} \), and \( \text{N}_2 \)). Furthermore, about 30% of livestock products originate from the North China Plain (NCP), which further increases the \( \text{NH}_3 \) emission in this highly polluted region. Over China, Liu et al. [253] used IASI satellite \( \text{NH}_3 \) \( \text{NH}_3 \) retrievals and vertical \( \text{NH}_3 \) profiles from MOZART to provide a comprehensive estimate of surface \( \text{NH}_3 \) over the region. The spatial distribution of \( \text{NH}_3 \) was as expected, aligned with intensive agricultural areas. The deviations from this distribution were explainable by concentrated animal farming locations, where livestock wastes contributed more than fertilizer use on farms. \( \text{NH}_3 \) emission mitigation was not a focus for the nation until 2015, when a \( \text{NH}_3 \) monitoring network (AMoN-China) was established. Using this network’s data, Pan et al. [254]
identified the NCP as having the highest [NH$_3$] followed by smaller hotspots over the Tarim basin, Chengdu Plain, and Guanzhong Plain coinciding with intensive agricultural activity.

5. Temporal Trends

The concentration of NH$_3$ in the atmosphere is mainly determined by its emission. Emission is highly human activity and temperature dependent. It is expected then that the temporal trends of NH$_3$ would be strongly correlated to those of human activity and temperature. Below we discuss the trends in [NH$_3$] at different temporal scales.

5.1. Diurnal

Diurnal variation in ambient [NH$_3$] has been observed to varying degrees. Variation in temperature is the primary reason for this. It may mainly affect local emissions, which are temperature-dependent and may also modulate the boundary layer depth and consequently the concentrations at the surface. Additionally, gas–particle partitioning generally peaks in the afternoon. Thus, the NH$_3$ mixing ratios are typically at a minimum in the early morning, peak near mid-day, and decrease during the night (e.g., [255]). On the flip side, Alkezweeny et al. [256] were among the earliest to demonstrate lower daytime[NH$_3$], as the nighttime shows shallower boundary layer depth. Erisman et al. [257] lent further evidence of this by observing a strong decrease in [NH$_3$] with height, which was steeper at night likely due to temperature inversions preventing the upward transport of NH$_3$.

Delving into this discrepancy, Buijsman et al. [258] were able to identify the expected diurnal variation occurring in low NH$_3$ emission sites and the atypical elevated nighttime concentrations over high emission areas. The relatively lower daytime [NH$_3$] in high emission areas was due to higher wind speeds and more favorable mixing conditions, while at night there would be an accumulation in the shallow boundary layer. For background stations, the opposite observations were due to the transport of NH$_3$ from emission areas; nighttime removal of NH$_3$ through dry deposition and conversion exceeded the transport contribution. During daytime, [NH$_3$] increased as atmospheric conditions permitted the vertical transport of tropospheric NH$_3$. Additionally, the difference in [NH$_3$] between day and night is larger during spring and summer [259], suggesting the influence of higher emissions during warmer months. No clear diurnal profile of [NH$_3$] was observed in a study by Parmar et al. [260] for different seasons in an urban area with elevated [NH$_3$]. Early morning decrease in [NH$_3$] was attributed to dew formation, which is a significant sink for soluble gases [261].

Perrino et al. [262] noted that NH$_3$ in an urban location was much higher than at a nearby rural location and higher than an urban background station with no vehicular traffic. Additionally, the concentration and temporal trend of NH$_3$ and CO were well correlated, indicating NH$_3$ may originate from vehicular emissions and its concentration is dependent on the mixing in the atmosphere. Measurements by Ianniello et al. [263] in Beijing, China showed no observed diurnal variability for [NH$_3$] in both summer and winter. The highest [NH$_3$] were observed in the early morning during summer (∼150 ppbv) when atmospheric conditions were stable. The diurnal trends of [NH$_3$] were weakly dependent on air temperature and were affected by wind direction, indicating the influence of local and regional sources. [NH$_3$] showed a correlation with boundary layer mixing and with NO$_2$, CO and PM$_{2.5}$, supporting their hypothesis that vehicular traffic may be a significant NH$_3$ source in Beijing. Similarly, Gong et al. [264] uncovered the contribution of vehicular emissions to the morning rise in the diurnal profile of NH$_3$ mixing ratios only in winter over urban and suburban areas of Texas, US. Notable spikes were likely due to transport from a coal power plant and some other possible sources. Large differences in NH$_3$ diurnal profiles between weekdays weekends were likely due to higher weekday industrial activities. Road traffic was also identified by Pandolfi et al. [265] as a significant source of NH$_3$, with a typical bimodal-traffic-driven NH$_3$ diurnal cycle at an urban site. Significant lowering of mixing height during nighttime in an urban area may lead to higher measured [NH$_3$] during nighttime [266]. Some trends due to daytime increases in transportation were also observed.
Wang et al. [267] observed the diurnal profile of [NH$_3$] in the urban atmosphere over Shanghai, China to demonstrate a typical bimodal cycle. The two modes occurred during morning and evening traffic emissions and were modulated by atmospheric boundary layer development. On the contrary, atmospheric NH$_3$ at a rural site showed a single mode (late morning), primarily due to volatilization from agricultural emissions as temperatures increased. In an industrial area, the diurnal profile of [NH$_3$] was irregular and showed no bimodal or unimodal pattern due to large industrial emission pulses, which were variable and mainly during nighttime.

As evinced by the literature reviewed, the diurnal variation in [NH$_3$] is not straightforward. Conventional wisdom leads to the expectation of a diurnal profile of NH$_3$ increasing from dawn till afternoon and then decreasing due to the effect of temperature on emissions. There may be many other factors at play, such as transport, boundary layer height, deposition, fertilizer application time, traffic emissions and the interplay of all of these that result in unexpected or even opposite diurnal profiles of NH$_3$.

5.2. Seasonal

The numerous studies in Table S1 show maximum (minimum) [NH$_3$] occurring during warm (cold) months. The seasonal cycle in [NH$_3$] is predominantly a result of the temperature dependence of: (1) gas–particle partitioning between NH$_3$ and NH$_4^+$, (2) NH$_3$ emissions from vegetation, organic wastes, and fertilizers due to Henry’s law equilibrium between aqueous and gas phases NH$_3$ [195,268], (3) turbulence, and (4) humidity [269].

Most of the listed observations show the expected maximum of gas-phase [NH$_3$] in the warmest summer months. Robarge et al. [270] examined the various meteorological factors that could affect [NH$_3$], i.e., air temperature, relative humidity, and wind speed and direction. They determined temperature to be the most significant meteorological parameter that determines [NH$_3$]. Bari et al. [271] also demonstrated that the Manhattan summer to winter ratio was 1.5. Anatolaki and Tsitouridou [272] observed that [NH$_3$] were slightly higher during the warm months in Greece. Usually, this is explained by the shift of gas–particle partitioning equilibrium from NH$_4$NO$_3$ towards HNO$_3$ and NH$_3$ at high temperatures [273]. However, in the conditions at their measurement site, particulate NH$_4$NO$_3$ was only expected in the cold period. They suggest that photochemistry for nitric acid and local NH$_3$ emissions from a fertilizer factory or agricultural activities could explain higher [NH$_3$]. This is indicative of the potential of factors other than temperature, most importantly the presence of local sources, to play a role in determining the apparent seasonal variation of [NH$_3$].

An important non-meteorological factor is the application of fertilizers for agriculture, especially in the spring. Hoell et al. [274] were among the earliest to identify enhanced NH$_3$ levels in March, possibly due to NH$_4$NO$_3$ volatilization from fertilizer. This springtime application of fertilizer may cause deviation from the ammonium nitrate equilibrium constant due to local sources and non-equilibrium conditions caused by high RH and the presence of sulfate acid aerosol, as observed by Cadle et al. [275].

However, Burkhardt et al. [276], noted that local sources could possibly explain the maximum seasonal arithmetic mean [NH$_3$] in spring and autumn, but the seasonal geometric means [NH$_3$] were largest in summer. Measurement of [NH$_3$] is generally reported as an arithmetic mean, which is affected to a higher degree by spikes associated with local sources, whereas the geometric mean is more effective at representing background [NH$_3$]. Thus, summertime NH$_3$ is in reality larger than spring and autumn values, where spikes associated with local agricultural emission sources appear to elevate the background [NH$_3$].

Alebic-Juretic [277] showed that in a residential area, within a cultivated garden, the seasonal maxima in [NH$_3$] are obtained during the warmer months of spring and summer. However, near industrial sources, higher [NH$_3$] is seen in winter and autumn. This is indicative of the role of the boundary layer depth in the high NH$_3$ background in industrial source regions and the role of emissions from the green space in the lower NH$_3$ background residential region.
These numerous studies and others listed in Table S1 demonstrate that seasonal variation of NH$_3$ is, as expected, correlated with variation of temperature. It is most concentrated in the summer months and decreases as it gets colder. There may, however, be the effect of boundary layer height: in the winter, a lower boundary height for mixing of NH$_3$ means more concentration. Deviations are also usually observed in spring, where it may be higher than in summer, during the period of manure and fertilizer application. The dependence on emissions also extends to the type of source. When there is a constant source of emissions, such as an industrial area, maximum [NH$_3$] may occur in winter due to the shallower atmospheric boundary layer. A key consideration to make is that the arithmetic seasonal mean may be greatly affected by outlier NH$_3$ emission events, deeming the geometric mean as more representative of background concentrations.

5.3. Long-Term Trends

Although the effect of temperature and relative humidity is evident in the temporal trends discussed thus far, in the long term, due to the expected smoothing out of meteorological factors, long-term trends in NH$_3$, if they exist, are mainly due to other factor(s). There have been observations of an increasing trend in NH$_3$ levels in the atmosphere in recent years, despite almost constant or reducing emissions. It is likely that the changing chemical environment due to reducing acidic gases (SO$_2$ and NO$_x$) means that more NH$_3$ remains in the gas-phase with reduced available reducible species to form NH$_4^+$ in the particle phase.

There are numerous studies (see Table S1) indicating the long-term trends in [NH$_3$] over North America. The concentrations of NH$_3$ have been increasing across the US despite the nearly constant emission of NH$_3$. Butler et al. [56] using AMoN and Yao and Zhang [278] using NAPS, CAPMoN and AMoN network data provided evidence for this increase. Analysis of satellite (AIRS)-derived NH$_3$ retrievals [105] shows that [NH$_3$] has increased by 0.056 ± 0.012 ppbv yr$^{-1}$ (≈2.61 % yr$^{-1}$) over the US from 2002–2016. Ground-based measurements in Toronto, Canada showed no significant increasing trend in 2003–2011 [279] and an increasing (≈20%) trend [278]; however, some sites in the US showed an increasing trend by up to 200%. Over the US, Yu et al. [157] examined long-term model simulated surface [NH$_3$], validated with network measurements. Their observation of an increasing long-term trend of [NH$_3$] was demonstrated to be due to the decreasing emissions of SO$_2$ and NO$_x$ contributing to roughly 2/3 and 1/3 of its increase over the US, respectively.

In China, the burgeoning population leads to increasing demand for animal and agricultural products. To meet this demand, there has been a sharp increase in the use of fertilizers for agriculture and concentrated animal feeding operations for producing livestock. This has led to a sharp increase of NH$_3$ emissions in China, especially in the North China Plain (NCP). A long-term record of NH$_3$ is lacking for the nation. However, analysis of satellite (AIRS)-derived long-term NH$_3$ retrievals [105] shows that NH$_3$ concentrations have increased over China by 0.076 ± 0.020 ppbv yr$^{-1}$ (≈2.27 % yr$^{-1}$).

In the European Union, NH$_3$ emissions fell by 23% between 1990 and 2015 but increased between 2014 and 2015 by 1.8%, mainly because of increases in Germany, Spain, France, and the United Kingdom. In Germany, there has been a rising trend in NH$_3$ emissions, especially in the period since 2009. This is attributable mainly to inorganic nitrogen fertilizers. It must be noted that among the main pollutants in the EU, [NH$_3$] showed the least reduction (23%).

One of the longest and earliest long-term records of [NH$_3$] measurements, determined spectrophotometrically by Nesslerization, was available in Rijeka, Croatia from 1983. Alebic-Juretic [277] examined this record for 1983–2005 and the long-term trend of gas-phase NH$_3$ showed a weak declining trend in two sites in the vicinity of the city over the period, despite an estimated emissions reduction of >20%. Analysis of the 25-year long-term measured [NH$_3$] in Hungary by Horvath et al. [117] showed no decrease even in the period of large NH$_3$ emission reduction (from 1989); in fact, a small increase was observed. Over the United Kingdom, similar observations were made by Tang et al. [280] for the period from September 1996 to December 2005 despite a ≈12% reduction in emissions during 1990 to 2004. NH$_3$ emissions in Sweden decreased by 20.6% during 1993–2009,
and yet Ferm and Hellsten [281] observed an increase in [NH₃]. The strict control on NH₃ emissions in the Netherlands saw a decrease in [NH₃] for 1993–2014, but there has been a subsequent increase despite emission reduction [148]. An analysis of satellite (AIRS)-derived NH₃ retrievals [105] showed that NH₃ concentrations have increased over Western Europe by 0.053 ± 0.021 ppbv yr⁻¹ (~1.83 % yr⁻¹).

Sutton et al. [282] posited that interactions with SO₂, which has shown decreasing concentrations, mask the expected decrease in [NH₃] due to slower rates of conversion from NH₃ to NH₄⁺ leading to longer atmospheric residence time of the gaseous NH₃. This is the strongest reason explaining the various observed temporal variations of [NH₃]. Additionally, reduced acidic species, through an increase of cuticular resistance for NH₃ uptake, can limit the potential for co-deposition (with SO₂) and therefore reduce the dry deposition velocity, which leads to increased [NH₃]. Recent examinations of measurements, remote sensing, and modeling data [98,105,148,156,157,169,250] provide evidence for the effect of reducing acidic precursor gases explaining most of the observed increases in [NH₃] despite its reduced/constant emissions in certain regions.

The temporal variation in [NH₃] is therefore highly dependent on the sources of emission and its temperature dependence. Warmer periods see elevated [NH₃]. Wetter periods see reduced [NH₃] due to lower temperatures and increased deposition. However, in the long term, these meteorological effects are smoothed out. Additionally, due to generally reducing emissions, there should be a negative trend in [NH₃]. The studies detailed above investigated the observed opposite trend of NH₃ concentration and emission and substantial evidence was presented for the significance of the changing chemical environment due to pollution-control strategies.

6. Conclusions and Research Needs

This paper reviewed around 540 publications dealing with the quantification of atmospheric ammonia concentrations in the atmosphere through in situ measurements, satellite remote sensing inference, and model simulations. We summarize key points in line with the aims established at the beginning of this review:

NH₃ has been in the spotlight considering its increasing concentrations despite reducing emissions over most regions of the globe. It is important to examine this chemical species due to its role in PM₂.₅ formation, and mainly in the formation of NH₄NO₃. Its role in atmospheric new particle formation is of special interest as well. The main determinant of the concentration of ammonia in the atmosphere is its emissions. Due to the short lifetime of the gas-phase form, NH₃ is concentrated over regions of intensive agriculture and concentrated animal feeding operations, such as the North China Plain, the US Midwest, the Indo-Gangetic plains, and pastoral lands of Europe. In some of these regions, [NH₃] can exceed 40 ppbv.

The variations in [NH₃] are expected to be temperature-dependent—a virtue of its strong dependence on emission. Typically, at the diurnal scale, [NH₃] varies with temperature, which is generally a function of insolation. There may be effects of vehicular emissions in pushing up [NH₃] and creating a bimodal daily cycle. However, many other factors, such as transport, boundary layer height, deposition, fertilizer application time, traffic emissions, and their interactions may result in unexpected variations of its atmospheric concentration. Seasonal and inter-annual variations are heavily meteorology dependent—warmer periods have higher [NH₃] and wetter/colder periods will have lesser [NH₃]. When examining long-term trends, which have been increasing over the last several years in most regions, there is mounting evidence for the importance of the chemical environment in determining the concentration of NH₃ in the atmosphere. In the long term, the effect of meteorology is generally smoothed out and these long-term trends are mainly dictated by the chemical environment. Due to stringent regulations for acid precursor gases (SO₂ and NOₓ) in most parts of the world, as well as the comparatively constant NH₃ emissions, less NH₃ is taken up into the particle phase. Thus, the concentration of NH₃ is increased in the atmosphere. Sutton et al. [282] additionally suggests that
dry deposition velocity is reduced due to reduced acidic precursor gases’ concentrations limiting the potential for co-deposition.

The quantification of $[\text{NH}_3]$ through in situ observations, satellite remote sensing, and modeling comes with certain caveats. The main issue with in situ measurement of $[\text{NH}_3]$ at the surface is the high cost of comprehensive spatiotemporal coverage. $[\text{NH}_3]$ is highly spatially variable, requiring any monitoring network to have a dense distribution of measurement stations. Online analyzers, while ideal, are not cost-effective to implement. Furthermore, there may be a non-linear negative bias in measured $[\text{NH}_3]$ due to its stickiness (polar nature) to instrument surfaces. There is, therefore, a dearth of in situ measurements over remote areas on land, and especially over the oceans. Satellite-based instruments are currently unable to resolve this gap due to the non-optimal thermal contrasts of the oceans for inference of ammonia mixing ratios from measured spectral radiances. Satellite remote sensing approaches need to be tuned specifically for the measurement of $[\text{NH}_3]$. Inference made from spectral radiances may be erroneous in circumstances such as nighttime and times with cloudiness. The assumed fixed vertical $\text{NH}_3$ profile for the conversion of radiances to $[\text{NH}_3]$ is problematic as well. There are other issues, such as the fact that most remote sensing data are not from a geostationary constellation that provides continuous global coverage. Furthermore, the spatial resolution is on the scale of several kilometers in diameter, a scale over which there can be significant variability in $[\text{NH}_3]$. Successful modeling is highly dependent on the accurate representation of the processes discussed in Section 3. Modeling of $[\text{NH}_3]$ in the atmosphere over oceans is non-optimal due to negligible empirical datasets for validation, uncertain marine emissions, photolysis of dissolved organic nitrogen in the surface water or in the atmosphere [154], and different chemical environments (more acidic aerosols, more fine mode particles, sea salt alkalinity) affecting partitioning. Heretofore, the ammonia concentration over oceans (pptv levels) and remote areas (low ppbv levels) was considered insignificant. New research Yu et al. [5] suggests that nucleation is enhanced in the presence of pptv levels of atmospheric $\text{NH}_3$, making it important to understand ammonia over regions of its low concentration. Despite these issues, the three varied approaches compensate for each other’s limitations to a fair degree and continue improving. A synergistic approach of measurements↔satellite-inference↔modeling is the current research need, which will contribute towards improved understanding of ammonia in the atmosphere.

**Supplementary Materials:** The following are available online at http://www.mdpi.com/2073-4433/11/10/1092/s1, Table S1: In situ measurements of $[\text{NH}_3]$, Table S2: Gas–Aerosol Equilibrium Models used in Chemical Transport Models.

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