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SAQN Awards End of Project Report

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Project Title	
Improving Satellite Observations of Ammonia by Integrating Chemical Transport Modelling	
Project Team	
Name	Role (PI / Co-I)
Max Priestman	PI
Lucy Ventress	Co-I
Barry Latter	Co-I
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Proposed activities (copy from your project proposal)	

Aims and Objectives
This project builds on a previous SAQN scoping study [2] and has been co-developed with RAL. It aims to improve the spatial and temporal scale of satellite-derived NH_3 available to RAL and the wider research and policy community. Specific objectives are:

1. Establish the current level of agreement between satellite-derived NH_3 estimates with state-of-the-art regional chemical transport model (CMAQ).
2. Develop satellite NH_3 estimates at improved spatial and temporal scales through the integration of modelled vertical profiles.
3. Improve the agreement between satellite observations and ground-level measurements for individual episodes and longer timescales with a focus on intensive agricultural activity.

Work Packages:
WP A: Satellite and model estimates comparison for 2018-2020.

1. Evaluation of the CMAQ NH_3 estimates using ground-level measurements.
    1. Collect ground-level NH_3 high-time resolution measurements from NERC and EMEP supersites.
    2. Extract CMAQ model NH_3 estimates.
    3. Compare CMAQ model with ground-level measurements using metrics such as those used in DEFRA's model intercomparison [3] and Taylor diagrams [4].
2. Comparison of satellite-derived NH_3 estimates and CMAQ model.
    1. Temporal and geographical co-location of individual satellite soundings with CMAQ data.

2. Apply satellite vertical smearing (observation averaging kernel) to the CMAQ vertical profile output to calculate column average mixing ratio (ppb by volume).
3. Categorise individual satellite soundings into two groups "observable" and "non-observable" based on CMAQ column average mixing ratio and satellite detection threshold. The "observable" group will include soundings which have good signal-to-noise and model determined level of ammonia higher than threshold. The "non-observable" group will include soundings which the model determines as below the detectable ammonia threshold. This will provide vital data to RAL on satellite limit of detection (**Milestone 1**).
4. Aggregate and apply the satellite-derived and CMAQ column average mixing ratios on different spatial scales (national, regional, satellite sampling (~10km)).
5. Evaluate consistency and uncertainty between satellite and model by producing maps and statistics (MGE, MB, NMGE, NMB, RMSE and R). (a) sampling all data and (b) restricting to "observable" satellite and CMAQ data.

WP B: Improving spatial and temporal resolution of satellite estimates (**Milestone 2**).

1. Compare maps of satellite and model data and their differences gridded on various spatial (10km to 100km) and temporal (monthly to multi-annual) scales.
2. Compare satellite and model data aggregated on various spatial (e.g. geographical, land use, farming type), and temporal (daily to seasonal) scales.
    1. Identify high pollution, peak agricultural activity episodes [5] from model data.
    2. Collate satellite and modelled data for the high pollution episodes.
    3. Evaluate consistency between datasets spatially and temporally using metrics in [3], Taylor diagrams [4] and Theil-Sen method.

WP C: Comparing satellite and ground-level observations (**Milestone 3)**.

1. Co-locate individual satellite soundings to measurement stations, aggregate to available measurement timebase (hourly/monthly) and subset hourly ground-level data for satellite overpass times.
2. Compare satellite and ground-level measurements for individual stations aggregated over spatial scales, and for high pollution episodes.
3. Assess correspondence between satellite and surface measurements using metrics in [3] and Taylor diagrams [4].

Please report on the activities completed in the project

The project completed work related to Milestone 1 and part of Milestone 2. This report will detail the comparison of the CMAQ model data to ground-based NH_3 measurements, the investigation into filtering methods for the satellite data set and the comparison of satellite and CMAQ data different spatial and temporal resolutions. Data for 2018 and 2019 were analysed and many different filtering methods were investigated including cloud fractions of varying degrees: 5%, 20%, 50% and 100%, in addition filtering data based on the thermal contrast between the surface and 1km altitude level. We initially analysed CMAQ data at hourly and monthly resolution, however, it was concluded there were not a sufficient number of satellite soundings to investigate the different satellite filters and therefore data were averaged for 3-month seasons. The project has produced a large volume of data and results, and so only selected results are shown in this report.

Comparison of CMAQ model and ground-based sensors:

The CMAQ model data was evaluated with high-time resolved ground-based ammonia data from the EMEP network and NERC OSCA campaign, a total of 5 sites across the UK.

The EMEP data (Auchencorth Moss and Chilbolton) was available for 2018-2020 and NERC OSCA (Honor Oak Park, Manchester and Birmingham) data was available for 2019-2020. The ground-based data was evaluated with the 2km CMAQ data set for the corresponding grid square. The evaluation was initially run for hourly data, however there was a large amount of variation in both the ground-based and modelled data sets. Monthly averaged data also showed a large amount of variation (Figure 1). For sites apart from Auchencorth Moss, the ground-based data had consistently higher concentrations than the CMAQ data set, with a large difference between two data sets for the urban sites.

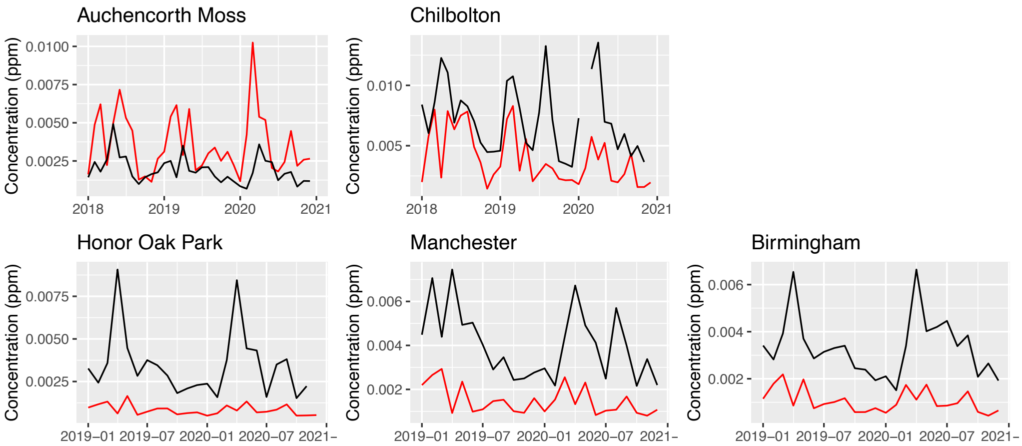

Figure 1: Time series of monthly average CMAQ model data (red) and ground-based data (black) for the 5 EMEP and NERC high-time resolved monitoring sites.

Figure 2 shows the error for the urban sites, with the largest amount of error occurring in April for all sites in both years. Figure 3 shows a less clear pattern in the rural data sets, with the largest error for Auchencorth Moss occurring in March 2020, and two peaks in error in March and May 2019. The largest error for Chilbolton occurs in August 2019 and April 2020. The CMAQ model uses an assumed profile to allocate the total emissions of NH_3 from the NAEI throughout the year, currently peaking in May. NH_3 emissions are temperature dependent, and therefore the error in April present in 4 out of 5 sites suggests that more investigation needs to be undertaken to understand whether the temperature for years 2018-2020 differing from the norm and therefore if the profile needs to be changed. Additionally, urban NH_3 emissions in the NAEI are small, however the plots in Figures 1-3 suggest that this may not be the case and highlights the need for further research to investigate the reasons for this and update traffic NH_3 emissions.

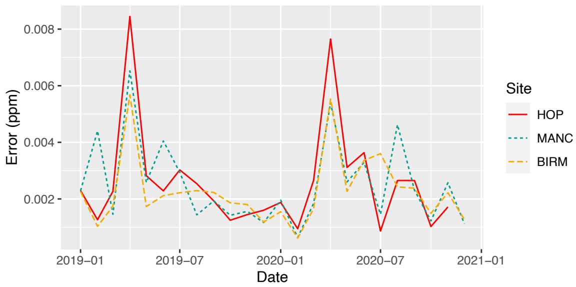

Figure 2: Time series plot of monthly averaged error between the CMAQ model data and ground-based data for the three NERC high-time resolved urban monitoring sites.

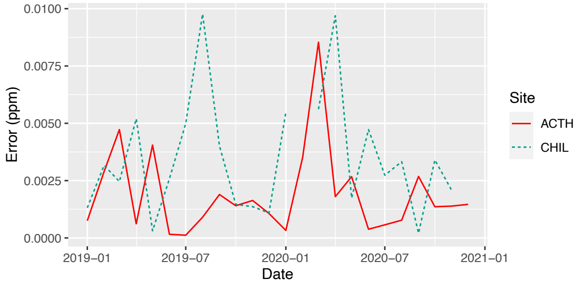

Figure 3: Time series plot of monthly averaged error between the CMAQ model data and ground-based data for the two EMEP high-time resolved rural monitoring sites.

The evaluation statistics, Table 1, shows that there is a weak relationship between the monthly averaged ground-based high time resolved monitoring site data and CMAQ model data. The normalised mean bias (NMB) and normalised mean gross error (NMGE) are similar for the urban sites (HOP, MANC, BIRM), with the NMB values suggesting that the CMAQ data set is underpredicting by between 62% to 75% for the urban areas and the NMGE shows that there is a large amount of variability in both data sets (62% to 75%). Whereas the rural site ACTH has the largest amount of variability (NMGE: 95%) with the CMAQ data set underpredicting by 89%. The rural Chilbolton (CHIL) site has the lowest NMB and NMGE statistics. Overall, the correlation between the CMAQ data and ground-based monitoring site data varies from 0.2 for Honor Oak Park (HOP) to 0.46 for Chilbolton (CHIL).

Table 1: Validation statistics for the CMAQ model data sets compared to the ground-based high-time resolved monitoring sites data, both data sets are monthly averages.

Site	**Monthly**		
	NMB (%)	NMGE (%)	R
ACTH	0.89	0.95	0.34
CHIL	-0.44	0.44	0.46
HOP	-0.75	0.75	0.20
MANC	-0.62	0.62	0.30
BIRM	-0.68	0.68	0.31

Comparison of satellite-derived NH_3 estimates and CMAQ model (Milestone 1) and investigation of spatial and temporal resolution of satellite estimates (Milestone 2).

Satellite data on ammonia has been provided by the Remote-Sensing Group at RAL Space using their Infrared and Microwave Sounder (IMS) retrieval scheme applied to the Cross-track Infrared Sounder (CrIS) and Across-track Microwave Sounder (ATMS) on the

Suomi-NPP satellite. Each individual sounding of ammonia column-average volume mixing ratio was accompanied by its corresponding averaging kernel (AK) describing vertical sensitivity along with estimated random error and auxiliary information on cloud, temperature profile, other co-retrieved variables and data quality indicators.

The satellite vertical sensitivity to NH_3 was investigated by integrating with the chemical transport model CMAQ data. Each satellite sounding was co-located with the corresponding CMAQ grid cell. The satellite averaging kernels were then applied to the z-dimension of the CMAQ data at the equivalent heights and the column average mixing ratio (CAMR) was calculated (CMAQ with Satellite sensitivity CAMR) from the resulting profile. The CMAQ with Satellite sensitivity CAMR was compared to the satellite CAMR and CMAQ CAMR.

The comparisons were conducted using CMAQ model data gridded at spatial resolutions of 10km and 50km, for each season in the years 2018-2019. Different satellite filters were applied to the CMAQ data, investigating the effect of using various levels of cloud fraction and thermal contrast between the surface and 1km altitude level. Cloud fractions of 5%, 20%, 50% and 100% were applied as well as a temperature contrast filter of greater than 10°K (DT1000). All the tests included a measure of the spectral fit precision (cost function) too (only values <1000 were permitted).

Using a cloud fraction of 20%, the satellite CAMR and CMAQ with satellite sensitivity CAMR plots for the summer season (June, July, August) 2018 (Figure 4) were broadly consistent with each other, with both data sets picking out emission hotspots such as the agricultural emissions in the Netherlands. The CMAQ CAMR plots (Figure 4) have higher NH_3 concentrations compared to the Satellite CAMR plots. However, the satellite vertical sensitivity is dependent on the thermal contrast between the surface and atmosphere, and therefore has reduced sensitivity to NH_3 at the ground-level. This is shown by the satellite averaging kernels in Figure 5, which highlights the low satellite sensitivity at ground-level where the CMAQ NH_3 concentrations are highest. This feature explains why it is not unexpected to see a lower value for the satellite CAMR compared to the CMAQ model data before averaging kernels have been applied to the model.

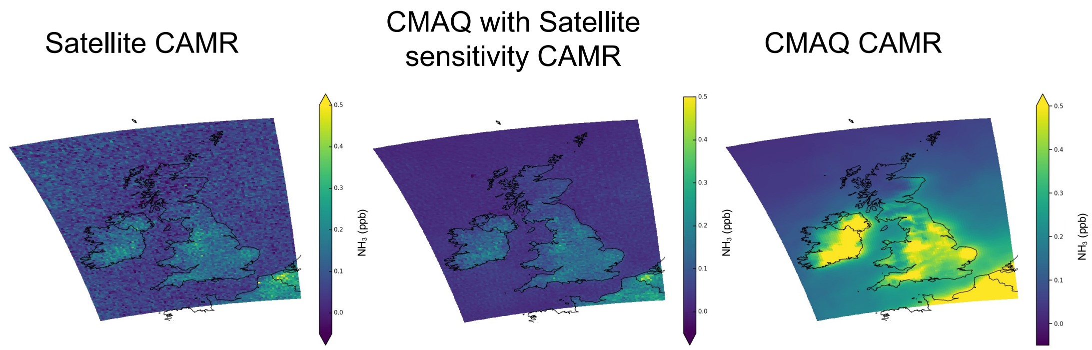

Figure 4: Comparison of season (June, July and August 2018) average Column Average Mixing Ratio (CAMR) plots for the different data sets. Satellite CAMR (left plots), the satellite vertical sensitivity applied to CMAQ z-dimension NH_3 estimates CAMR (middle plots) and CMAQ CAMR season average for 1pm (right plots) for the 10 km CMAQ model domain. The satellite data has been filtered with a cost value of 1000, a cloud fraction of 20% and without the DT1000 filter. The legends have been set to a maximum of 0.5 ppb and minimum of 0.05 ppb.

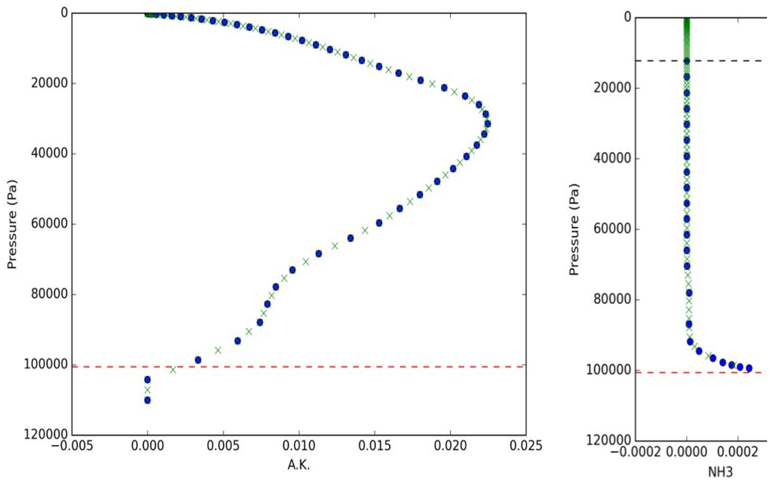

Figure 5: (a) Satellite sensitivity (A.K.) throughout the atmosphere. The red dashed line represents surface pressure, the circles represent the satellite sensitivity values provided with the data set and the crosses represent the interpolated sensitivity to the 101 pressure levels. (b) The blue circles show the CMAQ NH_3 concentrations at the 23 pressure levels throughout the atmosphere and the green crosses represent the interpolated CMAQ NH_3 concentrations at the satellite pressures (the black dashed line represents the extent of the CMAQ model).

To investigate the satellite sensitivity, different filters were compared. Table 2 shows validation statistics comparing the CMAQ with Satellite sensitivity CAMR to the satellite CAMR for the 10km summer season average (JJA) 2018 using a combination of different cloud fraction (CF) and surface to 1km temperature contrast (DT1000) filters. Any grid cells that fell outside of the UK boundaries (in the sea) were removed as NH_3 levels are expected to be very low over the sea surrounding the UK except from plumes arriving from the Netherlands and NW Europe. The highest R value of 0.76 was for the data set with a 20% cloud fraction and DT1000 >=10K filters, closely followed by the 50% (R = 0.66) and 20% (R = 0.65) cloud fraction filtered data set. The NMB shows that the CMAQ with Satellite sensitivity CAMR is underpredicted by 13% and 17% for the cloud fraction filters of 50% and 100% respectively. In contrast, the CMAQ with Satellite sensitivity CAMR is overpredicted by about 24% for the 20% cloud fraction and DT1000 >=10K data set. The NMB shows that the data produced with the most stringent cloud filters (5% and 20%) is the least biased, compared to more relaxed cloud filtered (50% and 100%) data which as anticipated is negatively biased. The NMGE values show that there is variability in all of the data sets, ranging from 42% to 62% in the 20% cloud fraction and DT1000 >=10K data set compared to the 100% cloud fraction data set.

Figure 6 shows scatterplots of the corresponding 10km summer season average (JJA) 2018 CMAQ-Satellite sensitivity CAMR and satellite CAMR plots for the different filters, where the points are coloured based on the corresponding NUTS-1 area they are contained within. The 20% cloud fraction and DT1000 >=10K data set plot shows that generally NH_3 estimates in the Satellite CAMR data set are lower compared to the CMAQ with Satellite sensitivity CAMR data set estimates, this is most apparent for estimates over Scotland (pink) which overall have the lowest NH_3 values and largest variability.

Table 2: Validation between CMAQ with Satellite sensitivity CAMR and Satellite CAMR for the summer season (June, July and August 2018) for the 10 km CMAQ model domain. Various different cloud fractions (CF: 5%, 20%, 50% and 100%) and air surface temperature contrast (DT1000) filters are shown (excluding grid cells outside of the UK land mass).

CF filter	DT1000 filter	NMB	NMGE	R
5%	none	0.080	0.493	0.595
20%	none	-0.040	0.355	0.653
50%	none	-0.133	0.321	0.659
100%	none	-0.165	0.621	0.346
20%	>=10K	0.237	0.421	0.757

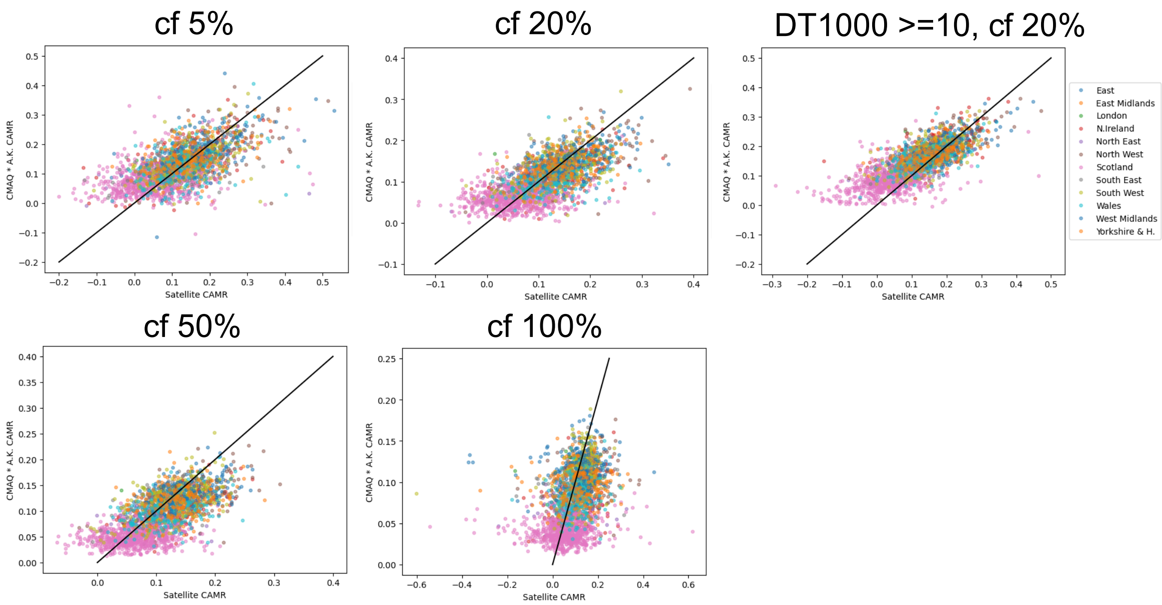

Figure 6: Scatterplots of the CMAQ with Satellite sensitivity CAMR and Satellite CAMR for the summer season (June, July and August 2018) for the 10 km CMAQ model domain. Various different cloud fractions (CF: 5%, 20%, 50% and 100%) and air surface temperature contrast (DT1000) filters are shown.

Comparing Figure 4, which shows data filtered with a cloud fraction of 20%, with Figure 7 which shows data filtered with a cloud fraction of 20% and a DT1000 filter of >=10°K, it can be seen that many of the satellite soundings over the sea do not have the required surface to 1km temperature difference of above 10°K and therefore the gridded dataset has gaps. In both data sets, there appears to be a more defined land sea boundary in the satellite data. The cause of this is under investigation but it may be due to cloud often occurring at low altitude over sea which is difficult to detect with thermal IR (and microwave) soundings alone.

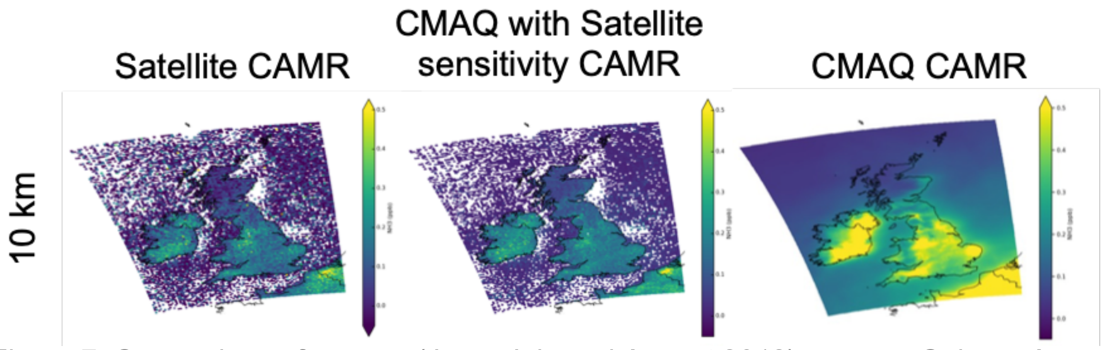

Figure 7: Comparison of season (June, July and August 2018) average Column Average Mixing Ratio (CAMR) plots for the different data sets. Satellite CAMR (left plots), the satellite vertical sensitivity applied to CMAQ z-dimension NH_3 estimates CAMR (middle plots) and CMAQ CAMR season average for 1pm (right plots) for the 10 km CMAQ model domain. The satellite data has been filtered with a cost value of 1000, a cloud fraction of 20% and with the DT1000 filter set to >=10°K. The legends have been set to a maximum of 0.5 ppb and minimum of 0.05 ppb.

Table 3 shows the evaluation statistics for two combinations of filters, the 20% cloud fraction filtered data set, and a 20% cloud fraction and DT1000>10K filtered data set. The evaluation statistics are shown for three different seasons, where for both data sets the summer (JJA) season summer (JJA) season summer (JJA) season has the highest R value (0.653 and 0.757 respectively), in addition to the lowest NMB and NMGE values. For both data sets, the worst statistics are for the autumn months (SON).

Table 3: Validation between CMAQ with Satellite sensitivity CAMR and Satellite CAMR at 10km CMAQ model domain for the different seasons in 2018 comparing a 20% cloud fraction filter to a combination of 20% cloud fraction and DT1000 >=10K filters (excluding grid cells outside of the UK land mass).

Season and year	CF filter	DT1000 filter	NMB	NMGE	R
2018 MAM	20%	none	0.254	0.557	0.544
2018 JJA	20%	none	-0.040	0.355	0.653
2018 SON	20%	none	-0.735	1.097	0.066
2018 MAM	20%	>=10K	0.386	0.625	0.567
2018 JJA	20%	>=10K	0.237	0.421	0.757
2018 SON	20%	>=10K	0.37	3.456	0.307

The 50km CMAQ JJA 2018 season average data had an error, and therefore to investigate the spatial scales MAM 2018 data was used. Comparing the 10km to 50km data (filtered using a 20% cloud fraction), the validation in Table 4 shows that the 50km data has the highest correlation (R = 0.77) which is anticipated as the increased sampling density will reduce the noise by a factor of ~5. The NMB values are similar for both data sets, showing that for both spatial scales the CMAQ with Satellite sensitivity CAMR is overpredicting by about 25%. However, the 50km data is more variable (56% compared to 38%), this is further shown in Figure 8. It should be noted that these comparisons are for one season in one year, to understand the influence of spatial scale on NH_3 estimates other seasons and years should be investigated.

Table 4: Validation between CMAQ with Satellite sensitivity CAMR and Satellite CAMR for the spring season (March, April and May 2018) using a cloud fraction filter of 20% comparing 10km and 50km CMAQ model domain data (excluding grid cells outside of the UK land mass).

Cloud Fraction	DT1000 filter	Spatial scale	NMB	NMGE	R
0.2	none	10km	0.254	0.557	0.544
0.2	none	50km	0.255	0.376	0.767

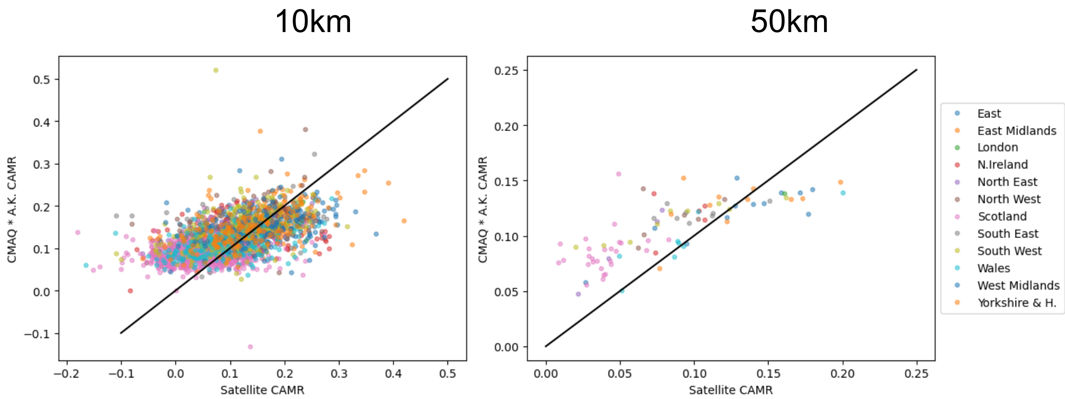

Figure 8: Scatterplots of the CMAQ with Satellite sensitivity CAMR and Satellite CAMR for the summer season (March, April, May 2018) using a cloud filter of 20% comparing the 10 km and 50km CMAQ model domain.

Future work could include the following:

- Optimise further the sampling and aggregation of both satellite and model data, with averaging kernels applied e.g. with respect to satellite soundings with near-surface sensitivity.
- Apply oversampling method to optimise both satellite and model data, with averaging kernels applied.
- Adjust satellite data for known bias.
- Extend comparison to 2020.
- Assess Satellite - 'CMAQ with Satellite sensitivity' difference maps to identify possible causes on both sides.
- Assess the low satellite sensitivity at ground-level in comparison to ground-based measurements.
- Apply all A.K. in CMAQ grid box to CMAQ domain rather than using the nearest satellite sounding.
- Test other filters including DT1000 >=15°K.
- Repeat analysis with improved, re-processed satellite data once available.
- Further research on urban sources of NH_3.
- Investigation of the CMAQ model assumed NH_3 yearly profile and the relationship with temperature.
- Evaluate the CMAQ model and satellite-derived NH_3 estimates with monthly UKEAP ground-based measurements.

Milestone 3 was not feasible to complete within the timeframe. Our plan for this work is to apply for any relevant funding sources or include the work within a bigger project and apply for UKRI NERC grant.
What are the next steps for this research? Will you be applying for further funding? What will you need to continue researching this topic?
We have applied for a STFC Knowledge Exchange Impact Award (via Imperial) for a project to create a network of stakeholders on satellite ammonia data for policy applications. We have plans to apply for UKRI grant on integrating ground-based measurements, chemical transport modelling and satellite observation, to quantify a range of sources of atmospheric NH_3 improving links with policy makers further.
Please outline the role of STFC in this project
RAL provided CrIS Level-2 satellite soundings, averaging kernels, filters for 2018-2020 and IDL code, as well as scientific support for data analysis and interpretation via weekly meetings and in-person meetings.
Please list a brief list of all outputs and impacts below. These may include papers, articles or blogs, presentations at events or conferences, meetings about future plans for the research. Please include links wherever possible
- Presented results in SAQN Ammonia networking meeting - RAL presenting at Clean Air Network Conference - Future opportunity (September/ October 2023) to present work in an event organised by the Clean Air Research Future's Group (CARFuG) work programme.
Were there any unexpected outcomes as part of the project?
The period needed to get acquainted with the "raw satellite soundings (L2)" file format, codes to read and aggregate data, access data from JASMIN was lengthier than foreseen. However, this project has now produced a code base which can be used for future analysis of this work, including existing IDL code translated into python. The comparison between CMAQ model and high-time resolved ground-based measurements showed an underprediction of NH_3 by the CMAQ model in urban areas. This result highlighted the need for further research into urban NH_3 emissions and update of these sources in the NAEI to reflect findings. Further, the comparison showed a large error in the CMAQ model estimates in April, highlighting the need for further evaluation using 85 monthly ground-based monitoring sites (UKEAP network) and research on the temperature dependence of NH_3 and inclusion of temperature impacts on assumed NH_3 yearly profile.
What are your plans to share the outcomes of this research with others? (Give details of any future meetings, conferences, papers or other dissemination planned)
We are planning to disseminate the outcomes of this research via a peer-reviewed journal paper and will present results at national and international conferences (subject to funding) such as the RSPSoc Wavelength conference and ESA Living Planet Symposium.

Project Impact: What is the most significant output/impact from this project?
This work is the first comparison over the UK of (these) satellite data with a regional CTM. The results indicate that the approach is worthwhile to pursue further; to identify and investigate discrepancies occurring on this scale and areas of potential improvement necessary to CMAQ and/or satellite data quality. Further work will benefit near-term from an improved version of the RAL ammonia retrieval scheme (in development) and subsequently from the new generation satellites to be launched in 2024 and 25 with higher spatial and temporal resolution and higher quality.