

UK ammonia emissions estimated with satellite observations and GEOS-Chem

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Key Points:

- Satellite observations of NH₃ from 2 sensors (IASI, CrIS) are used to estimate UK NH₃ emissions in Mar-Sep at fine scales (10 km, monthly)
- Satellite-derived NH₃ emissions total 272 Gg from IASI and 390 Gg from CrIS and exhibit a spring (April) and summer (July) peak
- Bottom-up inventories used for research and policy are 27-49% less than the satellite-derived estimates and miss the summer emissions peak

Abstract

Agricultural emissions of ammonia (NH_3) impact air quality, human health, and the vitality of aquatic and terrestrial ecosystems. In the UK, there are few direct policies regulating anthropogenic NH_3 emissions and development of sustainable mitigation measures necessitates reliable emissions estimates. Here we use observations of column densities of NH_3 from two space-based sensors (IASI and CrIS) with the GEOS-Chem model to derive top-down NH_3 emissions for the UK at fine spatial (~ 10 km) and time (monthly) scales. We focus on March–September when there is adequate spectral signal to reliably retrieve NH_3 . We estimate total emissions of 272 Gg from IASI and 390 Gg from CrIS. Bottom-up emissions are 27% less than IASI and 49% less than CrIS. There are also differences in seasonality. Top-down and bottom-up emissions agree on a spring April peak due to fertilizer and manure application, but there is also a comparable summer July peak in the top-down emissions that is not in bottom-up inventories and appears to be associated with dairy cattle farming. We estimate relative errors in the top-down emissions of 11–36% for IASI and 9–27% for CrIS, dominated by column density retrieval errors. The bottom-up versus top-down emissions discrepancies estimated in this work impact model predictions of the environmental damage caused by NH_3 emissions and warrant further investigation.

Plain Language Summary

Emissions of ammonia, mostly from agriculture, are often a dominant contributor to fine particles in countries with well-established policies that have led to large reductions in other precursors of such pollutants detrimental to our health. Here we use a model and observations of ammonia from two space-based sensors to estimate emissions in the UK where there are no direct policies regulating agricultural sources of ammonia. The satellite-derived emissions, limited to March–September when conditions are ideal for viewing ammonia from space, total 272 kilotonnes from an instrument that passes overhead in the morning and 390 kilotonnes from an instrument with a midday overpass. Though the emissions estimates differ for the two instruments, both exhibit a spring (April) peak due to fertilizer and manure use and summer (July) peak likely associated with dairy cattle farming. The summer peak is missing in bottom-up emission inventories and total March–September emissions from these inventories are also 27–49% less than those derived with satellites. Further research is needed to address these discrepancies, as such inventories are widely used for developing policies and assessing environmental damage caused by ammonia.

1 Introduction

Agricultural practices such as synthetic fertilizer and manure use and livestock farming release large quantities of ammonia (NH_3) to the atmosphere. Once emitted, NH_3 partitions to acidic aerosols to form ammonium that contributes to mass concentrations of fine particles ($\text{PM}_{2.5}$) hazardous to health (Cohen et al., 2017; Dockery et al., 1993; Vohra et al., 2021a). NH_3 and ammonium also deposit to the Earth's surface and drastically alter the natural nitrogen balance of terrestrial and aquatic ecosystems (Galloway, 1998; Johnson & Carpenter, 2010; Vitousek et al., 1997).

In the UK, agriculture is the dominant ($>80\%$) source of NH_3 emissions (Ricardo, 2018a), mostly from nitrogen fertilizer use, manure management, and farming of dairy and beef cattle (DEFRA, 2019). Modelling studies suggest that the largest and most extensive decline in $\text{PM}_{2.5}$

in the UK would be achieved by targeting NH_3 sources (Vieno et al., 2016), but only large pig and poultry farms are required to adopt best practices and technologies that reduce NH_3 emissions (DEFRA, 2019). There are additional policy options under consideration, such as limiting the use of solid urea fertilizer, a large source of NH_3 in the UK (DEFRA, 2020a). As a signatory of the United Nations Economic Commission for Europe (UNECE) Gothenburg protocol, the UK is committed to an anthropogenic NH_3 emissions ceiling of 297 Gg, informed by annual emissions estimates from the UK bottom-up National Atmospheric Emissions Inventory (NAEI). The UK is also required as part of the protocol to reduce NH_3 emissions by 8% in 2020 and beyond relative to emissions in 2005 (UNECE, 2019). The estimated decline in NH_3 emissions from 1980 to 2017 is $0.2\% \text{ a}^{-1}$ due to a steep decline in vehicular emissions of NH_3 in 1998-2007 and a recent increase in agricultural emissions since 2013 (Ricardo, 2020). Any future policies targeting NH_3 emissions would also need to consider increases in emissions as the atmosphere warms (Sutton et al., 2013).

Estimates of the contribution of NH_3 emissions to $\text{PM}_{2.5}$ and mobilization of nitrogen in aquatic and terrestrial ecosystems, assessment of attainment of UNECE emissions ceilings and trends, and decisions on effective mitigation measures demand accurate estimates of NH_3 emissions. The NAEI of annual total and mapped UK NH_3 emissions is published each year. These are at high spatial resolution (1 km) and incorporate detailed information about the farming activities that contribute to NH_3 emissions. The ability to validate the inventory is challenging, as there are no long-term measurements of NH_3 fluxes. There is a network of very reliable measurements of rural 24-hour mean surface concentrations of NH_3 that cover the full latitudinal extent of the UK from Cornwall in the south to Shetland in the north (Tang et al., 2018), but there are large monitoring gaps in-between. Satellite observations of NH_3 retrieved from infrared spectral measurements offer complete coverage of the UK and routine daily measurements in the absence of clouds and under good retrieval conditions. Satellites observe NH_3 molecules throughout the atmospheric column, but the majority are within the planetary boundary layer and most of the variability in the column is typically due to NH_3 at or near the surface (Clarisse et al., 2010; Nowak et al., 2010; Schiferl et al., 2016; Vohra et al., 2021b).

Retrieval of NH_3 from space-based instruments was first described by Beer et al. (2008) for the Tropospheric Emission Spectrometer (TES) instrument. Satellite NH_3 retrieval products have since undergone substantial retrieval development (Clarisse et al., 2009; Shephard et al., 2011; 2020; Shephard & Cady-Pereira, 2015; Van Damme et al., 2014b; 2017; 2021; Whitburn et al., 2016a), intercomparisons (Dammers et al., 2019), and validation against ground-based observations of total atmospheric column densities and surface concentrations of NH_3 (Dammers et al., 2016; 2017; Van Damme et al., 2015a; Vohra et al., 2021b). These products have also seen extensive use in characterizing NH_3 emissions. This includes detecting global and regional NH_3 emission hotspots (Cady-Pereira et al., 2017; Clarisse et al., 2019; Dammers et al., 2019; Shephard et al., 2020; Van Damme et al., 2018), constraining NH_3 emissions from biomass burning (Adams et al., 2019; Whitburn et al., 2016b), evaluating regional emission inventories (Chen et al., 2021; Fortems-Cheiney et al., 2020), identifying underestimated or missing NH_3 sources in widely used global and regional emission inventories and models (Heald et al., 2012; Hickman et al., 2018; Van Damme et al., 2014a), and determining long-term local and regional trends and variability in NH_3 (Hickman et al., 2020; Van Damme et al., 2015b; 2021; Vohra et al., 2021b; Wichink Kruit et al., 2017).

Here we use satellite observations of NH_3 and the GEOS-Chem chemical transport model (CTM) to derive top-down NH_3 emissions for the UK and evaluate the bottom-up NAEI emissions inventory and current understanding of seasonality in emissions. This includes the use of surface observations from the UK monitoring network to evaluate GEOS-Chem and corroborate findings from the satellite observations.

2 Space-based observations of column densities of NH_3

Satellite observations of NH_3 retrieved in the infrared portion of the light spectrum rely on the spectral signal that depends on the atmospheric state, such as abundance and vertical distribution of NH_3 and thermal contrast between the surface of the Earth and the overlying atmosphere (Clarisse et al., 2010; Shephard et al., 2011). Two prominent products are available from contemporary space-based instruments that pass overhead in the morning (the Infrared Atmospheric Sounding Interferometer or IASI) and midday (the Cross-track Infrared Sounder or CrIS). These products use distinct retrieval approaches for us to assess the constraints these provide on NH_3 emissions in the UK.

2.1 Infrared Atmospheric Sounding Interferometer NH_3

The IASI instrument onboard the Metop-A satellite was launched into low-Earth polar sun synchronous orbit in October 2006. The instrument has two overpass times in the morning (09h30 local solar time or LST) and at night (21h30 LST), providing global coverage twice a day. The elliptical IASI pixels range in ground pixel resolution from $12 \text{ km} \times 12 \text{ km}$ at nadir (directly below the instrument) to about $20 \text{ km} \times 39 \text{ km}$ at the edges of the 2200-km-wide swath (Clarisse et al., 2011). The data product we use is the Level 2 cloud-free reanalysis product of total column NH_3 (version 3R-ERA5) (Van Damme et al., 2021). The retrieval uses machine learning, specifically a neural network trained relationship between column NH_3 and a so-called hyperspectral range index or HRI, where the HRI is a measure of the relative enhancement in the spectral signature due to NH_3 (Van Damme et al., 2014b; 2017; Whitburn et al., 2016a). The data product includes reported retrieval errors estimated by perturbing individual input parameters in the neural network framework (Whitburn et al., 2016a). The neural network retrieved product has been validated against global and regional networks of ground-based NH_3 observations of surface concentrations and column densities (Dammers et al., 2016; Guo et al., 2021; Vohra et al., 2021b; Whitburn et al., 2016a). In general, IASI NH_3 reproduces the temporal variability in surface concentrations of NH_3 , but exhibits a low bias of 25-50% (Dammers et al., 2017; Whitburn et al., 2016a).

We use daytime (09h30 LST) IASI NH_3 for 2008-2018 to obtain multiyear monthly means. This dampens influence of interannual variability and ensures consistency with NAEI NH_3 emissions that are estimated with 30-year mean meteorology (Ricardo, 2019b). We grid the data to finer spatial resolution ($0.1^\circ \times 0.1^\circ$) than the native resolution of the instrument using the tessellation oversampling technique described in Zhu et al. (2017) and Sun et al. (2018). This takes advantage of the spatial variability in coverage of individual orbits and the long data record from IASI to reduce noise and smooth out spatial gradients in the gridded product (Sun et al., 2018). Briefly, tessellation involves weighting individual IASI pixels by the area of overlap with the target grid and also includes error-weighting using the reported retrieval error. In our application of the tessellation gridding technique, we approximate the area of IASI pixels as a

quadrilateral polygon, where the corners of each polygon are estimated as the distance midway between the centres of neighbouring IASI pixels.

Retrieval of NH_3 over the UK is challenging, due to persistent clouds and relatively cool conditions. Extreme retrievals, identified as absolute columns $> 5 \times 10^{17}$ molecules cm^{-2} , are removed. We also exclude IASI NH_3 columns retrieved on 26-27 July 2018, coincident with the summer 2018 heat wave (McCarthy et al., 2019). Record high temperatures ($> 30^\circ\text{C}$) lead to UK IASI NH_3 column densities 4-times greater ($\sim 4 \times 10^{16}$ molecules cm^{-2}) than the UK July multiyear mean ($\sim 1 \times 10^{16}$ molecules cm^{-2}) and cause an 11% positive bias in the July multiyear mean. A similarly large influence of heat waves on IASI NH_3 columns resulted from the summer 2010 heat wave over mainland Europe (Van Damme et al., 2014a). Following oversampling, the gridded multiyear means with large relative error ($> 50\%$) are removed. This leads to loss of the majority of IASI NH_3 columns in October-February, so only March-September multiyear means are considered. According to the monthly NH_3 emissions estimated by Hellsten et al. (2007), March-September accounts for $\sim 60\%$ of the annual total. Additional outlier filtering is applied to the gridded multiyear monthly means to remove columns $< -1 \times 10^{16}$ molecules cm^{-2} and $> 1 \times 10^{17}$ molecules cm^{-2} . These account for $< 0.1\%$ of the March-September data.

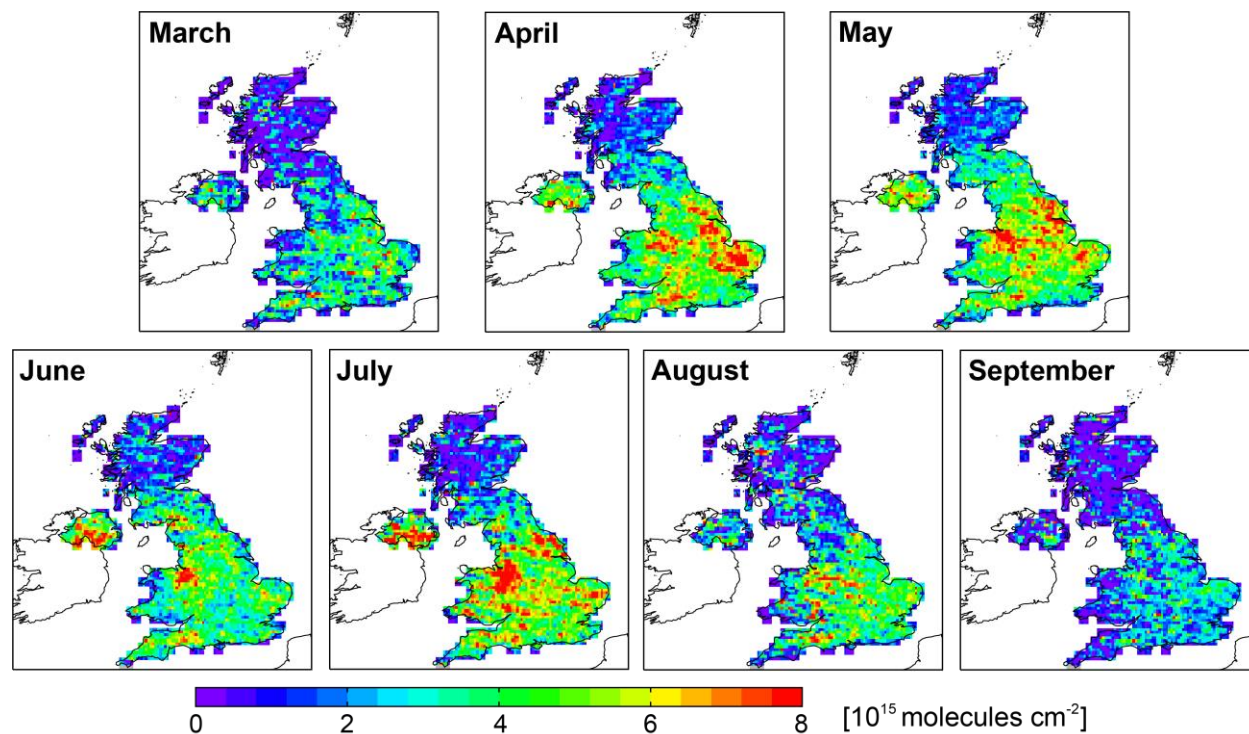


Figure 1. Monthly multiyear (2008-2018) mean IASI NH_3 . Data are gridded to $0.1^\circ \times 0.1^\circ$ using oversampling (see text for details). Grey grids have < 10 observations.

Figure 1 shows the gridded March-September multiyear monthly mean IASI NH_3 columns. The number of observations in each grid ranges from 11 to 128. Values over Scotland are very low (typically $< 2 \times 10^{15}$ molecules cm^{-2}) due to weak signal and lower agricultural

activity than the rest of the UK. The range in IASI NH_3 over the rest of the country of $4\text{--}8 \times 10^{15}$ molecules cm^{-2} is much less than the NH_3 hotspots in other parts of the world. Columns over global hotspots such as North China, West Africa, the Po Valley (Italy), and the Indo-Gangetic Plain (India) exceed 2×10^{16} molecules cm^{-2} (Cady-Pereira et al., 2017; Dammers et al., 2019; Van Damme et al., 2014a; 2018). These are associated with industrial and agricultural activity in India and China and intense seasonal open burning of biomass in West Africa and northern India, aided by warm temperatures increasing NH_3 emissions and enhancing the spectral signal.

2.2 Cross-track Infrared Sounder NH_3

The first CrIS sensor launched into low-Earth polar sun synchronous orbit in October 2011 is onboard the NOAA Suomi-NPP satellite. Like IASI, CrIS observes the Earth twice daily, though in the early afternoon (13h30 LST) and after midnight (01h30 LST) (Goldberg et al., 2013). It has the same swath width as IASI and similar ground pixel resolution (14 km circular pixels at nadir). The fast physical retrieval (CFPR) approach used to retrieve NH_3 columns is described in detail in Shephard & Cady-Pereira (2015) and Shephard et al. (2020). Briefly, it is based on conventional optimal estimation that involves minimizing the difference between observed and calculated outgoing spectral radiances with a priori vertical profiles of NH_3 (Rodgers, 2000). CFPR uses three prior NH_3 profiles representing polluted, moderately polluted, and remote conditions (Shephard et al., 2020) that are selected based on the ammonia spectral signal. This is different to standard optimal estimation that uses prior information that is independent of the observations and imposes spatial and temporal information. The CFPR retrieval generates averaging kernels that quantify the vertical sensitivity of the retrieval. These typically peak between 900 and 750 hPa ($\sim 1\text{--}2.5$ km altitude) (Dammers et al., 2017; Shephard & Cady-Pereira, 2015).

We use the Level 2 CrIS NH_3 CFPR version 1.6 product for 2013–2018. The predecessor product (version 1.5) exhibited a positive bias for $\text{NH}_3 < 1 \times 10^{16}$ molecules cm^{-2} , as values were only retrieved over scenes exceeding the instrument detection limit of $\sim 2 \times 10^{15}$ molecules cm^{-2} (Dammers et al., 2017; Shephard & Cady-Pereira, 2015). This approach filtered out cloud-free scenes below the instrument detection limit and indirectly removed cloudy scenes when the NH_3 signal below clouds could not be detected. In version 1.6 clouds are explicitly identified with information from the space-based Visible Infrared Imaging Radiometer Suite (VIIRS) (White et al., 2021). We use daytime cloud-free CrIS observations with quality flag ≥ 4 (Shephard et al., 2020) and thermal contrast > 0 K, where thermal contrast is the difference between the reported temperatures at the surface and the lowest atmospheric layer. We identify and correct for a positive trend in the CrIS baseline that appears to be erroneous, as it is not apparent in the IASI data. We do this by estimating a statistically significant ($p\text{-value} = 0.02$) increase in monthly mean background NH_3 columns over Scotland (Figure S1) of 2.33×10^{13} molecules cm^{-2} per month (amounting to 1.7×10^{15} molecules cm^{-2} over the whole record) and subtract this from individual CrIS NH_3 column retrievals. We grid the corrected data to $0.1^\circ \times 0.1^\circ$ using the same tessellation code used for IASI, but without error weighting. The individual total column errors include measurement and representative errors and cover a much narrower range (5–55% (Shephard et al., 2020)) than those for IASI (5% to $>100\%$). As a result, higher relative weighting would be applied to low column densities, leading to anomalously low gridded values in the CrIS multiyear means. For consistency with IASI, we only consider March to September.

Figure 2 shows the gridded March-September CrIS NH_3 multiyear monthly mean columns. As with IASI, we filter for outliers in the multiyear means (column densities $< -1 \times 10^{16}$ molecules cm^{-2} and $> 1 \times 10^{17}$ molecules cm^{-2}), removing $< 0.1\%$ of the gridded data. Unlike IASI, the observations during the July 2018 heatwave only increase the July multiyear mean by 1.6%, so these days are retained. The number of CrIS retrievals in each grid ranges from 11 to 96. The CrIS multiyear means are roughly double those for IASI (Figure 1), in part because CrIS passes overhead at midday when higher ambient temperatures lead to greater volatilization of NH_3 . Differences in vertical sensitivity and distinct retrieval approaches likely also contribute. This difference is particularly large in September when background NH_3 is 5.3×10^{15} molecules cm^{-2} more in CrIS than IASI, obtained as the intercept from regressing CrIS against IASI. The spatial correlation between CrIS and IASI multiyear means is $R < 0.5$ in most months (March, June-September), $R = 0.53$ in May, and $R = 0.55$ in April.

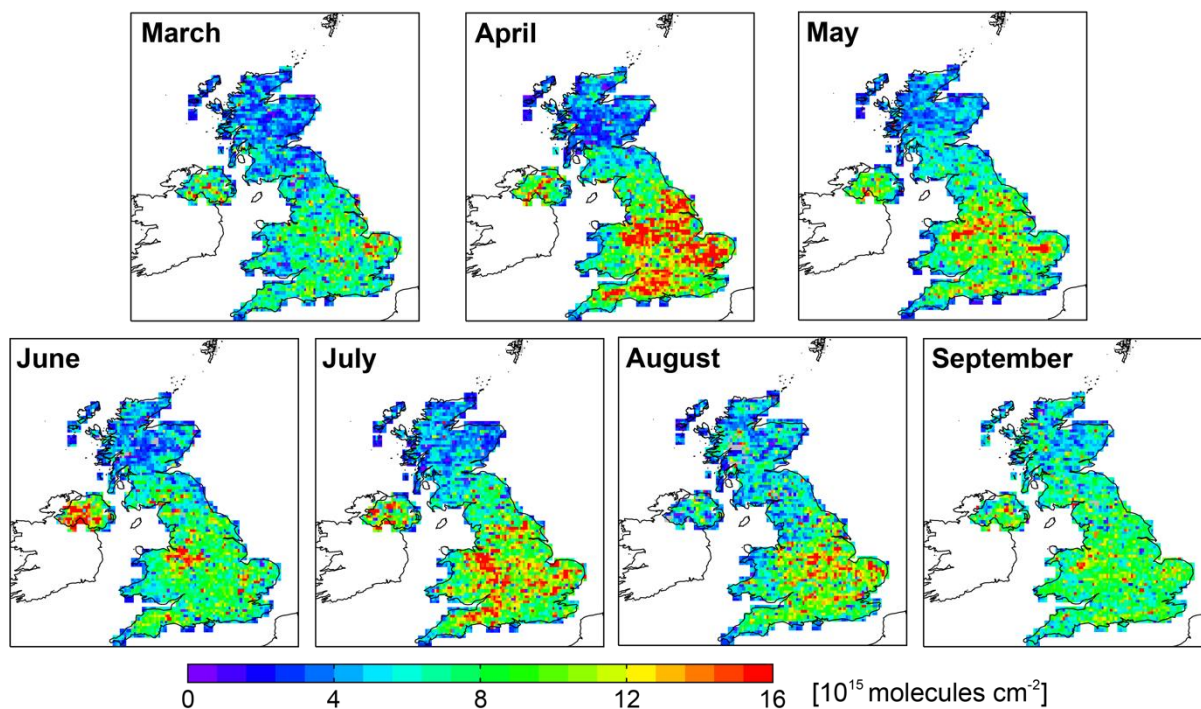


Figure 2. Monthly multiyear (2013-2018) mean CrIS NH_3 . Data are gridded to $0.1^\circ \times 0.1^\circ$ with oversampling (see text for details). Grey grids have < 10 observations.

3 The GEOS-Chem chemical transport model

We use the GEOS-Chem CTM version 12.1.0 (<https://doi.org/10.5281/zenodo.1553349>) to interpret constraints on UK NH_3 emissions from IASI and CrIS. The model is driven with NASA GEOS-FP assimilated meteorology from the Global Modeling and Assimilation Office (GMAO). Model simulations are conducted on a horizontal grid at $0.25^\circ \times 0.3125^\circ$ (latitude \times longitude) nested over western Europe ($32.75\text{--}61.25^\circ\text{N}$, $15^\circ\text{W--}40^\circ\text{E}$). The model extends over 47 vertical layers from the Earth's surface to 0.01 hPa. Dynamic (3-hourly) boundary conditions are from a global GEOS-Chem simulation at $4^\circ \times 5^\circ$.

Anthropogenic emissions over the UK, including from agriculture, are updated in GEOS-Chem to include gridded emissions from the NAEI for 2016 (Ricardo, 2018b). These are annual totals on a $1\text{ km} \times 1\text{ km}$ grid available at <https://naei.beis.gov.uk/data/map-uk-das> (last accessed August 2019). The agricultural NH_3 emissions incorporated in the NAEI are calculated at coarser resolution (5 km) than the NAEI with the nitrogen balance models of Webb & Misselbrook (2004) for livestock sources and Misselbrook et al. (2006) for fertilizer sources. These models are driven with 30-year mean meteorology for 1981–2010, so the NH_3 emissions represent a climatological mean (Ricardo, 2019b). Other anthropogenic NH_3 emissions are typically calculated as the product of emission and activity factors representative of the year of interest and mapped to the 1 km NAEI emissions grid (Ricardo, 2018a). Temporal variability of annual NAEI NH_3 emissions is represented in GEOS-Chem with gridded monthly scaling factors already included in the model and spatially uniform diurnal scaling factors calculated by Zhu et al. (2015) using information about the dependence of NH_3 on aerodynamic resistance, surface temperature and Henry's law. As a result of these, 30% of NH_3 is emitted at midday (noon–2pm LST) coincident with the CrIS overpass and 20% in the morning (9am–noon LST) coincident with the IASI overpass. Monthly scaling factors are from gridded data derived by Friedrich (2000) and lead to peak NH_3 emissions in April. Natural NH_3 sources are from inventories already in GEOS-Chem. These include natural emissions from soils and the ocean from the Global Emissions Initiative (GEIA) inventory (Bouwman et al., 1997) and inland and coastal seabird emissions from the Riddick et al. (2012) inventory. We halve the GEIA inventory emissions, as in Paulot et al. (2014), informed by a 50% overestimate identified by Simpson et al. (1999).

NH_3 is a semi-volatile acid buffer that neutralizes acidic sulfate and nitrate aerosols, so its abundance depends on the abundance of these acidic aerosols. Sulfate forms from oxidation of SO_2 and nitrates from aerosol uptake of nitric acid formed from oxidation of NO_x . The version of the NAEI we use includes outdated mapping of the location of ships and no vertical or temporal information for aircraft emissions. To address these issues, we separate ship and aircraft emissions from other sources in the lumped “Other Transport and Mobile Machinery” category of the NAEI emissions inventory and replace ship emissions with updated estimates that use geospatial information from the automatic identification system (Ricardo, 2017). We convert the NAEI aircraft emissions to monthly estimates and distribute these vertically up to 1 km (the altitude limit of the NAEI emissions) by deriving vertical and temporal scaling factors from the global Aviation Emissions Inventory version 2.0 (AEIv2) used in GEOS-Chem (Stettler et al., 2011). Above 1 km, the AEIv2 emissions are used. The existing temporal scaling factors in GEOS-Chem that are applied to NAEI SO_2 and NO_x emissions lead to peak emissions in winter, due to an increase in energy demand. SO_2 is emitted in the model as 95% SO_2 and 5% sulfate,

using sulfate-to-SO₂ emission ratios for Europe reported by Chin et al. (2000). NAEI emissions are gridded to a uniform 0.1° × 0.1° grid for input to the Harmonized Emissions Component (HEMCO) processing package version 2.1.010 (Keller et al., 2014) that maps all emissions to the model grid and applies relevant scaling factors.

The model includes detailed coupled gas- and aerosol-phase chemistry. Sulfate aerosols are formed in the model from oxidation of SO₂ in the gas phase by OH and in the aqueous phase in clouds by ozone and hydrogen peroxide (Park et al., 2004). Partitioning of NH₃ between the gas and acidic aerosol phase is determined dynamically with the thermodynamic equilibrium model ISORROPIA-II (Fountoukis & Nenes, 2007). Wet and dry deposition, terminal sinks of NH₃, are represented with a standard resistances-in-series scheme for dry deposition (Wesley et al., 1998) and, for wet deposition, includes scavenging in and below clouds (Amos et al., 2012).

We use network site measurements of trace gases and aerosols to evaluate model accuracy at reproducing surface concentrations of NH₃, SO₂, and sulfate. These include 2 rural sites (Auchencorth Moss in Scotland, Chilbolton Observatory in southern England) that form part of the European Monitoring and Evaluation Programme (EMEP) network and the mostly rural UK Eutrophying and Acidifying Atmospheric Pollutants (UKEAP) network. The 2 EMEP sites include hourly measurements from Monitor for AeRosols and Gases in Air (MARGA) instruments (Stieger et al., 2017; ten Brink et al., 2007; Twigg et al., 2015; Walker et al., 2019). The UKEAP network sites include monthly measurements from low-cost denuder filter sampling packs (Tang et al., 2018). The MARGA data are from the EMEP Chemical Coordinating Centre EBAS database (<http://ebas.nilu.no/>; last accessed February 2020) (Tørseth et al., 2012) and the UKEAP data are from the UK-AIR data archive (<https://uk-air.defra.gov.uk/data/data-availability>; last accessed November 2020).

To ensure consistency between the model and observations, the model is sampled from the lowest to the top model layer during satellite overpasses for comparison to IASI (08-11 LST) and CrIS (12-15 LST), and as monthly 24-hour means in the lowest model layer for comparison to the surface observations. The model is sampled in March-September 2016 following a 2-month spin-up for chemical initialization.

4 UK bottom-up emissions of NH₃

Figure 3 shows the spatial distribution of annual UK NH₃ emissions for 2016 from the NAEI. Table 1 gives the breakdown by sector. Annual emissions for 2016 total 298 Gg, mostly (84%) from agriculture. Natural emissions of 21.6 Gg (7% of the total) are consistent with annual total natural emissions in GEOS-Chem of 21.8 Gg. According to GEOS-Chem, these include soils, vegetation and the ocean (together 18.7 Gg) and seabirds (3.10 Gg). NAEI anthropogenic NH₃ emissions total 276 Gg, 21 Gg less than the UNECE Gothenburg protocol emissions ceiling of 297 Gg (UNECE, 2019). The NAEI version we implement in GEOS-Chem and evaluate against top-down estimates was released in 2018. Two NAEI versions have been released since. Reported differences in NH₃ emissions across these versions for consistent years is minor, just 1-3% (Ricardo, 2019a; 2020).

The spatial patterns in Figure 3 coincide with farming activities that dominate NH₃ emissions according to the modelling study by Hellsten et al. (2008) that uses the same Webb &

Misselbrook (2004) nitrogen balance model as the NAEI. Dominant agricultural sources include sheep farming along the Welsh border where emissions are low, and large sources like pig and poultry farming and fertilizer use in east England and dairy and beef cattle farming in west England and Northern Ireland. Hellsten et al. (2008) used agricultural activity data for 2000. Detailed geospatial farming activity data is proprietary and publicly available data are limited to decadal maps of farming activities in England for 2000 and 2010 and annual regional and national statistics. The decadal maps suggest that locations of intensive crop and livestock farming in England are relatively unchanged (DEFRA, 2016b; a). The regional statistics document large changes in the number of livestock and the amount of nitrogen fertilizer used from 2000 to 2016 that would affect trends in emissions. In general, livestock numbers in the UK have declined by 20% for sheep, 11% for dairy and beef cattle, and 25% for pigs (DEFRA, 2020b). Poultry, specifically table chickens, have increased by 10% in the UK, with the largest increase of 42% in Northern Ireland (DEFRA, 2020b). Nitrogen-based fertilizer usage, a dominant NH_3 source in east England (Hellsten et al., 2008), declined by 19% in the UK, ranging from a 3% increase in Scotland to a 37% decrease in Northern Ireland (AIC, 2020).

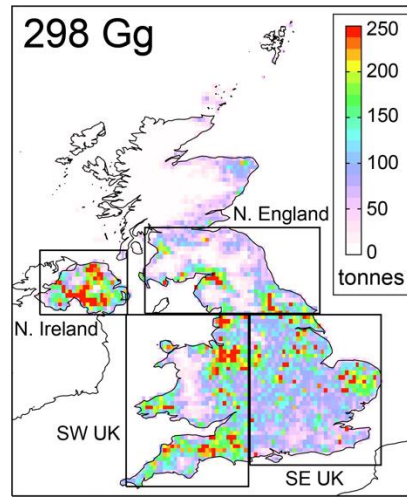


Figure 3. Annual UK NH_3 emissions for 2016. Data are in tonnes per year from the NAEI gridded to $0.1^\circ \times 0.1^\circ$. Inset value is the UK annual total. Boxes demarcate regions with broadly similar NH_3 sources: Northern Ireland (N. Ireland), Northern England and a portion of southern Scotland (N. England), southwest UK (SW UK), and southeast UK (SE UK).

Table 1. UK sector emissions of NH_3 according to the NAEI ^a

| Sources | NH_3 [Gg a^{-1}] |
|----------------------|--------------------------------------|
| Agriculture | 248.9 |
| Natural ^b | 21.6 |
| Waste | 14.2 |
| Point sources | 4.4 |
| Road transport | 4.4 |

| | |
|--------------------|--------------|
| Other ^c | 4.2 |
| Total | 297.7 |

^a Spatial distribution of UK NAEI NH₃ emissions are in Figure 3. ^b Contributors to natural emissions, according to GEOS-Chem, are soils, vegetation and the ocean (together 18.7 Gg) and seabirds (3.1 Gg). ^c Other is industrial and domestic combustion (2.9 Gg) and solvent use (1.3 Gg).

Inversion of column densities of NH₃ to estimate top-down surface emissions can be complicated by dependence of NH₃ abundance on acidic sulfate aerosols formed from oxidation of SO₂. UK SO₂ emissions are dominated by large industrial and energy sector point sources, ships, domestic and industrial combustion, and traffic (Ricardo, 2018a). We find that monthly mean March-September 2016 GEOS-Chem SO₂ concentrations driven with the NAEI are much greater than those measured at EMEP and UKEAP network sites (Figure S2). The model normalized median bias is >500% for modelled SO₂ > 2 µg m⁻³ at sites influenced by point sources in Yorkshire and ~280% for modelled SO₂ < 2 µg m⁻³. Modelled sulfate is also greater than the observations (normalized mean bias or NMB of 17%) (Figure S2). This would enhance partitioning of NH₃ to acidic aerosols to form ammonium, leading to a positive bias in the relative amount of NH_x (NH₃ + ammonium) present as ammonium.

Positive model biases in both SO₂ and sulfate (Figure S2) suggest an overestimate in NAEI SO₂ emissions, though other factors may contribute. These include, but are not limited to, misrepresentation of the height at which SO₂ is emitted from tall stacks, a reported positive bias in mainland Europe SO₂ emissions (Luo et al., 2020), and uncertainties in wet deposition (Luo et al., 2019). We conducted sensitivity simulations to assess the contribution of these uncertainties to modelled SO₂ and sulfate. Details of these simulations and the effect on SO₂ and sulfate concentrations are in the accompanying Supplementary. The factor we find to have the largest influence relative to the model bias is wet deposition. The more efficient wet deposition scheme of Luo et al. (2019) leads to an 11% decrease in sulfate concentrations.

Errors in NAEI SO₂ emissions could be due to uncertainties in emissions from domestic and industrial biomass combustion. The third of six generating units at the 3.9 GW generating capacity Drax power station in Yorkshire transitioned from burning coal to biomass in 2016 (Simet, 2017). SO₂ emissions from biomass combustion depend on fuel sulfur content and combustion efficiency. Reported emission factors range widely from 1 to 110 mg SO₂ MJ⁻¹ (Boersma et al., 2008; Paulrud et al., 2006) and so offer limited constraints. To reduce the influence of a possible bias in SO₂ emissions on GEOS-Chem simulation of abundance of sulfate and NH₃, we decrease land-based gridded (0.1° × 0.1°) NAEI SO₂ emissions by a factor of 3 for grids dominated by point sources (identified as grids with SO₂ emissions > 10 g m⁻² a⁻¹) and by a factor of 1.3 for all other land-based grids. This reduces the original NAEI SO₂ emissions over land by 49% from 164 Gg to 84.1 Gg. With shipping, the updated annual NAEI SO₂ emissions for the domain shown in Figure 3 total 94.5 Gg. The March-September modelled sulfate NMB changes from +17% (Figure S2) to -8.8%. We use the scaled SO₂ emissions in all subsequent simulations.

5 Top-down NH₃ emissions and comparison to bottom-up estimates

We calculate top-down NH₃ emissions by multiplying the satellite NH₃ multiyear monthly mean columns (Figure 1 for IASI, Figure 2 for CrIS) by GEOS-Chem ratios of 24-hour

mean NH_3 emissions to 3-hour mean columns (08-11 LST for IASI, 12-15 LST for CrIS) after interpolating the model fields to $0.1^\circ \times 0.1^\circ$. The mass-balance approach that we use to infer emissions can be susceptible to spatial misattribution of emissions due to displacement of NH_3 from the source. The global mean lifetime of NH_3 is ~ 15 h (Hauglustaine et al., 2014), ranging from ~ 2 h near large sources (Dammers et al., 2019) to ~ 36 h far from emission sources (Van Damme et al., 2018). The displacement length, the distance for NH_3 to decay to $\sim 63\%$ of the original concentration of the emission source (Marais et al., 2012; Palmer et al., 2003), is similar to the resolution of the satellite-derived emissions (10-12 km) for calm conditions (wind speeds of $5\text{--}6 \text{ km h}^{-1}$) and short NH_3 lifetimes (2 h). Conditions are relatively stable in the UK in summer (Figure A1f.3 of BEIS (2016)), so meteorological mean wind speeds typically reach $\sim 7 \text{ km h}^{-1}$. At these slightly windier conditions and for a longer NH_3 lifetime of 15 h, the displacement length increases to 105 km.

Maps of the resultant top-down monthly NH_3 emissions are shown in Figure 4 for IASI and Figure 5 for CrIS. Emissions for retained grid squares total 271.5 Gg for IASI, whereas these are 44% more from CrIS (389.6 Gg). CrIS monthly emissions are 19-37% more than IASI for March-July, similar in magnitude to the reported 25-50% low bias in IASI columns (Dammers et al., 2017; Whitburn et al., 2016a). The percentage difference increases to 56% for August and $>100\%$ for September. The large difference in September is due to $5.3 \times 10^{15} \text{ molecules cm}^{-2}$ greater background NH_3 in CrIS, even after correcting for the baseline trend (Section 2.2, Figure S1). CrIS emissions excluding September are 34% more than IASI. Qualitatively, both estimates exhibit similar spatial patterns to the NAEI (Figure 3). This includes relatively low emissions along the Welsh border, and peak emissions in Northern Ireland, the northern portion of the English side of the Welsh border, and in Norfolk in the east.

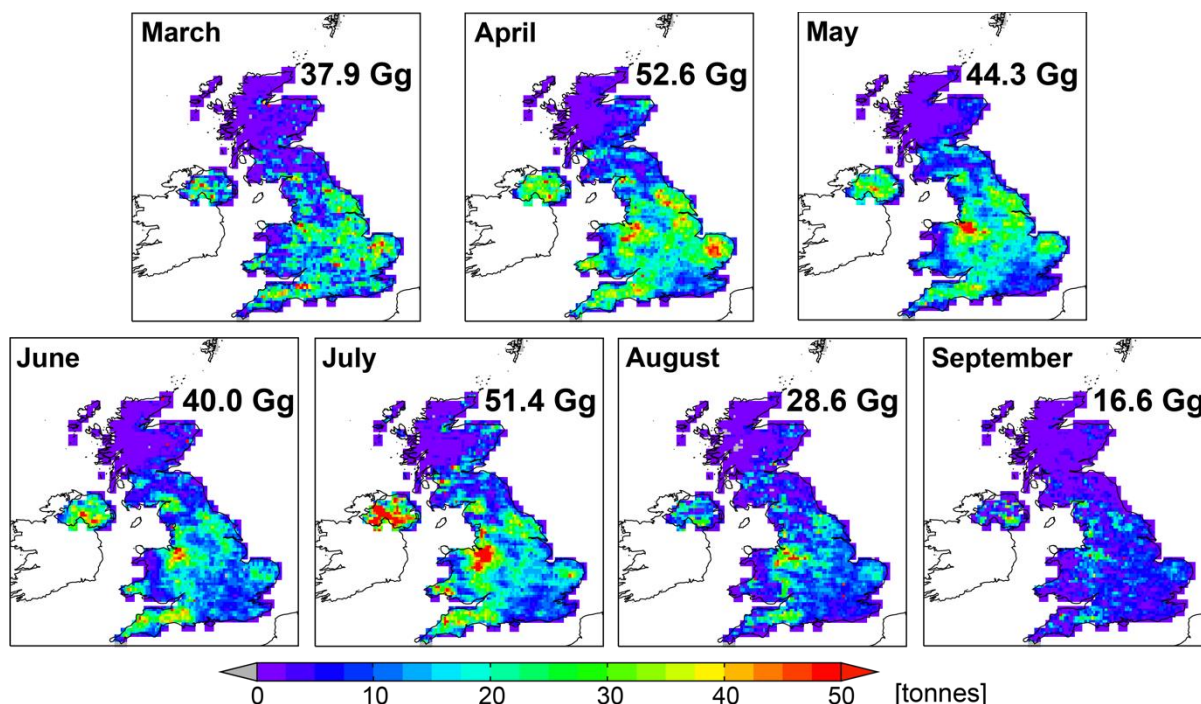


Figure 4. IASI-derived NH_3 emissions for March-September. Maps are at $0.1^\circ \times 0.1^\circ$. Inset values are monthly emissions that sum to 271.5 Gg.

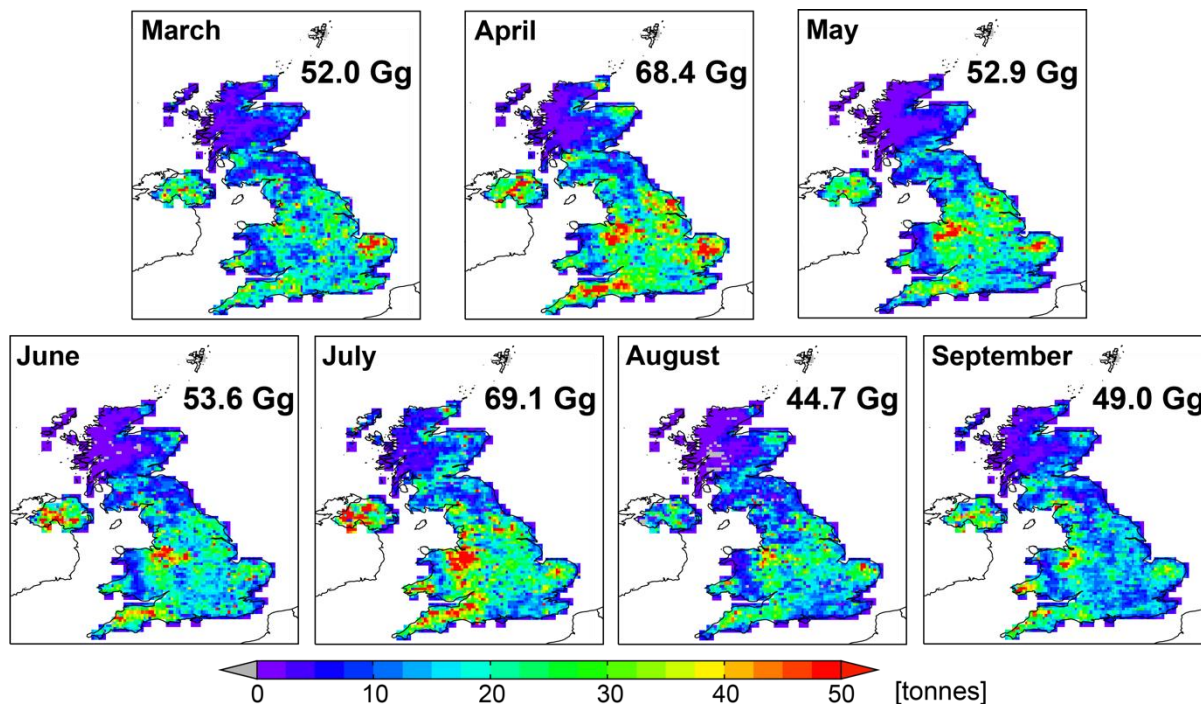


Figure 5. CrIS-derived NH_3 emissions for March-September. Maps are at $0.1^\circ \times 0.1^\circ$. Inset values are monthly emissions that sum to 389.6 Gg.

For comparison of monthly top-down and bottom-up emissions, we estimate bottom-up emissions as the product of the annual NAEI emissions in Figure 3 and ratios of GEOS-Chem monthly to annual NH_3 emissions interpolated onto the $0.1^\circ \times 0.1^\circ$ grid. Figure 6 shows the resultant monthly bottom-up NH_3 emissions for April and July. The other months are in the supplementary (Figure S3). The bottom-up emissions peak in April (~14% of the annual total) coincident with fertilizer application (Hellsten et al., 2007; Paulot et al., 2014). The gridded difference between top-down and bottom-up emissions are also shown in Figure 6 for April and July and Figure S3 for the other months. Locations where bottom-up emissions exceed those from the top-down approach (red grids) mostly occur where emissions are low. The largest difference is in July when top-down emissions are 30 Gg more (IASI) and 48 Gg more (CrIS) than the bottom-up inventory. Pronounced regional differences include lower bottom-up values in eastern England, particularly in April, where fertilizer use and pigs and poultry farming are dominant sources, as well as in western England and Northern Ireland, particularly in July, where dairy cattle farming dominates (Hellsten et al., 2008). The spatial correlation between top-down and bottom-up gridded emissions in general ranges from $R = 0.5$ to $R = 0.7$, except for IASI in September ($R = 0.34$) when dynamic range in emissions is low.

The bottom-up emissions for March-September total 198.7 Gg. This is 27% less than IASI and 49% less than CrIS. According to the bottom-up inventory and Hellsten et al. (2007), March-September captures 60-67% of annual emissions. If we use this to scale IASI and CrIS to

annual totals, this suggests annual NH_3 emissions of 405-453 Gg according to IASI and 581-649 Gg according to CrIS. Natural NH_3 emissions total ~22 Gg in the UK (Section 3), so top-down annual anthropogenic NH_3 emissions are 383-431 Gg according to IASI and 559-627 Gg according to CrIS. Both top-down estimates exceed annual total anthropogenic emissions from the NAEI of 276 Gg (Section 3) and the UNECE Gothenburg emissions ceiling of 297 Gg.

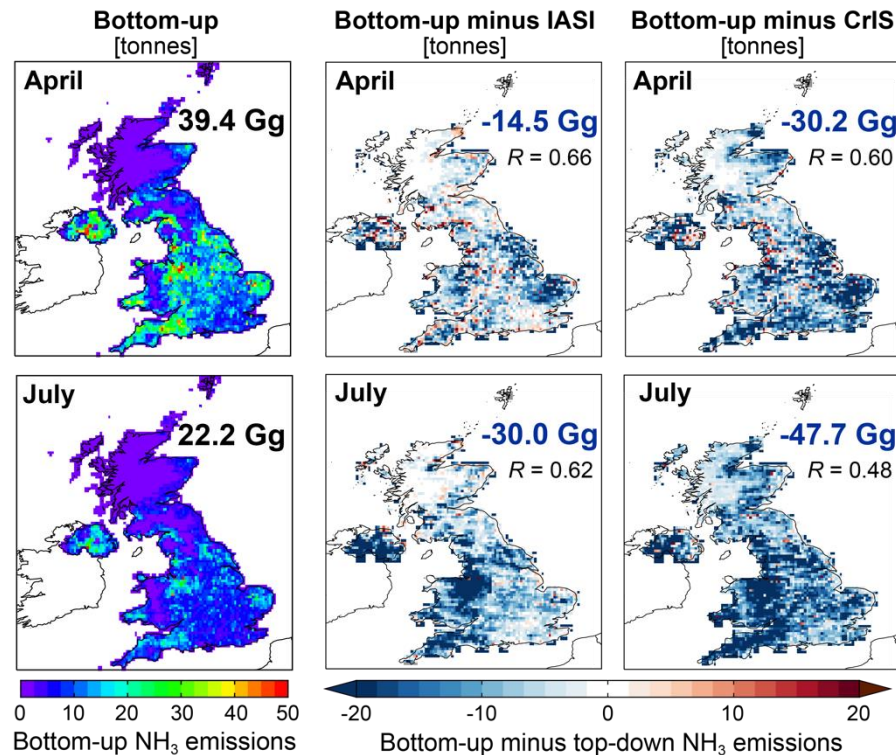


Figure 6. Comparison of bottom-up and top-down NH_3 emissions for April and July. Panels are bottom-up emissions (left), and the difference between top-down and bottom-up emissions for IASI (middle) and CrIS (right) in April (top row) and July (bottom row). Grids are blue for bottom-up < top-down and red for bottom-up > top-down. Values inset are bottom-up total (left) and differences in (middle and right) monthly emissions and the Pearson's spatial correlation (R) between top-down and bottom-up emissions.

Figure 7 compares regional seasonality in UK NH_3 emissions from bottom-up and top-down estimates as the percent change in monthly means relative to June. Regional seasonality in the top-down emissions is very similar in March-August in all regions except Northern Ireland. The mismatch between IASI and CrIS in September is due to a positive offset in CrIS relative to IASI columns. The July peak in emissions in Northern Ireland is more pronounced in IASI than CrIS. This is also apparent in the seasonality in the column densities (Figure S4). This may be due to differences in temporal coverage of the two sensors (2008-2018 for IASI, 2013-2018 for CrIS) over a location that has experienced dramatic changes in agricultural activity. This includes increases in livestock numbers of 45% for pigs and 42% for table chickens and a decline in nitrogen fertilizer of 37% from 2000 to 2016 (DEFRA, 2020b). All emission estimates exhibit a spring peak in April due to intensive fertilizer and manure application in March-April (Hellsten

et al., 2007). Paulot et al. (2014) also identified this April peak in NH_3 emissions inferred from ammonium wet deposition measurements, though a recent study questions the utility of these measurements for constraining NH_3 emissions (Tan et al., 2020). A second summer peak in the top-down emissions in July that is not present in the bottom-up inventory could be due to manure spreading and dairy farming (Hellsten et al., 2007). The likely contribution from dairy farming is supported by spatial consistency between the July top-down emissions (Figures 4 and 5) and locations dominated by emissions from dairy cattle (Hellsten et al., 2008).

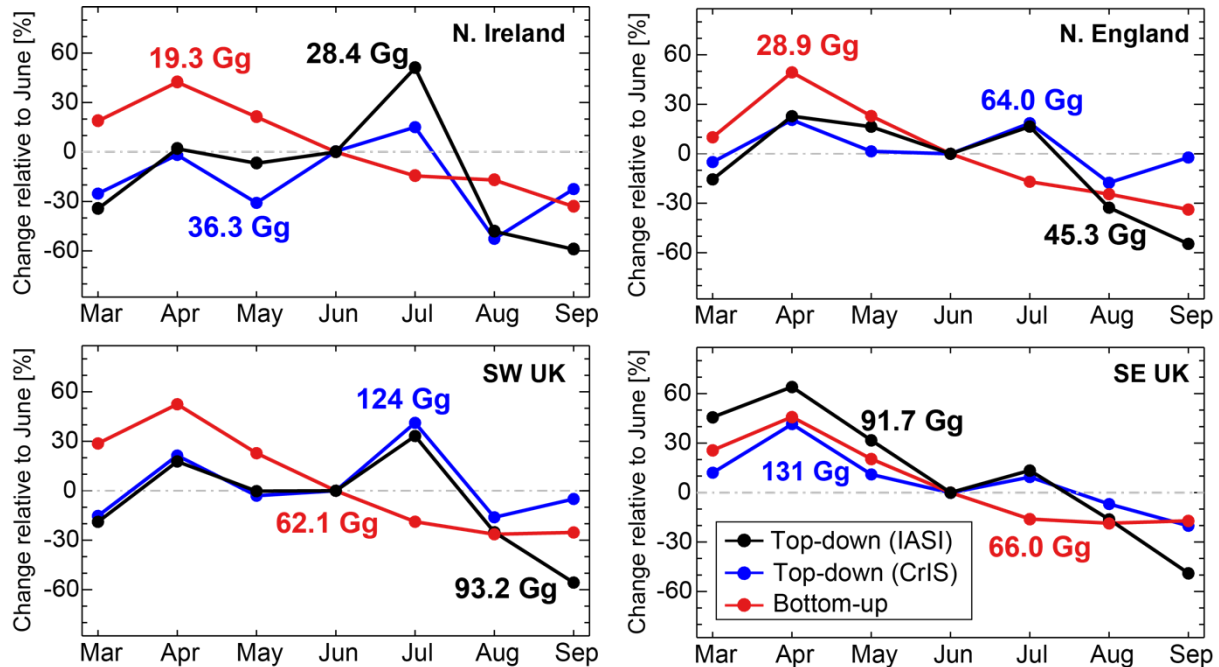


Figure 7. Regional seasonality in March-September NH_3 emissions. Points are the percentage change in monthly emissions relative to June for top-down emissions from IASI (black) and CrIS (blue), and from the bottom-up inventory (red). Regions sampled are in Figure 3. Inset values are March-September totals for each region from each estimate.

In Figure 8, we compare March-September 2016 mean modelled and observed surface concentrations of NH_3 to determine if the model driven with NAEI NH_3 emissions and prior assumptions of NH_3 seasonality and diurnal variability corroborates the results obtained with the satellite observations. Monthly means from model grids coincident with the surface sites are reasonably spatially consistent with the surface observations ($R = 0.54$) and the model is 38.3% less than the observations. This is midway between the NAEI comparison to the top-down emissions of 27% less than IASI and 49% less than CrIS. There are also rural low-cost passive sampler measurements of NH_3 concentrations, but these have low precision and are only reliable (within $\pm 10\%$ of reference measurements) at $\text{NH}_3 \geq 2 \mu\text{g m}^{-3}$ (Martin et al., 2019; Sutton et al., 2001). Even so, the model is similarly biased low (by 41.5%) compared to these measurements (not shown).

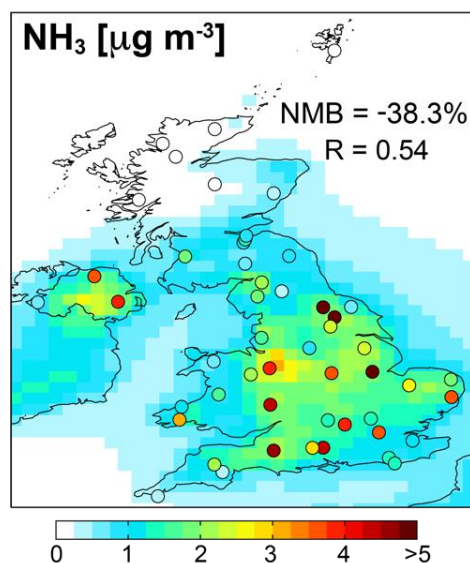


Figure 8. Comparison of observed and modelled surface concentrations of NH_3 . Data are EMEP and UKEAP site measurements (points) and the model (background) for March-September 2016. Inset values are the Pearson's spatio-temporal correlation coefficient (R) and the model NMB for coincident monthly means.

6 Error analysis of the top-down emissions

The reported relative error for NAEI NH_3 emissions is 31% (Ricardo, 2018a). Quantifiable random errors that contribute to total March-September satellite-derived emissions include uncertainties in retrieval of NH_3 , and in the modelled relationship between NH_3 emissions and column densities. For the latter we test sensitivity to modelled sulfate aerosol and nitric acid abundances and prior assumptions of the spatial and temporal variability of NH_3 emissions. IASI NH_3 retrieval errors for columns $\geq 2 \times 10^{15}$ molecules cm^{-2} range from 0.7-34%. Retrieval errors larger than 34% do occur, but are in locations with very low emissions. The CrIS NH_3 column errors across all grids range from 0.2-25%. Error contributions from uncertainties in sulfate and nitric acid are small compared to column density retrievals. We estimate these as the change in top-down emissions due to a perturbation in SO_2 emissions for

sulfate and NO_x emissions for nitric acid. The percent change in top-down emissions from a 50% decrease in SO_2 emissions is 4-5%. A 50% increase in NO_x emissions increases nitric acid by 14%, aerosol nitrate by 11%, and satellite-derived NH_3 emissions by 8-9%. The limited sensitivity to sulfate and nitrate in the UK is because NH_3 is in excess due to the success of emission controls targeting SO_2 and NO_x sources and absence of these for NH_3 sources. We also find that our top-down emissions estimates are relatively insensitive to NH_3 emissions perturbations. A 50% increase in NH_3 emissions causes a small (3%) decrease in satellite-derived NH_3 emissions. The total relative error from adding these individual errors in quadrature is 11-36% for IASI and 9-27% for CrIS and is dominated by errors in retrieval of the columns. Total emissions for March-September are 198.7 ± 61.6 Gg for the bottom-up inventory and up to 271.5 ± 97.7 Gg for IASI and 389.6 ± 105.2 Gg for CrIS.

There are also known systematic biases. Some studies reported that IASI NH_3 column densities are biased low by 25-50% compared to ground-based measurements (Dammers et al., 2017; Whitburn et al., 2016a). However, these comparisons were for earlier versions of the IASI NH_3 product. The version used here is consistent with columns derived with aircraft observations (Guo et al., 2021), though Guo et al. (2021) caution that their comparison is limited in time (summer) and location (Colorado, US) and sensitive to errors in column estimates from integrating aircraft measurements. The CrIS column amounts display a gradual increase with time (Figure S1) that we correct for in this work, though further work is required to determine the cause. Both satellite products preferentially sample clear-sky conditions. The bias that this may cause is challenging to quantify. Warmer temperatures and absence of clouds would reduce the amount of NH_3 that partitions to the aqueous phase (Stelson & Seinfeld, 1982; Walters et al., 2018), but NH_3 emissions would also increase (Sutton et al., 2013). The largest impact of clear-sky sampling in the UK may be in July, when boundary-layer clear-sky air temperatures, according to GEOS-Chem, are $\sim 6^\circ\text{C}$ warmer than all-sky scenes.

5 Conclusions

Emissions of ammonia (NH_3) in the UK are mostly (>80%) from agriculture and are challenging to estimate with bottom-up approaches and validate exclusively with current ground-based networks. Here we used satellite observations of NH_3 in March-September for multiple years from the Infrared Atmospheric Sounding Interferometer (IASI) (2008-2018) and the Cross-track Infrared Sounder (CrIS) (2013-2018) with the GEOS-Chem chemical transport model to derive top-down monthly emissions across the UK at high spatial resolution (~ 10 km).

Total top-down March-September emissions are 272 Gg from IASI and 390 Gg from CrIS. Bottom-up emissions estimated with the UK National Atmospheric Emission Inventory (NAEI) annual emissions and GEOS-Chem monthly scaling factors are 27% less than IASI-derived emissions and 49% less than CrIS-derived emissions. This is supported by a 38-42% underestimate in surface NH_3 concentrations from GEOS-Chem driven with the NAEI. We infer UK top-down annual anthropogenic NH_3 emissions of 383-431 Gg from IASI and 559-627 Gg from CrIS compared to 276 Gg from the NAEI. Seasonality in the top-down emissions confirms the well-known spring April peak from fertilizer and manure use, but there is also a summer July peak coincident with intensive dairy farming that is absent in the bottom-up inventory.

The relative error in the top-down emissions, mostly due to NH₃ column retrieval errors, is 11-36% for IASI and 9-27% for CrIS and is similar to the error reported for the NAEI (31%). The top-down emissions estimates are relatively insensitive to model uncertainties in SO₂, NO_x and NH₃ emissions, as NH₃ is in excess and the relationship between modelled NH₃ columns and emissions is near-linear.

Our study demonstrates the tremendous potential to use satellite observations to derive NH₃ emissions and assess bottom-up inventories under particularly challenging observing conditions (cloudy, cool) in the UK. This is critical for assessing reliability of these inventories for informing policies and mitigation strategies. The discrepancy between bottom-up and top-down emissions identified here warrants further investigation of both approaches.

Acknowledgments, Samples, and Data

The authors are grateful for helpful discussions with Daven Henze and Hansen Cao. EAM and AKP acknowledge funding from DEFRA (contract reference ecm_55415) and EAM acknowledges additional funding from NERC/EP SRC (grant number EP/R513465/1). The research in Belgium was funded by the F.R.S.-FNRS and the Belgian State Federal Office for Scientific, Technical and Cultural Affairs (Prodex 645 arrangement IASI.FLOW). Both MVD and LC are supported by the Belgian F.R.S.-FNRS.

The top-down and bottom-up emissions estimated in this work are publicly available from the UCL Data Repository (<https://doi.org/10.5522/04/14566635>). The CrIS CFPR NH₃ data are created by Environment and Climate Change Canada and hosted by the Meteorological Service of Canada (MSC) Datamart. Access to the CrIS NH₃ data can be requested from MWS (mark.shephard@canada.ca). The IASI NH₃ data are publicly available from the IASI data catalogue (<https://iasi.aeris-data.fr/nh3/>).

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