S1: Emissions and station incidence estimates

Land-use information is derived from the 2024 Graphical Parcel Register (Registre Parcellaire Graphique, RPG : https://artificialisation.developpement-durable.gouv.fr/bases-donnees/registre-parcellaire-graphique), produced by the French National Institute of Geographic and Forest Information (IGN). This dataset provides detailed information on agricultural land use at the parcel scale, enabling a precise description of declared crop types across the study area. To characterize the agricultural environment surrounding each surface measurements, the following methodology is implemented:

- Creation of two buffer zones with radii of 500 m and 1 km around each monitoring site;
- Extraction of all RPG parcels intersecting these buffer zones;
- Aggregation of individual crop types into broader and more homogeneous categories (e.g., winter wheat, permanent grassland, grain maize);
- Calculation of the relative share (%) of each crop category within the buffer area;
- Identification of the dominant crop types surrounding each monitoring location.

This approach provides a robust description of local agricultural practices and their potential influence on atmospheric ammonia concentrations, particularly through activities such as fertilization and manure spreading (Figure A1).

Livestock Data – Annual Agricultural Statistics (SAA)

Livestock information is obtained from the Annual Agricultural Statistics (Statistique Agricole Annuelle, SAA : https://agreste.agriculture.gouv.fr/agreste-web/methodon/S-SAA/methodon/), distributed by Atmo France through the national PRISME platform (https://www.atmo-france.org/actualite/prisme-la-plateforme-air-climat-energie-pour-la-transition-ecologique). Similar to RPG, the following steps is applied:

- Extraction of livestock population data at the municipal scale;
- Conversion of livestock numbers into Livestock Units (LU) to ensure consistency across the different animal species;
- Calculation of the relative share (%) of each livestock category within each municipality;
- Spatial allocation of livestock data to monitoring sites using municipal boundaries and spatial overlay techniques;
- Identification of the dominant livestock production system surrounding each passive sampler.

Site Typology and Dominant Influence Classification

The combined analysis of land-use information from the RPG, livestock data from the SAA, and field observations (site visits and local contextual knowledge) enabled the development of a classification framework for the monitoring sites according to their surrounding environment and dominant emission influences.
Two complementary levels of classification are defined:

- Site Typology, describing the general characteristics of the monitoring environment (background, rural, traffic-influenced, etc.);
- Dominant Influence, identifying the primary source expected to affect observed concentrations (livestock farming, crop cultivation, road traffic, or regional background).

Determination of Dominant Influences

The dominant influence assigned to each site is established through a combination of spatial indicators and expert assessment. For agricultural environments, the following criteria were considered:

- Agricultural land cover exceeding 70% of the area within the 500 m or 1 km buffer;
- Presence of significant crop production and/or livestock activities, as identified from RPG and SAA datasets;
- Absence of major urban, industrial, or traffic-related influences.

The distinction between crop-dominated and livestock-dominated environments is based on the relative importance of cultivated land and livestock density indicators. Sites characterized by extensive grasslands and high livestock-unit densities are classified as livestock-influenced, whereas sites dominated by arable crops and lower livestock densities were classified as crop-influenced. Traffic-influenced sites were identified through their proximity to major roads and transport corridors, while regional background sites corresponded to locations with limited local anthropogenic influences.

Reference Framework for Rural Site Classification

The criteria used to define rural sites are consistent with established European and French monitoring frameworks, notably the MERA (https://www.lcsqa.org/fr/le-dispositif-mera) network and the EMEP (https://unece.org/fileadmin/DAM/ie/capact/ppp/pdfs/rws2/emep_man_e.pdf) monitoring program. These reference systems recommend selecting locations representative of regional agricultural environments while minimizing the influence of local point sources.

Site selection was carried out in collaboration with IMT Nord Europe (IMT-NE), which coordinates the monitoring program. Given the highly agricultural nature of Brittany, identifying sites representative of rural background conditions required particular attention. The final selection aimed to balance representativeness of regional agricultural emissions with the need to avoid direct influence from nearby intensive emission sources.

This classification constitutes a key step in the interpretation of measured concentrations, the comparison of results between sites, and the assessment of the representativeness of the monitoring network with respect to the different emission sources present across the region.

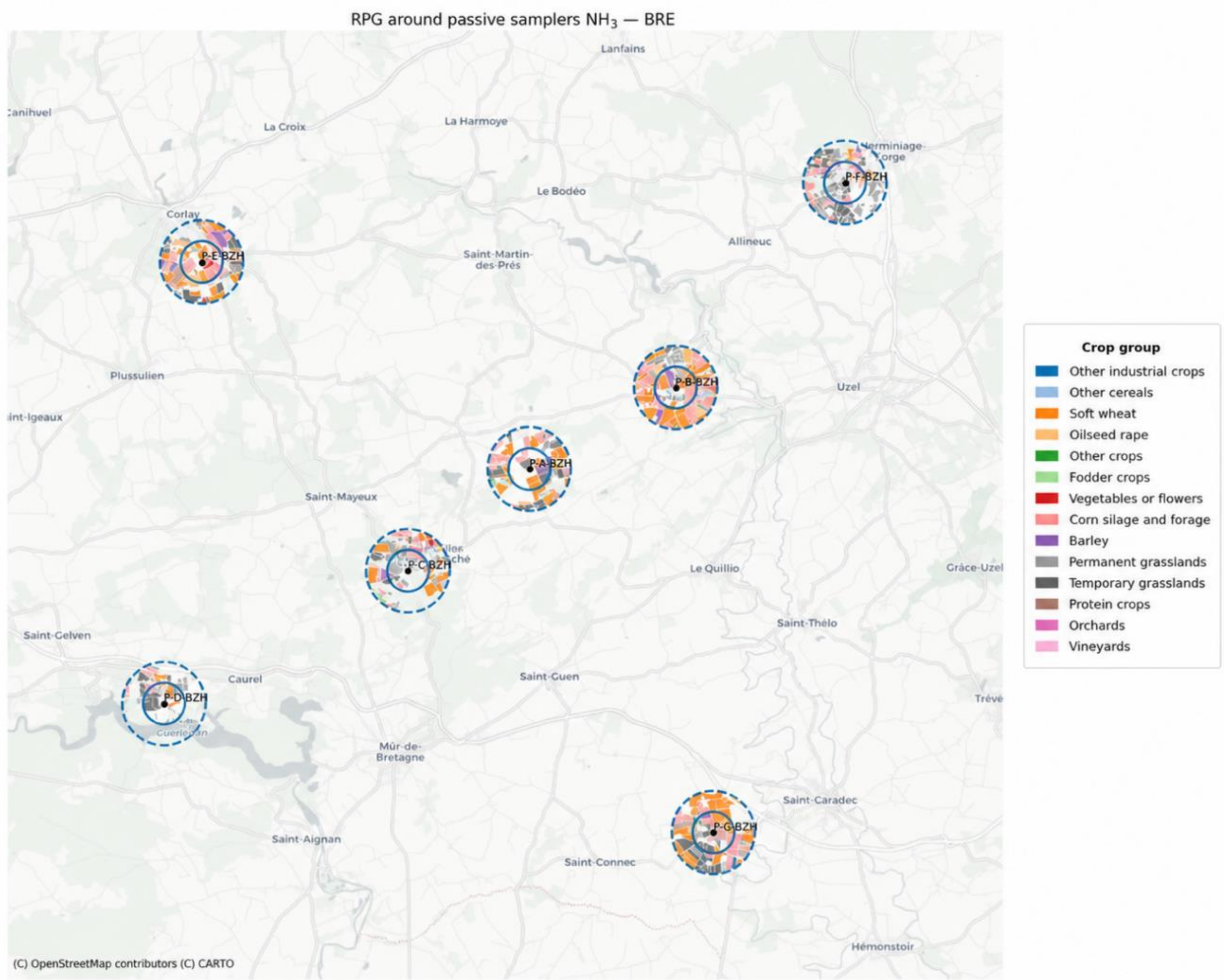

Figure S1: Map of Graphical Parcel Register (RPG) data around each ROSAS monitoring site in Brittany.

S2

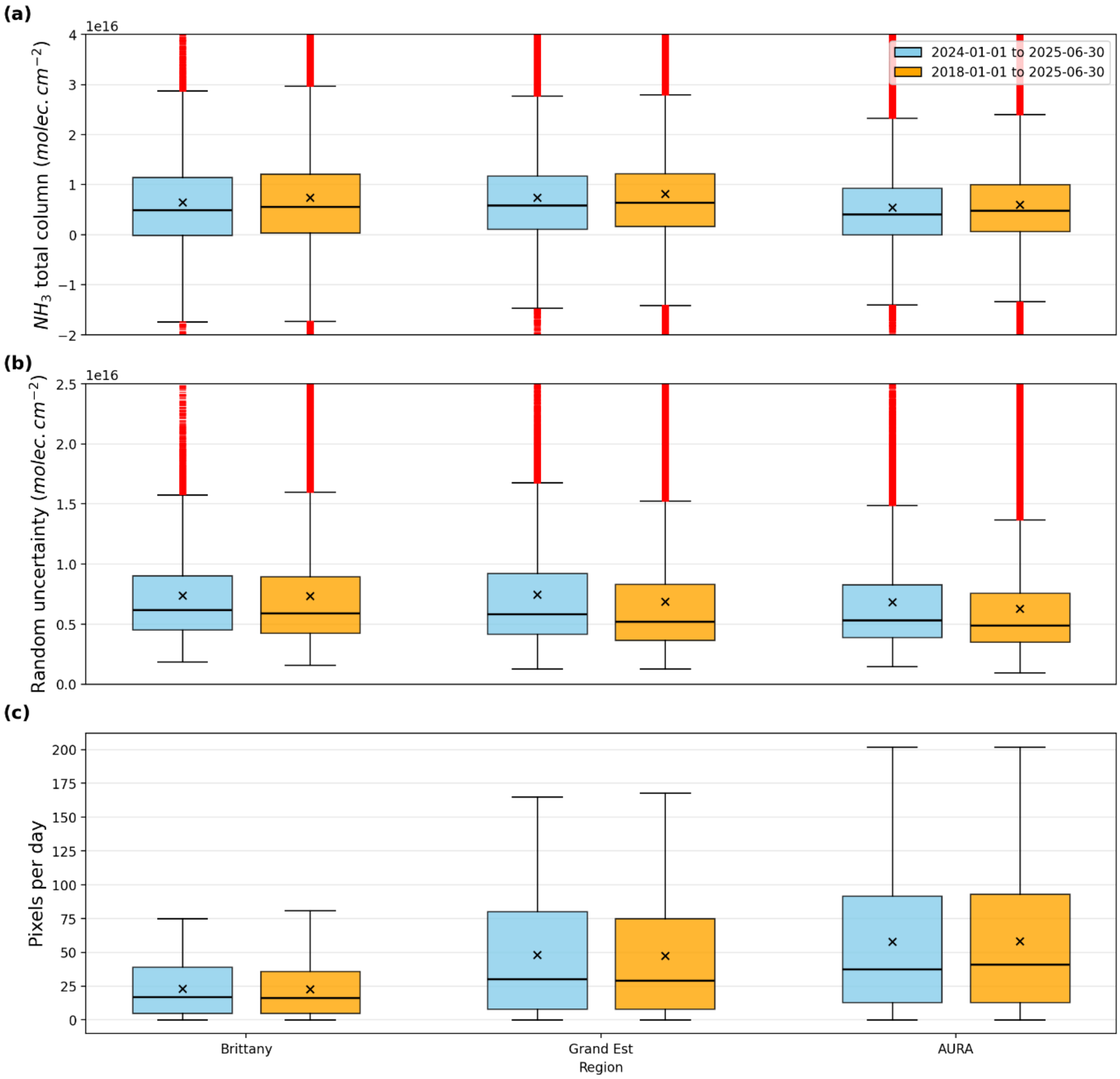

Figure S2: Statistical distributions of total NH_3 column values (a) from IASI, the associated random uncertainty values (b) and the number of pixels (c) across the three regions during the ROSAS campaign period (in blue) and from 1 January 2018 to 30 June 2025 (in orange). The whiskers represent the minimum and maximum values, the box edges correspond to the first and third quartiles, the horizontal line within the box indicates the median, the cross denotes the mean, and outliers are shown in red dots.

S3

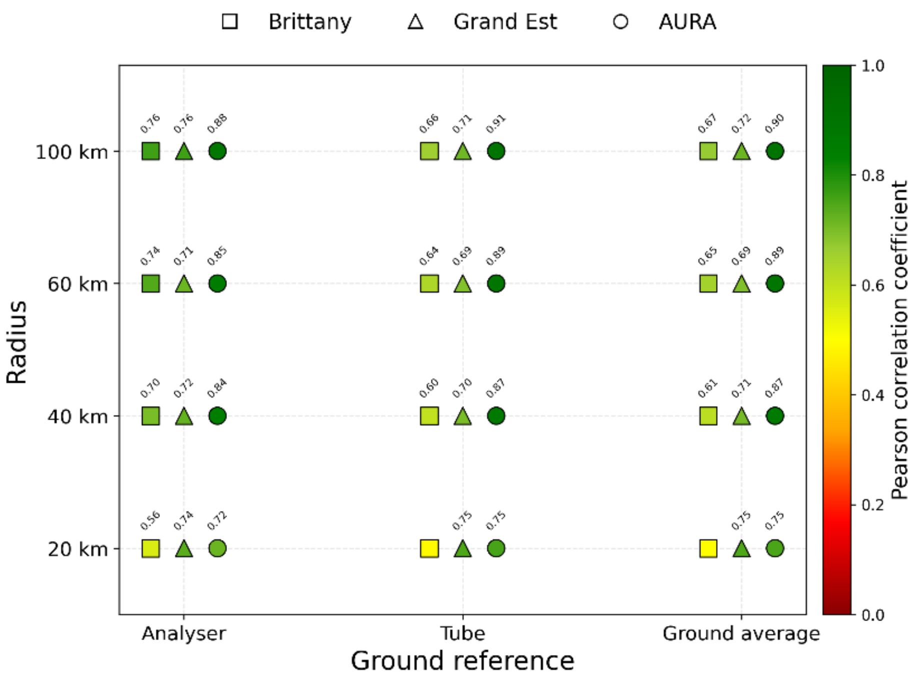

Figure S3: Space–time diagrams of color-coded Pearson correlation coefficient between a two-weeks average of IASI and surface NH_3 observations for the 3 regions (square for Brittany, triangle for Grand Est, and circle for Auvergne-Rhône-Alpes) during the ROSAS campaign when comparing with the analyzer, the passive tube, both located at the centered of the region, or comparing with the averaged ground observations (including all passive tubes and analyzer). All correlations are displayed, but only those that are statistically significant (p-value < 0.05) are annotated.

S4

Table S4: Comparisons between IASI and all passive samplers deployed in each region during the ROSAS campaign. Pearson correlation coefficients (R) and p-values are shown together with the number of surface observations and IASI pixels used in the comparison, as well as mean NH_3 concentrations from both IASI (molec. cm^{-2}) and passive tubes ($\mu g.m^{-3}$).

Region	Passive Tube name	Pearson R	p_value	surface Nobs	IASI Npixels	IASI NH_3 (molec.cm^{-2})	Passive Tube NH_3 ($\mu g.m^{-3}$)
Brittany	P-B-BZH	0.66	1.38E-04	28	299	7.8793E+15	1.41
	P-G-BZH	0.62	7.43E-04	26	269	8.1186E+15	3.13
	P-A-BZH	0.53	5.16E-03	26	269	8.2963E+15	3.97
	P-E-BZH	0.47	1.21E-02	28	299	7.8793E+15	1.49
	P-F-BZH	0.44	1.82E-02	28	299	7.8793E+15	5.98
	P-C-BZH	0.35	6.75E-02	28	299	7.8793E+15	2.62
	P-D-BZH	0.32	1.03E-01	27	292	7.7298E+15	3.33
Grand Est	P-G-GE	0.80	2.15E-07	29	406	7.1282E+15	1.38
	P-C-GE	0.77	1.13E-06	29	406	7.1282E+15	2.62
	P-F-GE	0.74	4.45E-06	29	406	7.1282E+15	2.29
	P-A-GE	0.69	3.66E-05	29	406	7.1282E+15	5.16
	P-D-GE	0.69	3.70E-05	29	406	7.1282E+15	3.63
	P-E-GE	0.63	2.33E-04	29	406	7.1282E+15	2.72
	P-B-GE	0.59	9.17E-04	28	393	7.1278E+15	2.60
Auvergne-Rhône-Alpes	P-A-AURA	0.86	1.56E-09	29	415	4.307E+15	1.48
	P-G-AURA	0.86	3.15E-09	28	411	4.420E+15	1.88
	P-E-AURA	0.85	5.56E-09	29	415	4.307E+15	1.10
	P-B-AURA	0.84	3.72E-08	27	380	3.974E+15	1.71
	P-C-AURA	0.82	1.11E-07	28	397	4.112E+15	1.53
	P-D-AURA	0.81	1.88E-07	28	380	4.111E+15	1.31
	P-F-AURA	0.79	2.73E-07	29	415	4.307E+15	2.30