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Special Collection:

Advancements in Observing
Atmospheric Composition from
Hyperspectral Infrared Sensors

Key Points:

- We devise a new physics-constrained neural-network method for measuring atmospheric methane from Infrared Atmospheric Sounding Interferometer (IASI) satellite data and ancillary information
- The method detects near-surface enhancements and reconstructs the atmospheric methane profile
- The method supports the monitoring of natural and anthropogenic emissions for climate studies and is demonstrated on the Nord Stream case with IASI data

Supporting Information:

Supporting Information may be found in the online version of this article.

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A New Physics-Constrained Neural Network Approach to Retrieve CH₄ Vertical Profiles From Hyperspectral Infrared Data

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Abstract Methane (CH₄) is a critical greenhouse gas, and accurate monitoring of its vertical distribution is essential for understanding its radiative impact and identifying emission sources. This paper introduces a novel methodology using a physics-constrained neural network based on soft constraints to retrieve CH₄ vertical profiles from hyperspectral Infrared Atmospheric Sounding Interferometer (IASI) data. The methodology combines the advantages of Optimal Estimation and neural networks, leveraging physical constraints to enhance retrieval accuracy while maintaining computational efficiency. The approach integrates dimensionality reduction techniques, such as Principal Component Analysis and Averaging Kernel optimization, to maximize information content while reducing computational complexity. Two models were developed: one for general profile retrieval and another optimized for detecting surface level enhancements. The novel approach was validated on synthetic data and applied to real IASI observations, including the Nord Stream pipeline sabotage event, and demonstrated that the algorithm can confidently detect CH₄ enhancements as low as 50 ppbv near the surface and reconstruct profiles with average error of ~20 ppbv in the mid-troposphere and >100 ppbv in the stratosphere on the considered test set. While effective in capturing strong enhancements, the technique exhibits slightly reduced sensitivity near the surface due to limited spectral information, which is mitigated by inflating covariance matrices in the cost function. This methodology offers a promising tool for global CH₄ vertical monitoring with existing sensors and could be potentially extended to other compounds such as CO₂.

Plain Language Summary Methane is a greenhouse gas that plays a significant role in global warming. We have developed a new method based on artificial intelligence to measure its distribution in the atmosphere using satellite data. This method can detect increases in methane emissions near the ground and provide a vertical profile of methane, constituting a powerful aid to monitor emissions from both human activities and natural sources.

1. Introduction

Methane (CH₄) is the third most relevant greenhouse gas in the Earth atmosphere (after H₂O and CO₂) and its increase in the atmosphere in the last decades is unequivocally contributed mostly from human activity and influence (Ciais et al., 2026; Environment, 2022; IEA, 2022). The present knowledge of CH₄ temporal trends and spatial distribution relies on the integration of several kinds of measurements. On the ground, the NDACC (Network for the Detection of Atmospheric Composition Change) and the TCCON (Total Carbon Column Observing Network) provide sparse yet accurate measurements of GHGs including CH₄ and its vertical profiles, by direct measurement of the Solar radiation extinct by the atmosphere (Washenfelter et al., 2003). The vertical sensitivity of these measurements does depend upon the Solar Zenith Angle (SZA) (M. Zhou et al., 2019) and is highest below 20 km of altitude. NDACC and TCCON measurements are a valuable source of information to locally constrain CH₄ vertical distribution and total column, and as a ground truth for other investigations (Ji et al., 2020). Yet, it has been demonstrated (see Palmer et al., 2019) that the number of measuring sites needs to be increased and optimized to achieve much needed precision in the characterization of the radiative impact of GHGs

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globally. In situ methane measurements have long records, are abundant and have been usually applied to specific issues and case studies. Some refer to emissions from specific marine ecosystems (Saderne et al., 2023) or lakes (Xun et al., 2024) in which eutrophication phenomena are linked to climate change or the impact of human activities. Other in situ methane measurements are linked to specific industrial activities, particularly those connected to oil extraction (Al-Shalan et al., 2022; Deighton et al., 2020). In urban areas, numerous mappings of methane emissions have been carried out using sensors on mobile vehicles (cars, bicycles): examples are in the areas of Toronto (Ars et al., 2020), Paris (Defratyka et al., 2021), Utrecht and Hamburg (Maazallahi et al., 2020), Bucharest (Fernandez et al., 2022). Data collected in these field campaigns are generally used to refine, increase and correct the inventory of local emissions, yet they are highly dependent upon the characteristics of the area examined, as each urban area is unique in its topography, infrastructure, and energy needs, and the emissions results presented in a study cannot be easily extended to others. Nevertheless, for the purpose of verifying and testing the emission inventory estimates on a local scale, methods such as the one proposed in Fratticioli et al. (2023) are effective: in that specific study, to evaluate and test the inventory of CH₄ emissions of the Po Valley basin, the authors use CH₄ and CO data collected at the Monte Cimone observatory, and then combine them with estimates of CO emissions from two other types of emission inventories. On the other hand, satellite observations provide information to bridge the gaps in the spatial distribution of ground-based measurements. In the last 20 years, several satellite-derived data products have been developed that include estimation of the total column of CH₄, as well as some vertical profiles or partial tropospheric columns and profiles of CH₄. Among the platforms that provide such measurements, there is the Infrared Atmospheric Sounding Interferometer (IASI, Hilton et al., 2012) on board the METeorological OPERational satellites (MetOp) platforms, whose data are used to provide the MUSICA (Multi-platform remote Sensing of Isotopologues for investigating the Cycle of Atmospheric water) retrievals of CH₄ profiles and columns (García et al., 2018; Schneider, Ertl, Diekmann, et al., 2022); the TROPOMI (TROPOspheric Monitoring Instrument) which provides official Level 2 products including the CH₄ total column (Lorente et al., 2021; Schneider, Ertl, Tu, et al., 2022); and the GOSAT (Greenhouse gases Observing SATellite) TANSO-FTS, which provides CH₄ total columns and profiles (Belikov et al., 2021). Documented and well-characterized data sets of retrieved CH₄ columns and profiles also exist for the NASA Atmospheric InfraRed Sounder (AIRS, Xiong et al., 2008; L. Zhou et al., 2023), and for the Cross-track Infrared Sounder (CrIS, Alvarado, 2015): they both reach a characterization of the vertical profile and total column of CH₄ with a retrieval precision that is similar to the one achieved by IASI, covering the same spectral interval with a similar resolution.

A comprehensive look at the estimated accuracy and type of product provided by ground and satellite data is shown in Figure 1. Current state of the art under-explores CH₄ profiling from hyperspectral data, and improvements to its derivation are valuable to improve the accuracy of retrieved CH₄ profiles globally. Specifically, IASI measurements have the potential to constrain the vertical profile of CH₄ (e.g., Siddans et al., 2017) to the point that its vertical profile can be retrieved with 2 degrees of freedom (as shown in, e.g., (Dils et al., 2024; de Lange & Landgraf, 2018; Schneider, Ertl, Diekmann, et al., 2022)).

Common approaches to retrieve vertical profiles are based on Optimal Estimation (OE, Rodgers, 2000; Liuzzi et al., 2016), where through a Gauss-Newton iterative scheme one can retrieve the CH₄ profile and other atmospheric and surface parameters. Advanced implementations of the OE exist for IASI and other hyperspectral sounders, where one exploits strategies to compress the data and parameter space with Principal Component Analysis (PCA) and even more refined approaches such as Random Projections (RP, Serio et al., 2018), which are greatly effective in treating biases. Yet, OE-based algorithms are affected by the significant bottleneck of the large amount of radiative transfer calculations needed for convergence. In many cases, this is solved by applying OE or iterative algorithms to a restricted portion of the spectrum where species of interest absorb (e.g., at a wavelength of 7.9 μm in the case of CH₄ with IASI). In turn, this forces one either to retrieve (e.g., with OE) or to assume the other atmospheric parameters (Temperature, H₂O, N₂O in particular in the 7.9 μm region) from climate and meteorological models, yielding biases in retrieved CH₄ profiles, recognized in literature (De Wachter et al., 2017). Those biases can be larger than CH₄ average interannual variability of ~20 ppbv (e.g., https://gml.noaa.gov/ccgg/trends_ch4/).

The group of operating and past satellite-monitoring instruments for CH₄ includes also a few platforms dedicated to directly observing the vertical structure of the atmosphere, providing quasi-direct measurements of the vertical profile of several trace species at variable altitude. Among them there is the ACE-FTS (Atmospheric Chemistry Experiment-Fourier Transform Spectrometer), which worked until 2019 in solar occultation geometry, and is a

	Ground-based		Satellite-based Nadir				Satellite-based Occultation and Limb		
	 NDACC - FTIR	 TCCON	 MetOp - IASI	 SSP - TROPOMI	 TANSO - FTS	 JPSS - CrIS	 Aqua - AIRS	 ACE-FTS	 ENVISAT-MIPAS
Type of product	Profiles (0 – 50 km)	Profiles (0 – 60 km)	Profiles (0 – 15 km) Column	Total column	Profiles (0 – 15 km) Column	Profiles (0 – 15 km) Column	Profiles (0 – 15 km) Column	Profiles (5–70 km)	Profiles (5–70 km)
Wavelength	2610–2950 cm ⁻¹ (3.39–3.83 μm) MIR	3900–15500 cm ⁻¹ (0.64–2.56 μm) MIR NIR VIS	1200–1350 cm ⁻¹ (7.40–8.33 μm) TIR	5880–6250 cm ⁻¹ (1.6–1.7 μm) NIR	1200–1350 cm ⁻¹ (7.40–8.33 μm) TIR	1200–1350 cm ⁻¹ (7.40–8.33 μm) TIR	1200–1350 cm ⁻¹ (7.40–8.33 μm) TIR	1245–2888 cm ⁻¹ (3.46–8.03 μm) MIR TIR	685–2410 cm ⁻¹ (4.15–14.6 μm) MIR TIR
Nominal Precision	Column: 1.5% (25 ppbv)	Column: 0.4% (6 ppbv)	Column: 1% (20 ppbv)	Column: 1% (20 ppbv)	Column: 1% (20 ppbv)	Column: 1.5% (30 ppbv)	Column: 1% (20 ppbv)	Column: N/A	Column: N/A
	Profile: 5–15% ⁽¹⁾	Profile: 1–3% ⁽²⁾	Profile: ~7% ⁽³⁾	Profile: N/A	Profile: 5% ⁽⁴⁾	Profile: AK only ⁽⁷⁾	Profile: ~5% ⁽⁶⁾	Profile: 10–25% ⁽⁵⁾	Profile: 10–30% ⁽⁸⁾

¹ values in troposphere and stratosphere, from Ji et al., 2020, doi:10.1007/s00376-020-9233-4
² values in troposphere and stratosphere, from Zhou et al., 2019, doi: 10.5194/amt-12-6125-2019
³ from Schneider et al., 2022, doi:10.5194/essd-14-709-2022
⁴ values in troposphere (<10 km), from Belikov et al., 2021, doi:10.3390/rs13091677
⁵ values from De Mazière et al., 2008, doi:10.5194/acp-8-2421-2008
⁶ values combining bias and accuracy of retrievals from Xiong et al., 2008, doi: 10.1029/2007JG000500
⁷ from CrIS Atmospheric Chemistry Data User's Workshop report, 2014, doi:10.7289/V50V8955
⁸ from Pleningner et al., 2016, doi:10.5194/amt-9-765-2016

Figure 1. Summary of current state-of-the-art capabilities of CH₄ monitoring: ground networks and satellite-based instruments. Nominal precision is the typical uncertainty associated with the retrieved quantity (profile or column). In some cases profile is not retrieved (“N/A”) or only Averaging Kernels are provided (“AK only”). The value of the precision reported for IASI is a derived value rather than being directly reported by Schneider, Ertl, Diekmann, et al. (2022).

gold standard for CH₄ vertical profiles from the upper troposphere to the mesosphere at high vertical sampling (De Mazière et al., 2008). Those profiles are however spatially sparse due to the geometry of the measurement, which is restricted to the immediate surroundings of the terminator and cannot sound the lower troposphere due to the high opacity in occultation because of the path length. ENVISAT-MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) has also produced a multi-year record of vertical profile measurements of several constituents including methane, with a precision comparable to the one typically achieved by ACE-FTS (Plie- ninger et al., 2016).

When using nadir satellite data, approaches like OE are very efficient at extracting and characterizing vertical information (when present). Yet, machine learning can also offer promising solutions to retrieve full vertical information about trace gases from nadir data, and overcome the issues related to the large computational re- sources needed to do that with iterative algorithms. As an example, machine learning schemes based on neural networks have been applied to IASI data to retrieve NH₃ column (Whitburn et al., 2016) and temperature and water vapor profiles (Malmgren-Hansen et al., 2019) demonstrating that retrievals with neural networks can be used to quickly process a large amount of data on a global scale, to account for a-priori information about the atmosphere and to provide a coherent estimation of the uncertainties and to document eventual retrieval biases. A significant example in this sense is also provided by MIIDAPS-AI, (Maddy & Iturbide-Sanchez, 2025), which is based on a deep neural network for hyperspectral retrieval that also deals with a-posteriori parameters' covariance information.

In this work, we build upon these latest results to set up new pathways to drive neural networks to correctly retrieve CH₄ profiles from clear-sky hyperspectral infrared satellite data, with an application tailored on IASI data. The application we present is potentially very challenging, as the CH₄ ν₄ band used in this work is located in a spectral region characterized by numerous interfering signals, including but not limited to H₂O (and isotopes), N₂O, CF₄, SO₂, and temperature effects.

The objective of this work is to combine the advantages of OE approaches, that is, the ability to provide strong constraints on convergence and the use of context information regarding the covariance of parameters to retrieve, and suitable neural network-based approaches, which have the undeniable advantage of speed in data processing.

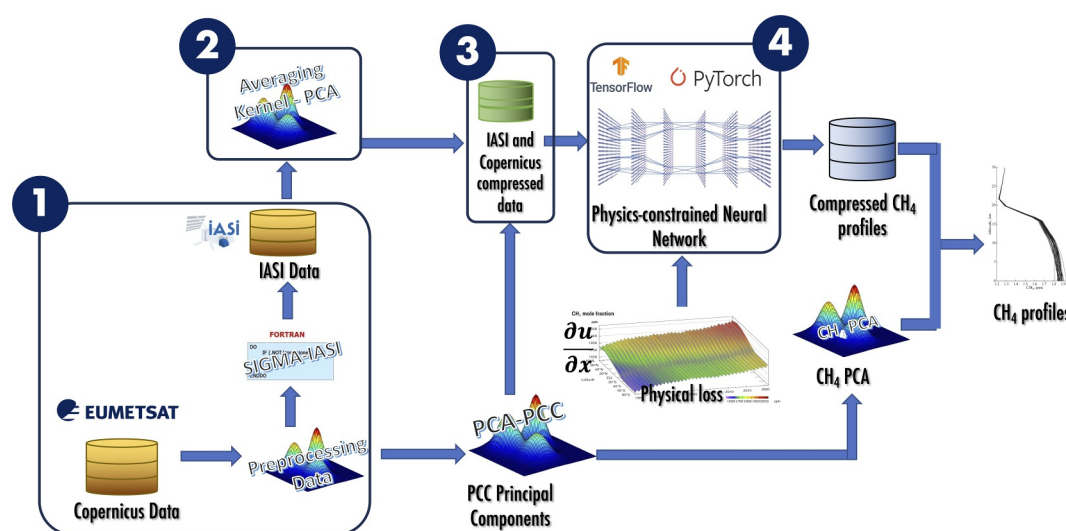


Figure 2. Outline of the scheme of training and retrievals based on the physics-constrained neural network used in this work. Parts 1–4 are illustrated in detail in the following subsections.

While OE algorithms typically take less than 10 s to invert IASI spectra for vertical profiles of temperature and gas constituents, the same problem is solved fractions of a second with neural networks. We combine the two approaches by introducing physics-based constraints in a neural network (from now on, simply “NN”), applied to an inverse problem using satellite data for the first time in this work.

The paper is structured as follows: in Section 2, we will explain what data have been used, and their pre-processing. Section 3 will present the architecture of the physics-constrained neural network elaborated in this work, and what physical constraints have been used. Section 4 will follow with sample retrieval results and the characterization of their accuracy. Conclusions will be drawn in the end.

2. Data and Methods

The retrieval scheme we present here is based on training a NN framework on simulated IASI data to retrieve CH_4 vertical profiles from IASI data. This is accomplished through several steps, beginning with the construction of a suitable data set of atmospheric profiles of CH_4 and other atmospheric profiles of gases whose absorption is relevant to the spectral interval covered by IASI. From those, we simulate IASI measurements using the σ -IASI/F2N Radiative Transfer Model (Masiello et al., 2024). The simulated spectral radiances (from now on, simply “radiance(s)”), along with other representative atmospheric state vector information for mid-range weather forecasting, are processed to create a data set (regressors) that serves as input and output for the NN, through which the vertical profiles of CH_4 are determined.

We show in Figure 2 the overall scheme of the NN training. This section is devoted to analyzing each step in detail and outlining the general methodology which is at the core of this work.

2.1. The Tool for IASI Data Simulation: The σ -IASI/F2N Model

The σ -IASI/F2N is an advanced radiative transfer tool for calculating radiances in the range $10\text{--}2760\text{ cm}^{-1}$, covering the entire spectrum from Far Infrared (FIR) to Near Infrared (NIR). The code was initially conceived and first published in 2002 (Amato et al., 2002), and it has since then undergone several stages of development, including the addition of several trace species (up to 12 different gases of variable concentration), the expansion of the spectral interval of operation to the FIR for application to future and current missions such as FORUM and PREFIRE (Kahn et al., 2020), and the latest update consisting in the addition of the capability to conduct accurate simulations of clouds and aerosols via the Tang and Chou schemes (Martinazzo et al., 2021), and with the inclusion of the Linear-in-T approximation to treat heavily inhomogeneous atmospheric layers.

Table 1

List of the Average Pressure Layers (60 Values) and Corresponding Level Boundaries Used in σ -IASI/F2N

Index	Edge [hPa]	Level [hPa]	Index	Edge [hPa]	Level [hPa]
0	1,013.0	—	31	175.17	187.6
1	1,004.99	1,009.0	32	150.0	162.6
2	1,000.0	1,002.5	33	122.05	136.0
3	985.76	992.9	34	100.0	111.0
4	972.71	979.2	35	85.23	92.6
5	959.7	966.2	36	70.0	77.6
6	945.74	952.7	37	64.67	67.3
7	932.82	939.3	38	59.8	62.2
8	925.0	928.9	39	57.5	58.6
9	912.95	919.0	40	55.29	56.4
10	900.0	906.5	41	53.17	54.2
11	874.73	887.4	42	51.14	52.2
12	850.0	862.4	43	50.0	50.6
13	833.17	841.6	44	48.79	49.4
14	813.83	823.5	45	47.29	48.0
15	795.0	804.4	46	45.84	46.6
16	748.23	771.6	47	40.0	42.9
17	700.0	724.1	48	30.0	35.0
18	649.78	674.9	49	25.0	27.5
19	600.0	624.9	50	20.0	22.5
20	549.96	575.0	51	15.0	17.5
21	500.0	525.0	52	10.0	12.5
22	465.63	482.8	53	7.01	8.5
23	432.01	448.8	54	5.0	6.0
24	400.0	416.0	55	3.0	4.0
25	349.93	375.0	56	2.0	2.5
26	300.0	325.0	57	1.0	1.5
27	274.95	287.5	58	0.5	0.8
28	250.0	262.5	59	0.1	0.3
29	225.0	237.5	60	0.005	0.1
30	200.0	212.5			

The model is applied to compute radiances in clear and cloudy sky conditions semi-analytically: low-degree polynomials are used to parametrize optical depths (OD) of gases with temperature, and optical properties of clouds and aerosols with the effective sizes of particles. This approach makes it possible to obtain analytical Jacobians too, which is relevant when it comes to performing retrievals through Optimal Estimation (Liuzzi et al., 2016; Serio et al., 2024). To make this possible, σ -IASI/F2N works on a fixed grid of 61 edges and 60 pressure levels (Table 1), on which calculations are always performed. The above-mentioned elements allow the model to maximize computational speed (taking 0.3s to simulate a single spectrum on modern CPUs), offering an efficient compromise between speed and accuracy. This makes it particularly suitable for operational applications, such as the one related to this work, where a high volume of simulations is required while maintaining a level of accuracy compatible with the requirements of space missions and atmospheric research. More details about the model can be found in published work (Masiello et al., 2024).

2.2. The Input Data Sources: Atmospheric Profiles

The initial task required to set the retrieval framework up is the creation of a large data set of simulated IASI radiances that can be used to train the scheme. To this end, we collected a large amount of vertical profiles of relevant gas species to feed the σ -IASI/F2N model. It is important to stress again that the retrieval scheme we build in this work is specifically designed to extract information about the vertical profile of CH₄, with a potential application to pinpoint, or even loosely constrain the eventual presence of surface sources (e.g. Bauduin et al., 2017). For this reason, the data set of simulated radiances we build considers only clear-sky scenes.

The atmospheric profiles data set was constructed based on the data available on the European Centre for Medium Range Weather Forecasts (ECMWF) databases. Data on skin temperature and specific humidity profiles were obtained from the Copernicus Climate Data Store ERA5 databases (Hersbach et al., 2020). These databases combine observations and advanced atmospheric models to provide an accurate global representation of surface and atmosphere. ERA5 provides a view of climate and weather since 1940 by integrating historical observations with physically based models, replacing the previous ERA-Interim re-analyses and significantly improving its quality and resolution. ERA5 data include hourly estimates of numerous atmospheric and surface parameters, with a spatial resolution of 0.25° for re-analyses and 0.5° for uncertainty estimates.

In this study, radiative transfer simulations require the following parameters:

- Skin temperature: it is extracted from the ERA5 hourly data from 1940 to the present (<https://cds.climate.copernicus.eu/datasets/reanalysis-era5-single-levels?tab=overview>).
- Specific humidity vertical profiles: they are obtained from the ERA5 hourly data on pressure levels data set (<https://cds.climate.copernicus.eu/datasets/reanalysis-era5-pressure-levels?tab=overview>).
- Atmospheric gases and temperature: data are downloaded from the Copernicus Atmosphere Monitoring Service (CAMS) (Agusti-Panareda et al., n.d; Granier et al., 2019; Inness et al., n.d; Peuch et al., 2022; Thompson et al., 2014) databases. In detail:
 - CO₂ and CH₄ are extracted from the CAMS global greenhouse gas reanalysis (EGG4) (<https://ads.atmosphere.copernicus.eu/datasets/cams-global-ghg-reanalysis-egg4?tab=overview>) data set;
 - CO, O₃ and Temperature profiles are extracted from the CAMS global reanalysis data set (EAC4) (<https://ads.atmosphere.copernicus.eu/datasets/cams-global-reanalysis-eac4?tab=overview>);
 - N₂O is obtained from the CAMS global greenhouse gas inversion (<https://ads.atmosphere.copernicus.eu/datasets/cams-global-greenhouse-gas-inversion?tab=overview>) data set.

This multi-dataset approach was specifically designed to ensure the highest possible fidelity for each atmospheric variable used in our simulations. The EGG4 data set was selected for CH₄ and CO₂ because it is a dedicated greenhouse gas reanalysis that actively assimilates satellite observations, providing tightly constrained concentration fields. Conversely, the EAC4 data set was chosen as it represents the state-of-the-art reanalysis for atmospheric thermodynamics (Temperature) and reactive gases (O₃, CO). Finally, since N₂O is not directly assimilated in the aforementioned data sets, we utilized the CAMS global greenhouse gas inversion product, which optimizes N₂O fields based on global surface observation networks.

For purposes of creating, testing and validating the NN retrieval scheme, we focused on a specific region encompassing the Mediterranean basin, extending from North Africa to Northern Europe, for the year 2020. To make sure that the NN is trained at its best to identify potential surface sources, the final data set should include a portion of profiles (and corresponding spectra) characterized by surface sources or noticeable slopes in the profile. To this end, new measurements were added from contexts with proven CH₄ sources, even if not referring to the area of interest, in order to make the data set less homogeneous so that the network can be trained with a substantial number of example sources.

The area of interest is defined by the following geographical boundaries: 60° North, 20° West, 20° South, and 20° East. Source subsets refer to:

- Subset 1: Deepwater Horizon Platform Disaster (2010);
- Subset 2: San Bruno Gas Pipeline Explosion (2010);
- Subset 3: Elgin Platform Accident (2012);
- Subset 4: Methane Leaks from Aliso Canyon Storage Reservoirs (2015–2016) and Emissions at Oil and Gas Fields in the U.S. (Permian Basin);

Table 2

Top: Subsets Created, With Reference Period, Region Considered, and Number of Measurements, Bottom: Overview of Data Sets Used, With Type of Data, Spatial, Temporal, and Vertical Resolution

Subset	Year, month	Region considered	Number of measurements	
Principal subsets	2020, all	N 70, S 25, W −20, E 42.25	1,359,072	
Subset 1	2010, 04-05	N 38, S 18, W −98, E −78	9,325	
Subset 2	2010, 09	N 47, S 27, W −132, E −112	2,260	
Subset 3	2012, 03-04-05	N 67, S 47, W −12, E 12	22,035	
Subset 4	2015, 10–11–12; 2016, 01–02	N 44, S 24, W −128, E −108	30,413	
Subset 5	2014, all; 2015, all; 2016, all; 2017, all; 2018, all	N 70, S 25, W −20, E 42.25	351,796	
Data set	Data	Spatial resolution	Time resolution	Vertical structure
CAMS ghg gas reanalysis	CO ₂ , CH ₄	0.75° × 0.75°	3-Hourly	60 model levels
CAMS global reanalysis	CO, T, O ₃	0.75° × 0.75°	3-Hourly	60 model levels
CAMS ghg gas inversion	N ₂ O	2.5° × 1.27°	3-Hourly	60 model levels
ERA5 single levels	Skin temperature	0.25° × 0.25°	Hourly	/
ERA5 pressure levels	Specific humidity	0.25° × 0.25°	Hourly	37 pressure levels

- Subset 5: Data for the area of interest, from 2014 through 2018. Measurements that did not refer to surface sources were removed from this subset, retaining only measurements with a higher concentration around 1,000 hPa. To minimize possible generalization errors, the subsets include profiles with surface enhancements up to 5 ppm, with a CH₄ concentration 4x higher than global average.

The final data set consisting of the 5 subsets created and the main subset related to 2020, which we will call “Principal subsets”, contains 1,774,901 measurements. Details of the subsets are given in Table 2. Figure 3 right, shows that the subsets exhibit a higher methane concentration near the surface compared to the Principal data set. The inclusion of these measurements has consequently increased the data set variance, with source-related observations now accounting for approximately 23% of the total. This fraction refers to all starting layers: for profiles with starting layer ≤ 3 , which include all the sea surface soundings analyzed in Section 3.2, the structurally comparable fraction of the training set is smaller.

2.3. Step 1: Data Pre-Processing and Radiance Simulation

The five data sources used in this work have different spatial resolutions (see Table 2). To ensure consistency among data sets, we filtered the profiles: specifically, N₂O required an additional preprocessing step because its

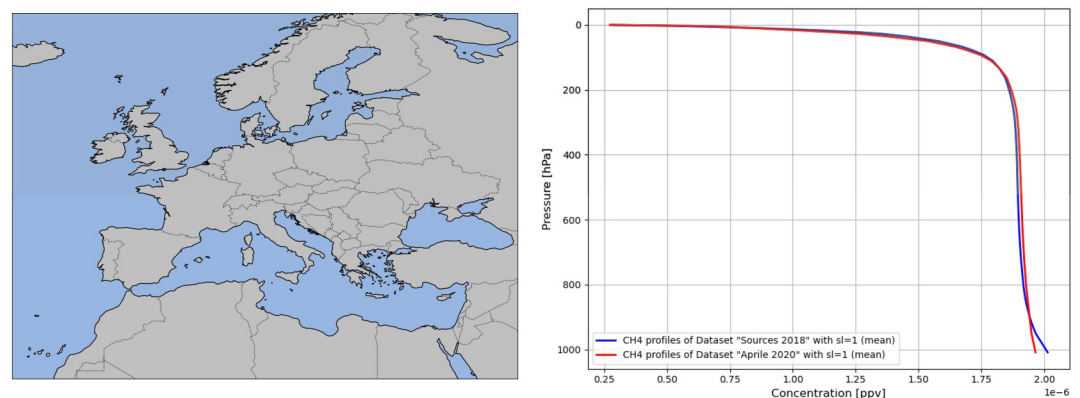


Figure 3. Left: Geographic area on which the data set was constructed, net of the extra set of profiles containing sources. Right: Comparison of the average of April 2020 methane profiles and the average of methane profiles characterized as sources for the entire year 2018. Both averages refer only to profiles with starting layer = 1.

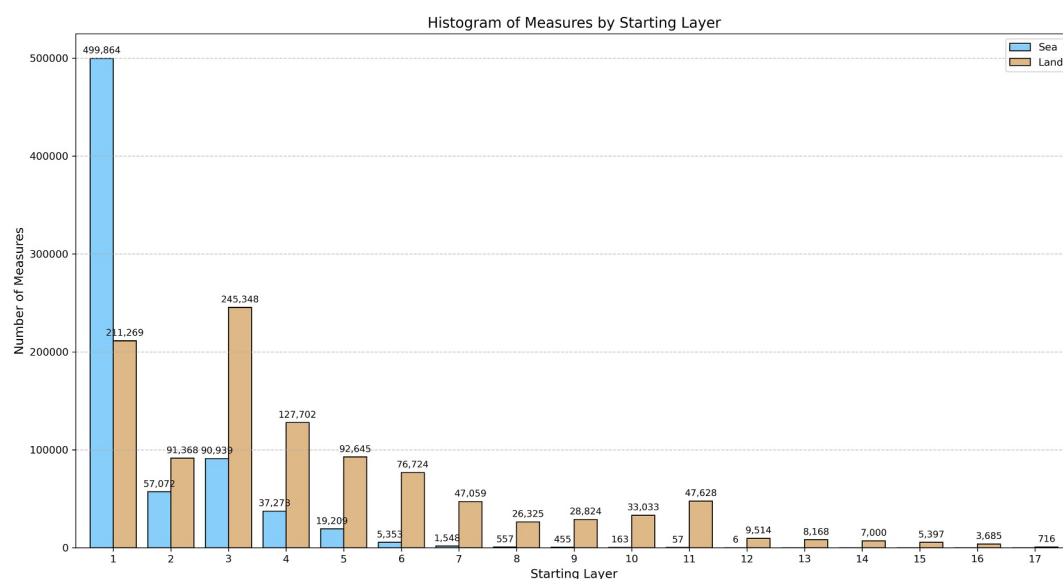


Figure 4. Histogram of the number of spectra according to starting layer, divided by land and water surface coverage.

spatial values rarely coincided exactly with those of the other data sets, showing discrepancies as large as 0.05° . To resolve this inconsistency, the N_2O data set was remapped to the coordinates of the CAMS data (data sets with lower spatial resolution). In other words, each mismatched measurement was associated with the value of the geographically closest N_2O relative to the same time instant. This technique is called Inverse Distance Weighting (G. Y. Lu & Wong, 2008). Then data sets were filtered by retaining only the common spatiotemporal measurements (i.e., same latitude-longitude pair and time). From the perspective of temporal resolution of the final data set, we chose a six-hour window, as a compromise between the CAMS data, which have a temporal resolution of 3 hours, and the ERA5 data, provided as hourly. Specifically, we chose the 4 synoptic times 00-06-12-18 UTC. All data sets refer to the 1,000-1 hPa pressure range. However, while the CAMS data include 60 vertical levels, the ERA5 data are given on 37 fixed pressure vertical levels only. Because the pressure grids differ from the σ -IASI/F2N forward model pressure grid (see Table 1), it was necessary to interpolate the data to the forward model layering.

As mentioned in the introductory section, IASI radiances, already widely used in the literature in trace gas studies (Atkinson et al., 2010), are simulated by the σ -IASI/F2N model. To obtain radiances routinely, σ -IASI/F2N is run through a Python wrapper.

This approach pays particular attention at simulating IASI-like clear sky radiances in all aspects, including the choice of Viewing Zenith Angles (VZAs), which during the simulations are randomly chosen from the list of 15 IASI-specific VZAs, which range from 0 to $\pm 48.3^\circ$. This procedure outputs a NetCDF file containing geophysical parameters and associated IASI-like simulated radiances (645 cm^{-1} – 2760 cm^{-1} , with a spectral sampling of 0.25 cm^{-1} , and a resolution of 0.5 cm^{-1}).

A normally distributed noise of amplitude as prescribed by IASI L1C noise specifications was added to the simulated spectra. It is important to note that for each radiance generated, the pressure range used for the simulation is also provided, which depends on the input data provided, relative to the starting layer.

The starting layer is found by matching the σ -IASI/F2N pressure grid layer closest to the surface pressure of a latitude-longitude pair, which varies over time. The data set used in this work includes 17 different starting layers, corresponding to the first 17 pressure layers from the surface of the σ -IASI/F2N pressure grid.

Summarizing, 1,774,901 spectra were generated, the detail of which is shown in Figure 4. The number of measurements associated with the lowest starting layers (i.e., at higher pressures) is significantly higher than the others, a distribution attributable to the geographic configuration of the study area, with water pixels including also high-altitude lakes.

2.4. Step 2: Data Dimensionality Reduction: The Averaging Kernel-Based Methodology

Once the atmospheric profiles and their associated radiances have been obtained, a preliminary operation needed to increase the efficiency of the scheme is to reduce the dimensionality of both radiance and profiles, in order to make the NN work with smaller arrays and reduce eventual redundancies in the data. A study based on the Averaging Kernel (AK) concept was applied to the spectra used in this paper. In this context, AK are used to quantify the information content of IASI data regarding CH₄ profile as a function of the spectral range considered and the instrumental noise. In the context of Optimal Estimation, the AK matrix, which we denote here with **A**, is defined as the derivative of the optimal estimate of the state vector (i.e., the parameters that are retrieved) with respect to the “true” set of state variables. That is a function of the a-priori covariance matrix of the noise, S_e , the background covariance matrix (associated with the entire state vector), S_a , and the Jacobian matrix of the radiance with respect to the parameters to retrieve, **K**, computed at the optimal solution:

$$\mathbf{A} = \frac{\partial \hat{\mathbf{v}}}{\partial \mathbf{v}} = (\mathbf{K}^T \mathbf{S}_e^{-1} \mathbf{K} + \mathbf{S}_a^{-1})^{-1} \mathbf{K}^T \mathbf{S}_e^{-1} \mathbf{K} \quad (1)$$

Importantly, **K** is analytically calculated by the σ -IASI/F2N model. The AK matrix provides also the degrees of freedom (DoFs) of retrieved parameters, namely how many pieces of independent information can be retrieved solely from the data without relying on the background and prior information provided to the algorithm. In the case of a vertical profile, the DoFs are obtained—by definition—by the trace of the AK matrix. Clearly, the values of the AK matrix and the DoFs will inherently depend on the spectral range considered, which may or may not include spectral information useful to constrain the information content of a vertical profile in a certain atmospheric pressure interval.

In this study, we therefore decided to use the quantification of the AK and DoFs to pinpoint a suitable spectral interval containing the maximum amount of information about CH₄ vertical profile. In some sense, this can be taken as a compression technique driven by the quantification of the information content of the data. This concept was applied to the measurements by starting from the core of the CH₄ ν_4 band in the infrared (at 1305 cm⁻¹) and extending the spectral range under analysis step by step. For each step, the AK matrix was computed, and from there the DoFs, with the expectation that they increase as the considered spectral interval around the center of the CH₄ band expands.

The study identified the spectral range between 1180 cm⁻¹ and 1430.25 cm⁻¹ as optimal for analysis, carrying 1.9 DoFs for the CH₄ profile and it was done on a sample of IASI-simulated radiances with various surface pressures (therefore, airmasses). Details about the results can be found in Supporting Information S1. Extending the spectral range beyond these limits does not yield a significant increase in information content. DoFs calculated for atmospheric pressures 300 to 1,000 hPa, roughly corresponding to the first 7 km above the surface, show that most of the information content comes from this vertical region. The AK analysis reveals therefore a sort of “sweet spot” in the choice of the spectral interval to consider, and allows to truncate the original spectra of 8,461 channels into a spectral range centered at the CH₄ band of only 1,001 channels (1,180–1,430 cm⁻¹), as can be seen from Figure 5, left. Furthermore, we report the diagonal of **A** (Figure 5, right panel) corresponding to the selected spectral interval, informing about how the vertical information is distributed in detail. This result denotes that the spectral information related to CH₄ is low at two specific points. The first is in the vicinity of the pressure levels immediately following the surface (around 1,000 hPa), which makes the identification of CH₄ sources a difficult problem to solve; the second concerns the layers around the 32nd level (150–170 hPa), of the IASI pressure grid, which is of less interest for studying CH₄ in the troposphere, which is the main focus of this work. Previous results dealing CH₄ retrievals with IASI and GOSAT (de Lange & Landgraf, 2018; Dils et al., 2024) show that the maximum sensitivity to CH₄ is generally achieved between 200 and 400 hPa, while our analysis shows highest sensitivity lower in the troposphere at 400–600 hPa. This discrepancy mostly reflects the different spectral windows used in each work: ours exploits a larger spectral interval than previous ones, including the critical 1,230–1,270 cm⁻¹ interval, which probes the lower troposphere more effectively.

2.5. Step 3: Spectra and Profiles Compression With Principal Component Analysis

The process of dimensionality reduction of data by PCA begins with spectral normalization (Equations 2–4), which is critical because radiances have a very large dynamic range, negatively affecting the results. To prevent

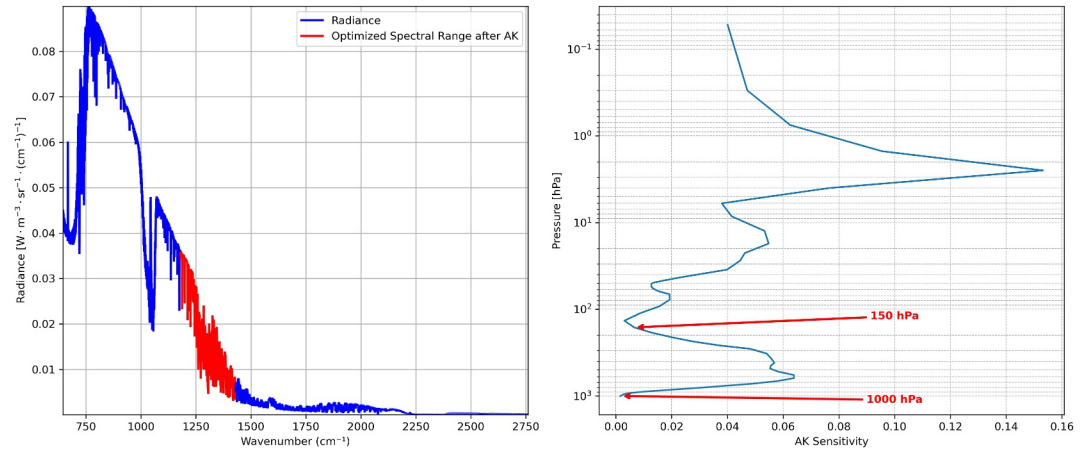


Figure 5. Left: Total spectrum and optimized spectral range on methane band obtained by Averaging Kernel. Right: AK diagonal profiles in the optimized spectral range. This specific example refers to radiances obtained by profiles with starting layer 1.

variables with larger values from dominating the analysis, the mean (Equation 2) of each variable is subtracted and divided by the standard deviation (Equation 3), resulting in a normalized $\mathbf{z}$ matrix (with zero mean and unit standard deviation). Next, we calculate the covariance matrix of $\mathbf{z}$, $\mathbf{C}$ (Equation 5), which allows to identify correlations between variables and understand which combinations of them explain the most variability in the data. Singular Value Decomposition (SVD), which is coincident to eigendecomposition since $\mathbf{C}$ is symmetric positive-semidefinite is then applied to $\mathbf{C}$, producing three matrices: $\mathbf{U}_R$, $\mathbf{S}_R$, and $\mathbf{U}_R^T$ (Equation 6):

$$R_{\text{mean}}(i) = \frac{1}{N} \sum_{j=1}^N R(i, j) \quad (2)$$

$$R_{\text{std}}(i) = \sqrt{\frac{1}{N-1} \sum_{j=1}^N [R(i, j) - R_{\text{mean}}(i)]^2} \quad (3)$$

$$z(i, j) = \frac{R(i, j) - R_{\text{mean}}(i)}{R_{\text{std}}(i)}, \quad i \in [1, \dots, M]; j \in [1, \dots, N] \quad (4)$$

$$\mathbf{C} = \frac{1}{N} \mathbf{z} \mathbf{z}^T \quad (5)$$

$$\mathbf{U}_R, \mathbf{S}_R, \mathbf{U}_R^T = \text{svd}(\mathbf{C}) \quad (6)$$

In the equations above, bold type indicates matrices, and:

- $R(i, j)$ is the element of the matrix $\mathbf{R}$ of dimension $M \times N$, containing the N radiances over $M = 1001$ wavenumbers;
- $\mathbf{C}$ is a $M \times M$ matrix, while $\mathbf{z}$ is of dimensions $M \times N$;
- svd denotes the calculation of SVD.

The SVD allows us to reduce the dimensionality of the data set by focusing on the components that explain most of the variability: the $\mathbf{U}_R$ matrix contains the principal directions in which the data vary the most (eigenvectors), while the $\mathbf{S}_R$ matrix contains singular values, which represent the amount of variance explained by each principal direction. It is important to note that in our case the basis $\mathbf{U}_R$ is employed to train and develop a retrieval scheme. For this reason, synthetic radiances must be used; otherwise, the unique physical link between the observations and the data space would be lost (Masiello et al., 2012). In Figure 6, the cumulative explained variances with increasing principal components are shown. For each spectrum belonging to the training data set, we then consider the $\mathbf{U}_R$ basis, and we project the spectrum into the basis, following the below equations:

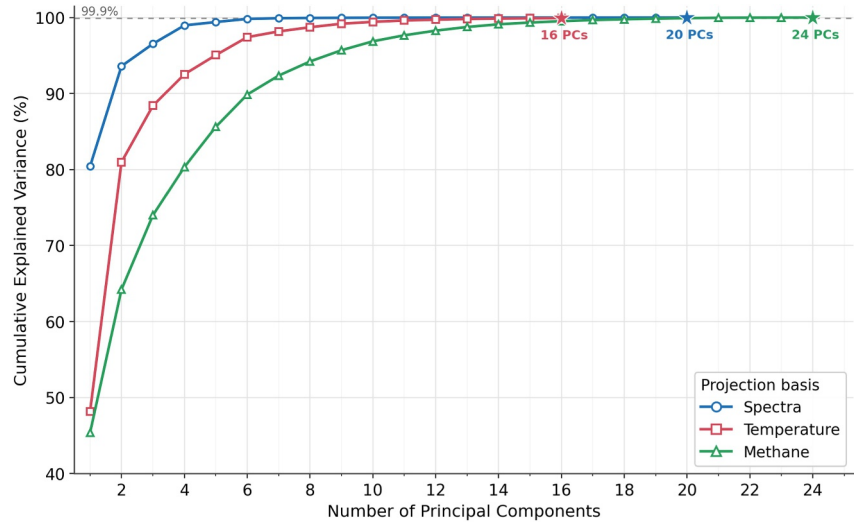


Figure 6. Cumulative explained variance as a function of the number of principal components for the three projection bases used in this work: AK-optimized spectra (blue), temperature profiles (red), and methane profiles (green). Stars mark the number of components retained (20, 16, and 24, respectively), all exceeding 99.9% of the explained variance (dashed line).

$$R(j)_{\text{norm}} = \frac{R(j) - R_{\text{mean}}}{R_{\text{std}}}, \quad j \in [1, \dots, N] \quad (7)$$

$$\mathbf{x}_j = \mathbf{U}_R^T R(j)_{\text{norm}} \quad (8)$$

where:

- $R_{\text{norm}}(j)$ is the normalized spectrum to mean and standard deviations;
- $\mathbf{U}_R$ is the truncated basis, a matrix of size $M \times N_{\text{PC}}$, with N_{PC} the number of components retained from the whole basis, made up of M eigenvectors;
- $\mathbf{x}_j$ is the array containing the values of the coefficients of the projection of $R_{j,\text{norm}}(\sigma)$ on the truncated basis $\mathbf{U}_R$.

The PCA reduces the dimensionality of the CH_4 band spectra, originally containing 1,001 wavenumbers, by projecting them into $N_{\text{PC}} = 20$ Principal Components (PCs), which are derived from the analysis whose result is shown in Figure 6. This reduction strikes an optimal balance between variability and dimensionality, retaining $> 99.99\%$ of the explained variance. PCA is also applied to the vertical profiles of CH_4 and temperature, with separate analyses conducted for each starting layer to account for variations in surface pressure. As mentioned earlier, the starting layer corresponds to the pressure layer closest to the surface pressure, and in this study the data, refer to 17 starting layers (1,013–748 hPa).

The projection bases are calculated by grouping all profiles associated with each starting layer, resulting in as many matrices as the number of starting layers in the data set. This aspect differentiates the projection process of vertical profiles from that for spectra. After normalization and covariance matrix computation, these matrices are decomposed using the SVD as before. Through the $U_{T,K}$ and $U_{\text{CH}_4,K}$ matrices the profiles are projected for a generic profile with a specific starting layer k ; indicating respectively with $q_{j,k}$ and $q_{j,k,\text{norm}}$ the j th vertical profile of CH_4 , and with $T_{j,k}$ and $T_{j,k,\text{norm}}$ the temperature ones with k th starting layer, and their normalized forms, we have:

$$q_{(j,k,\text{norm})}(p) = \frac{q_{j,k}(p) - q_{\text{mean},k}}{q_{\text{std},k}}, \quad j \in [1, \dots, N_k] \quad (9)$$

$$y_{j,k} = U_{\text{CH}_4,k}^T q_{j,k,\text{norm}}(p) \quad (10)$$

$$T_{j,k,\text{norm}}(p) = \frac{T_{j,k}(p) - T_{\text{mean},k}}{T_{\text{std},k}}, \quad j \in [1, \dots, N_k] \quad (11)$$

$$t_{j,k} = U'_{T,k} T_{j,k,\text{norm}}(p) \quad (12)$$

where:

- $q_{j,k,\text{norm}}(p)$ is the normalized j -th profile to mean and standard deviations of all N_k profiles with starting layer k , and it is a function of pressure p ;
- $U'_{(\text{CH}_4,k)}$ is the truncated basis, a matrix of size $M_L \times N_{\text{PC},\text{CH}_4}$, with $N_{\text{PC},\text{CH}_4}$ the number of components retained from the whole basis, made up of M_L eigenvectors, where $M_L = 60 - k + 1$ given the layering of the model made up of 60 layers;
- $y_{(j,k)}$ is the array containing the values of the coefficients of the projection of $q_{j,k,\text{norm}}(p)$ on the truncated basis $U'_{\text{CH}_4,k}$;
- Similar symbols are used for the temperature profile.

This approach ensures that the compression distinctly considers each profile class associated with the different starting layers, while best preserving the specific characteristics of each of them. The PCA of CH_4 and temperature shows that the PC numbers needed to obtain 99.9% of explained variance are $N_{\text{PC},\text{CH}_4} = 24$ and $N_{\text{PC},T} = 16$, respectively. As an example, Figure 6 (bottom panels) refers to the projection bases with respect to the starting layer 1 of methane and temperature. For the other starting layers results are almost identical, which allows us to take a fixed number of PCs and have a simpler and efficient network retrieval model homogeneous for all starting layers; this is indeed a necessary condition to construct the input and output tensors of the network and implement it.

In addition, with the idea of grouping the relevant quantities and arrays according to their starting layer, that is done also for covariance matrices of the profiles, $S_{(a,k)}$, resulting in a set of 24×24 matrices, one for each starting layer for CH_4 , and a set of 16×16 matrices for temperature, which we indicate here with $\Sigma_{(a,k)}$. To calculate them, we consider the covariance matrix of each set of profiles, and then we project them into the corresponding basis:

$$S_{a,k} = \frac{1}{N} (\mathbf{q}_k(p) - \mathbf{q}_{\text{mean},k}) (\mathbf{q}_k(p) - \mathbf{q}_{\text{mean},k})^T \quad (13)$$

$$\Sigma_{a,k} = U'_k S_{a,k} U_k^T \quad (14)$$

Because the eigenvectors of each basis are orthogonal by construction, the result will be a set of matrices $\Sigma_{(a,k)}$ which are diagonal. Their use will be discussed in detail in Section 2.6.2, which describes the cost function formulation and implementation.

2.6. Step 4: Physics-Constrained Neural Network Setting and Construction

Before detailing the construction of the NN, we briefly discuss here the selection of the input parameters to the network. The selection of features to be used in the composition of the input regressors is done using a Pearson correlation coefficient analysis (Guyon & Elisseeff, 2003; Schober et al., 2018), supplemented with assessments regarding the nature and composition of the data. For example, although VZA, latitude and longitude have a negligible correlation with CH_4 concentration, they were still included: radiances were generated with random values of VZA among the set of IASI observation angles, because two radiances simulated with the same atmosphere at the same place and time, but with different VZA, will be slightly different, as transmittance depends upon $\exp(-OD/\cos(VZA))$. The low correlation occurs because VZA, per se, is not directly related to CH_4 concentration, but it may be useful for the network to detect hidden patterns (Crowley & Aboubakr, 2006) between radiance and CH_4 concentration. The input regressor consists of features related to spectrum, temperature, latitude, longitude, starting layer, time and VZA (see Figure 7).

Unlike CO_2 , CH_4 emissions have a more complex geographic distribution and do not show a strong correlation with latitude. While CO_2 emissions are closely associated with industrial and human activities, which tend to be concentrated in specific latitudinal regions (Helyer & Courtney, 2018), CH_4 sources, both natural (e.g., wetlands)

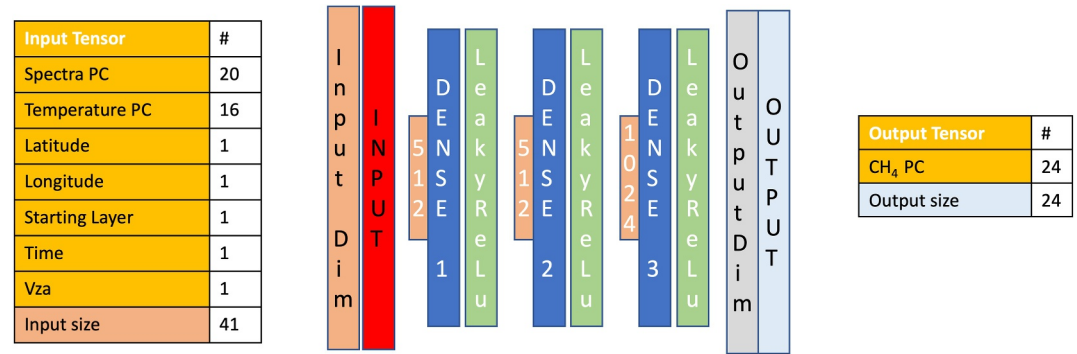


Figure 7. Physics-constrained neural network structure with associated input (left) and output (right) tensors.

and anthropogenic (e.g., agriculture and natural gas extraction), are influenced by a variety of factors, including climate and agricultural practices, rather than by latitude itself. Therefore, CH₄ emissions do not follow a geographically uniform distribution pattern related to latitude (Saunois et al., 2020). However, in this work, both latitude and longitude are included as inputs to enable the network to possibly pick up on hidden correlation with sources, thus aiding their identification. The sensitivity of the retrieved profiles to the geolocation inputs is assessed in Supporting Information S1, by perturbing latitude and longitude while keeping the measured spectra fixed.

2.6.1. Physics-Informed Neural Networks (PINNs): Fundamentals and Relation to This Work

PINNs are neural networks designed to solve problems governed by differential equations, such as partial differential equations (PDEs). Instead of relying exclusively on labeled data, PINNs incorporate physical knowledge, represented by mathematical equations, into their loss function, combining machine learning approaches with physical constraints (Cuomo et al., 2022). A problem governed by a PDE can be expressed as:

$$\begin{cases} F(u(z); \gamma) = f(z), & z \in \Omega, \\ B(u(z)) = g(z), & z \in \partial\Omega. \end{cases} \quad (15)$$

where:

- $z = [x_1, \dots, x_{d-1}; t]$ represents space-time coordinates;
- $u(z)$ is the unknown solution;
- γ represents physical parameters;
- F is the nonlinear differential operator that describes the system;
- B represents the initial or boundary conditions.

The neural network approximates the solution $u(z)$ by $\hat{u}_\theta(z)$, where θ represents the network parameters. The overall loss function is:

$$L(\theta) = \omega_f L_f(\theta) + \omega_B L_B(\theta) + \omega_d L_{data}(\theta) \quad (16)$$

where the total loss function $L(\theta)$ consists of the physics-informed loss L_f (computed from the physical residuals r_F), the boundary condition loss L_b (computed from the residuals R_B), and finally the data loss L_{data} evaluating the discrepancy between the training data and the NN predictions. Residuals r_F and R_B can be defined as:

$$r_F \hat{u}_\theta = F(\hat{u}_\theta(z); \gamma) - f(z) \quad (17)$$

$$R_B \hat{u}_\theta = B(\hat{u}_\theta(z)) - g(z) \quad (18)$$

PINNs represent an innovative and versatile approach that combines machine learning with the laws of physics described by a differential equation F directly into the loss function, performing well even with limited or noisy

data. In this work, we adapt this approach in an original way to solve an inverse problem, working on a formulation of a physical condition simpler than the introduction of a full differential equation in the network training.

The initial configuration of the neural network model is obtained using AutoKeras, an advanced automated machine learning (AutoML) framework, which allows different neural network architectures to be automatically tested and quickly explored to identify the one best suited to the data provided (Jin et al., 2019; Zöller & Huber, 2021). This strategy can be very useful in complex problems such as the one under consideration.

To make sure that no input variables disproportionately affect the learning process, the data are normalized using the StandardScaler function of the sklearn. preprocessing library. This procedure ensures that all features have zero mean and unit standard deviation, making each piece of data comparable to each other and improving numerical stability during training. Accurate normalization is particularly important in deep learning models because it helps prevent features with different scales from dominating the optimization of network weights (Lecun et al., 1998). The neural network model obtained through AutoKeras is structured in three dense layers; each layer has a Leaky ReLU activation function, which is useful to limit the “dying neurons” problem (L. Lu et al., 2020). The dimensions of the input layers are as follows:

- First layer: 512 neurons;
- Second layer: 512 neurons;
- Third layer: 1,024 neurons.

Starting from this basic implementation, the model was further customized to include physical constraints specific to the problem under analysis and thus obtain a network capable of predicting the PCs of CH₄ vertical profiles from an input tensor as shown in Figure 7.

2.6.2. Loss and Cost Function: A Physics-Based Approach

The cost function measures the difference between the true value and the value predicted by the network (see Equation 19). The cost function is an intrinsic quantity needed by any inversion scheme, whether it is OE or AI-based, as a metric to drive the scheme toward the optimal solution. We resort to a scheme where we can exploit physical constraints on the system to impose a cost function which is physically based. The obvious choice in this sense would be to use as cost function the full chi-square (χ^2) as implemented in Optimal Estimation (Carissimo et al., 2005; Rodgers, 2000; Serio et al., 2024), however that presents the issue that one needs to have a forward scheme that computes Jacobians and radiances, which the NN does not do in this work. In fact, the objective is to have just the inverse model without the burden of a forward model, which typically is the bottleneck of retrievals.

For these reasons, the implementation of the physical condition in the cost function of our scheme is given by the parametric term of the traditional (χ^2), which acts as a differential term to drive the solution to a physically reasonable one. By embedding this into the loss function, the network is forced to find solutions that are physically consistent with atmospheric thermodynamics and chemistry. As noted by Karniadakis et al. (2021), using such prior physical knowledge to guide the learning process is a defining characteristic of “Physics-Informed” systems. In our case, prior knowledge (like the covariance of atmospheric states) is used to drive the learning process: differently from “Hard Constraints” (where PDEs are built into the architecture) we adopt a “Soft Constraint” approach: rather than a differential physical formalism, the integration of an established, physically-based prior (Equation 19) into a fast neural network inverse model is at the core of this work.

In detail, given $\Sigma_{a,k}$ the covariance matrix of any given CH₄ profile starting from layer k , as defined in Equation 13, we define the additional term Q to the cost function in the following way during the training phase:

$$\mathbf{Q}_k = (\mathbf{q}_{k,\text{est}}(p) - \hat{\mathbf{q}}_k(p)) \Sigma_{a,k} (\mathbf{q}_{k,\text{est}}(p) - \hat{\mathbf{q}}_k(p))^T \quad (19)$$

$$\mathbf{Q} = \frac{1}{N_{\text{sl}}} \sum_{k=1}^{N_{\text{sl}}} \mathbf{Q}_k \quad (20)$$

where:

- $\mathbf{Q}_k$ is the cost function term computed for the profiles of the training set with starting layer k ;
- $\mathbf{q}_{k,\text{est}}(p)$ is the profile as estimated by the NN in the training phase, where the real profile $\hat{\mathbf{q}}_k(p)$ is known;
- N_{sl} is the number of starting layers in the training data set.

The covariance matrix $\Sigma_{a,k}$ is used in literature to optimize parameters in evolutionary scenarios; this concept can also be applied to deep learning models to optimize cost functions (Willjuice Iruthayarajan & Baskar, 2010). The total loss or losses for a training epoch is calculated as the average of the individual losses of all examples in the batch (see Equation 20). This approach provides a global measure of model error, balancing the impact of any anomalies or discrepancies in individual samples. Losses provide a scalar representation of model performance, which can be used to guide optimization during the training process.

The final loss was handled by the network using the Adam (Adaptive Moment Estimation) optimizer, which combines the benefits of stochastic gradient descent (SGD) with adaptive estimation of first- and second-order moments, that is, the mean and variance of the gradients. Adam updates the model weights using not only the gradient of the loss function but also the moving averages of the first and second moments. This approach allows adaptive learning speed for each parameter, improving convergence and reducing the impact of extreme or noisy gradients (Kingma & Ba, 2017).

2.6.3. The Need for Two Models

Using the AutoKeras and TensorFlow libraries, a Python code was developed to implement two models. The need for two models is justified by the analysis performed in Section 2.4 with the Averaging Kernels, which highlighted that IASI data have lower sensitivity to the shape of CH_4 profile near the surface, therefore diminishing the capabilities of a retrieval approach to detect surface sources if present.

To overcome this limitation, we borrow a typical approach followed in OE, which consists in increasing the covariance of the profile at pressure levels where data are less sensitive to it (for example, Liuzzi et al., 2016). While this gives more freedom of searching for the optimal solution in the atmospheric regions in which covariance is increased, generally this will not correspond to an increase in the total DoFs, which are dominated by instrumental noise and resolution. Therefore, this approach is ideal only to obtain a better identification of surface sources, with some loss of accuracy in reconstructing the whole profile. Keeping this in mind, we created two different retrieval models, which have been trained with two different sets of covariance matrices $\Sigma_{a,k}$:

- The first model, labeled Model 0, was designed to predict the vertical profile of CH_4 and its general shape. It uses a cost function which incorporates the covariance matrix $\mathbf{S}_{(a,k)}$ obtained from the data set of training profiles;
- The second model, labeled Inflated model, was optimized to estimate the surface concentration of the gas, and thus the identification of its sources. It uses a cost function which incorporates the covariance matrix obtained from the data set of training profiles, but with the elements corresponding to the first 10 layers above the surface multiplied by 10 (Equation 21). This is done to give more weight to the lowest pressure layers and to introduce an artificial correlation between the first pressure layers and the next ones, in order to facilitate the identification of surface CH_4 sources.

$$\mathbf{S}_{(a,k)}(p_i, p_j) = \begin{cases} \mathbf{S}_{(a,k)}(p_i, p_j) \cdot 10, & \text{if } 1 \leq i, j \leq 10, \\ \mathbf{S}_{(a,k)}(p_i, p_j), & \text{otherwise.} \end{cases} \quad (21)$$

3. Results and Statistics

3.1. Simulated Test Data Set

This section presents the results of the implemented models. The test was conducted on methane profiles, obtained by backprojecting the predicted PC scores. Each output tensor ($\mathbf{y}_{r,j,k}$), of size $N_{\text{PC},\text{CH}_4} = 24$ (corresponding to the number of PCs related to the methane profile), was subjected to the back-projection process (i.e., the reverse projection process). Given the matrix $\mathbf{U}_{\text{CH}_4,k}$ containing the principal components associated with the k -th starting layer, and the vector of predicted scores $\mathbf{y}_{r,j,k}$, the normalized reconstructed profile $\mathbf{q}_{r,j,k,\text{norm}}(p)$ is obtained by

Table 3

Test Metrics Related to the Developed Models, Using the Principal Components Obtained Through the Test Set Back-Projected Into the IASI Pressure Grid

Metrics	Inflated model	Model 0
RMSE	4.5147×10^{-8} ppv	4.5268×10^{-8} ppv
MAE	2.6447×10^{-8}	2.6532×10^{-8}
Relative error	2.79%	2.79%
R^2 score	99.18%	99.17%

projecting the scores back into the original feature space through matrix multiplication. This operation reverses the dimensionality reduction applied during the projection phase.

To recover the physical units of the original profile, the normalized profile is denormalized using the mean $\mathbf{q}_{\text{mean},k}$ and standard deviation $\mathbf{q}_{\text{std},k}$ associated with the k -th starting layer as follows:

$$\mathbf{qr}_{j,k,\text{norm}}(p) = \mathbf{U}_{\text{CH}_4,k} \mathbf{yr}_{j,k} \quad (22)$$

$$\mathbf{qr}_{j,k}(p) = \mathbf{qr}_{j,k,\text{norm}}(p) \cdot \mathbf{q}_{\text{std},k} + \mathbf{q}_{\text{mean},k} \quad (23)$$

where:

- $\mathbf{qr}_{j,k,\text{norm}}(p)$ is the normalized retroprojected j -th profile to mean and standard deviations of all N_k profiles with starting layer k , and it is a function of pressure p ;
- $\mathbf{U}_{\text{CH}_4,k}$ is the truncated basis, a matrix of size $M_L \times N_{\text{PC},\text{CH}_4}$, with $N_{\text{PC},\text{CH}_4}$ the number of components retained from the whole basis, made up of M_L eigenvectors, where $M_L = 60 - k + 1$ given the layering of the model made up of 60 layers;
- $\mathbf{yr}_{j,k}$ is the array containing the values of the coefficients found through the neural network model;
- $\mathbf{qr}_{j,k}(p)$ is the j -th profile back-projected onto the IASI pressure grid.

The analysis is based on established metrics, the root mean square error (RMSE) and relative error. Table 3 reports the values of the metrics calculated for the two models developed, for the entire vertical profile, consisting of the 60 atmospheric layers of the σ -IASI pressure grid. Both models were implemented and evaluated on the same data set divided as follows:

- 70% training-set;
- 15% validation-set;
- 15% test-set.

Table 3 shows that the two models, although built with a slightly different cost function to meet different objectives, provide almost identical results. For this reason, the study continued with the use of the Inflated model, which was originally designed for the identification of methane surface sources, but is also found to be very accurate for vertical gas profiling. The metrics considered, RMSE, MAE (Hodson, 2022), R^2 , relative error, are found to be slightly better for the Inflated model. Figure 8 shows the overall statistics of the Inflated model computed on our test-set for 3 sample starting layers. It can be seen that the variance of the data decreases as the pressure characterizing the starting layer decreases, and as correspondently the number of measurements associated with it decreases. The mean of the original profiles matches perfectly, confirming the results shown in Table 3.

It is important to quantify the uncertainty affecting the retrieved vertical profile of CH_4 , even if the NN architecture does not inherently provide it. In particular, the standard deviation of the differences between true and retrieved profiles, computed for each starting layer, can be considered a good indicator of the cumulative 1σ uncertainty affecting the retrieved profiles. Minding that the plots in Figure 8 report this difference multiplied by 10, we can say that the retrieved profiles have their lowest uncertainty in the mid-troposphere (where most of the signal in IASI spectra in the considered band comes from), which can be as low as 20 ppbv (or about 1%). The uncertainty increases to 100 ppbv (or 5%) in the stratosphere. Importantly, we can argue that the standard deviation of the differences between true and retrieved profiles in the lowest atmospheric layer is an indicator of

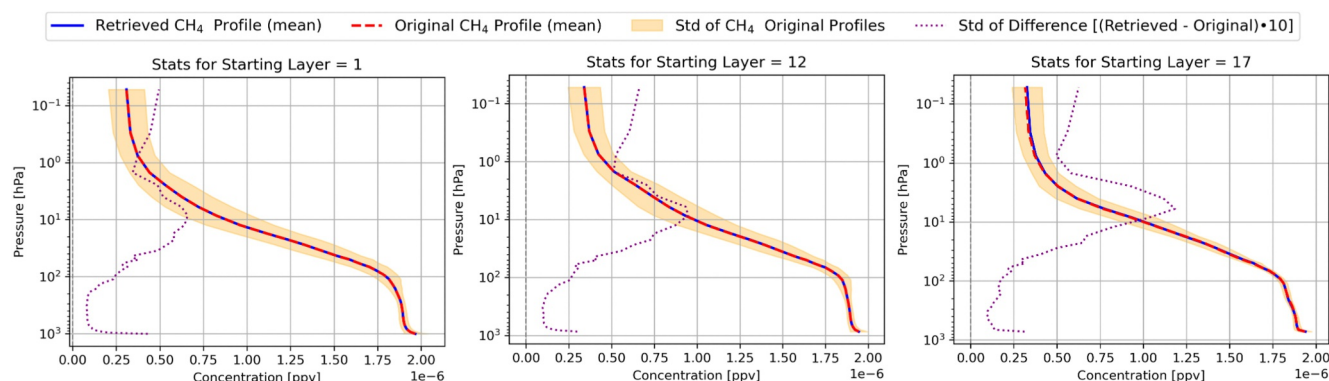


Figure 8. Evaluation of the Inflated model on the test set, for some sample classes defined by the starting layer. For each class, identified by the starting layer, the average of the original CH₄ concentration profiles (dashed red line) and the average of the profiles reconstructed by the model (solid blue line) were calculated. The orange shaded area represents the standard deviation of the original profiles within the class.

what level of CH₄ enhancement with respect to the atmospheric background we can measure with this framework applied to IASI data: this level is equal to about 50 ppbv, which can constitute a reference threshold to discriminate whether eventual near-surface enhancements retrieved with this technique are credible. In the Supporting Information S1 we also quantify the variance in the training data set of CH₄ versus N₂O as examples of interfering species, showing that 78% of the combined two-gas variance is due to CH₄.

3.2. Test Case: The Nord Stream Explosion Event

We validated the models on real IASI radiances on a specific test case focusing on northern Europe (50–75°N, 5°W–25°E), a region including the Baltic Sea near the island of Bornholm and the North and Norwegian Seas, for the period between late September and early October 2022, coinciding with the underwater explosions involving the Nord Stream gas pipeline. This specific test aims to evaluate the behavior of the Inflated model under extreme and routine conditions for our model, trying to identify surface sources and the evolution of atmospheric concentration over time. IASI radiances were downloaded via the EUMDAC service. Ancillary atmospheric information were retrieved with the δ -IASI model (Liuzzi et al., 2016; Serio et al., 2024), which is an Optimal Estimation-based retrieval scheme, through which we derived atmospheric temperature profile as inputs to the NN. We chose four time intervals over which to conduct the analysis:

- T1: September 23–September 25 (until the morning overpass), pre-event period (which is an example of a routine case);
- T2: September 26–September 27, explosions and first day of the release;
- T3: September 28–September 29, continuing release and transport of the plume;
- T4: September 30–October 3, post-event phase, characterized by interventions to stop gas leaks.

We extracted all pixels of IASI orbits falling in the region of interest and in the four defined time intervals, and retained those acquired under clear sky, over both sea and land surfaces. The NN is trained on both land and water scenes (Figure 4), and no systematic discontinuity across coastlines is found in the results (Figure 9). Each profile is backprojected individually, using a different basis depending on the reference starting layer. Each spectrum of each of the four subsets was then ingested and analyzed by the Inflated model.

To get an immediate view of the results, we computed, for each IASI sounding, the column-averaged CH₄ mixing ratio between the surface and 900 hPa (hereafter, near-surface partial column) from the retrieved profile; soundings with surface pressure lower than about 930 hPa are therefore excluded. As discussed in Section 2.4, the information content of the total column comes mostly from the mid-troposphere, which makes it much less sensitive to near-surface enhancements.

Considering the estimate of 450,000 tonnes of CH₄ released during the whole event (Mohrmann et al., 2025), and an area immediately interested by the release of about 10⁶ km², and the first 3 km of atmosphere above the surface, that amounts to an average enhancement of $9 \cdot 10^{-6}$ mol/m³, or 11% the natural background (200 ppbv). This value represents an upper estimate of the average enhancement one could expect, as it assumes that the whole

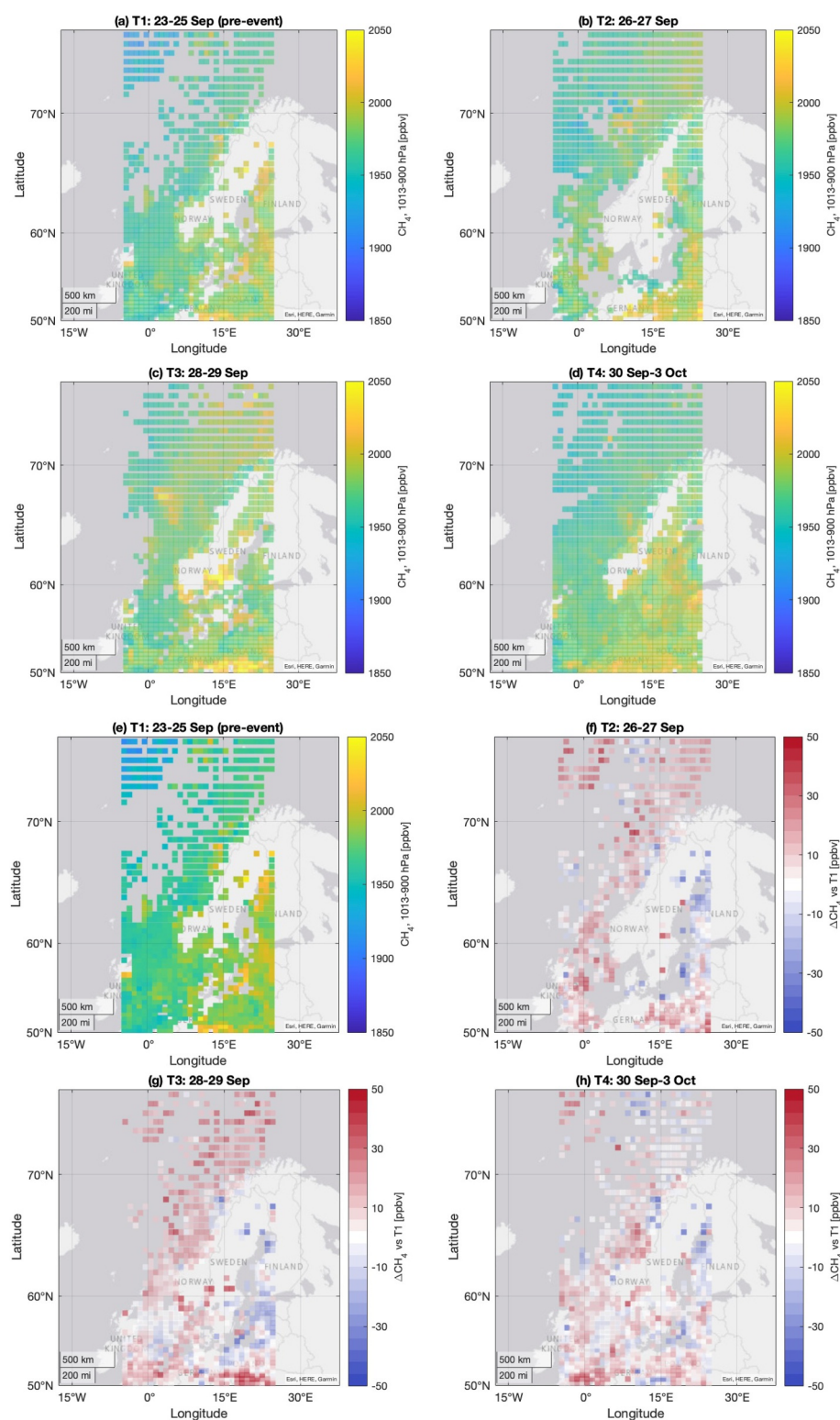


Figure 9. Top (a–d): Maps of the near-surface partial column (column-averaged CH_4 mixing ratio between the surface and 900 hPa) for the four time intervals chosen for analysis, averaged on a regular $0.5^\circ \times 1^\circ$ grid. Bottom (e–h): Variations of the near-surface partial column for time intervals T2, T3 and T4 compared with T1, used as the “background map.”

released mass is confined in that volume at the same time, and is compatible with the near-surface enhancements of a few tens of ppbv that we observe after spatial and temporal averaging (see below). Enhancements up to 200 ppbv are retrieved by (Wilson et al., 2024), using IASI data, and estimating a CH₄ release up to 427,000 tonnes. We should note, though, that our work is the only one dealing with atmospheric profile, and not just column retrieval.

A detailed view of the results at every IASI passage, together with the comparison between column abundances derived from this work and those derived by (Wilson et al., 2024) are shown in Supporting Information S1. To get more insights into the temporal evolution of CH₄ near-surface partial column over the whole region, we regridded the data on a regular spatial grid of $0.5^\circ \times 1^\circ$ (about 55×55 km at 60°N), averaging the values for each time interval (for each IASI overpass, we averaged the soundings falling in each grid cell, and then averaged the overpasses of each time interval with equal weights), obtaining a set of Level 3 maps. Figure 9 shows in the top panels the maps of the near-surface partial column, regridded on a regular spatial grid for each time interval. Before the explosions (T1), the near-surface partial column shows a persistent contrast between maritime air over the North Atlantic and the Norwegian Sea (about 1950 ppbv) and continental air over the Baltic region and central Europe (about 1990–2010 ppbv). During T3, values up to about 2050 ppbv are found over the Norwegian Sea off the Norwegian coast and over southern Scandinavia. Subtracting T1 from the subsequent maps (T2, T3, T4) it is finally possible to highlight the differences attributable to Nord Stream pipeline sabotage (Figure 9, bottom): this representation highlights that the enhancement of CH₄ near the surface at its peak (T3) is widespread over the Norwegian Sea, between about 62 and 72°N , with anomalies up to 40–50 ppbv relative to T1, while negative anomalies are found over the eastern Baltic Sea and the Gulf of Bothnia. Positive anomalies found over southern Poland are not related to the release, but most likely to the production of coking coal in Upper Silesia. Although all anomalies in Figure 9 are mostly below the 50 ppbv threshold derived in Section 3.1, the latter refers to individual soundings, whereas each value in the maps is the average of many soundings, which reduces the random component of the retrieval uncertainty. Yet, in the single-overpass maps reported in Supporting Information S1, individual soundings over the Norwegian Sea exceed the pre-event background by up to about 150 ppbv, well above the 50 ppbv threshold. It also helps to identify the areas of highest enhancement, located north-west of the release area, also seen in other studies, such as Wilson et al. (2024): as noted therein, the local circulation following the explosions transported a large part of the plume north-westward. This explains the enhancements observed in that part of the domain and the gradients seen at T4. Even if this is out of the scope of the present paper, the use of the retrieval framework illustrated in this paper could, in principle, allow for monitoring the 3-dimensional circulation patterns of CH₄ over large regions such as the one used in this case study. Nevertheless, we can focus here on the analysis of the time evolution of the average derived methane vertical profiles in two regions affected by the plume. To this end, we show in Figure 10 the daily average of retrieved vertical profiles of methane, calculated at the two IASI satellite acquisition time slots (morning and evening), to get a focused view of the temporal evolution of CH₄ concentration and enhancements over the Norwegian Sea (60 – 75°N , 5°W – 10°E), where the plume was transported according to Wilson et al. (2024), and over the southern Baltic Sea (52 – 58°N , 15 – 23°E), which includes the release area.

It should be noted that the average at each pressure level is computed over the soundings whose starting layer lies at or below that level, so that near the surface the set of contributing soundings changes from one overpass to another. In the southern Baltic Sea, in particular, the lowest layer is not sampled by clear-sky soundings between the evening of 26 September and the morning of 29 September, that is, during the most intense phase of the release. Therefore, the analysis was conducted while maintaining the atmospheric conformation of the domain, that is, respecting the spatial variability of surface pressure.

In detail, over the Norwegian Sea we observe a near-surface enhancement between 27 and 29 September, reaching about 25 ppbv relative to T1 on the evening of 28 September and confined below about 950 hPa. Over the southern Baltic Sea, a second enhancement is observed on October 1–2, reaching about 35 ppbv near the surface and mostly confined below about 850 hPa, in line with what has been shown in previous analyses from ground stations (Jia et al., 2022). The weaker positive anomaly found above 900 hPa in both regions after 28 September is nearly uniform with height and does not show the near-surface maximum expected from a surface source, therefore it cannot be straightforwardly attributed to the release.

Similarly to what is done for the spatial maps, we performed also an analysis of the concentration change attributable to the emissions caused by the explosions, using the average vertical profile of T1 as a background

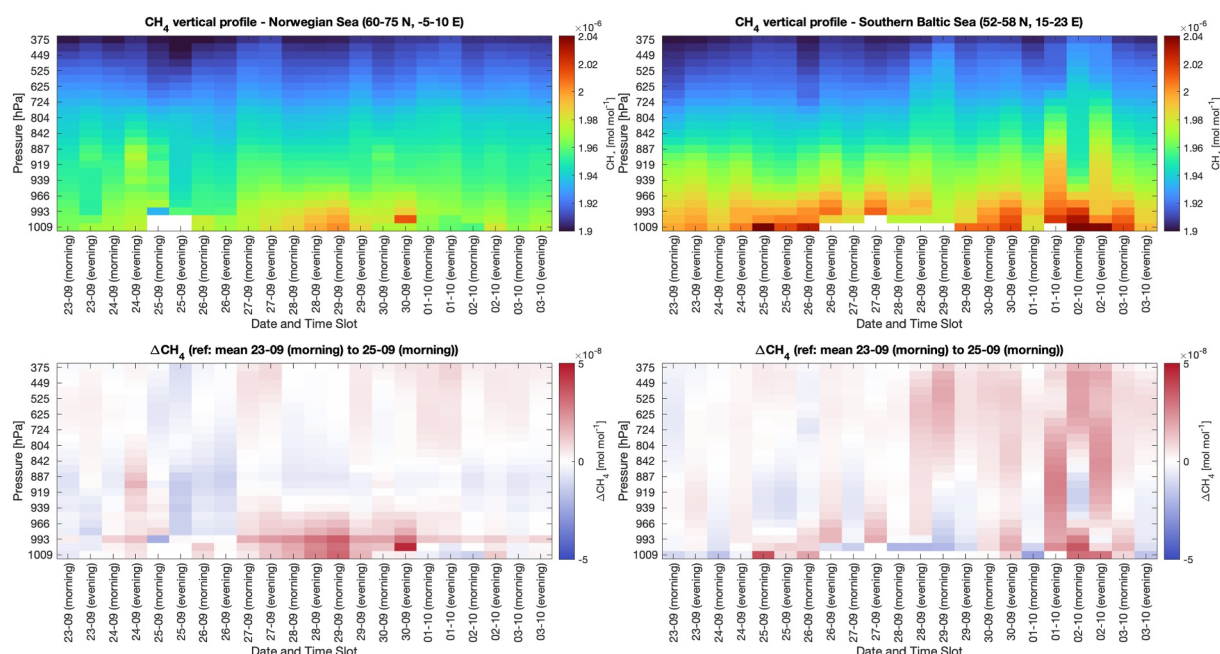


Figure 10. Left: Norwegian Sea (60–75°N, 5°W–10°E). Top: time series of the derived average vertical profiles of CH₄. This representation focuses on the lower troposphere and extends up to about 350 hPa. Data are averaged every 12 hr to reflect the actual temporal sampling of IASI. Bottom: In the same format, we show the time series of the variations of the average vertical profiles of CH₄ derived from this work with respect to the average vertical profile of T1 (from the morning of 23 September to the morning of 25 September). Right: Same as left, but for the southern Baltic Sea (52–58°N, 15–23°E).

reference to be subtracted from the concentration profiles of subsequent days. The result (Figure 10) shows that the changes are more pronounced in the near-surface atmospheric levels. As for the maps, these box-averaged anomalies are below the 50 ppbv threshold for individual soundings, but each of them is an average over a large number of soundings.

As said above, the NN cost function is tailored on the parametric term of a classic OE approach, leaving aside the one associated to observations. Therefore, to draw a definite conclusion about the consistency of this retrieval framework, it is essential to analyze spectral residuals in the range 1,180–1,430 cm^{−1}, interested by the NN retrieval. To this end, we show in Figure 11 the difference between the average residuals (Obs.-Calc.) obtained with an a-priori, uniform profile with a column abundance of 1,880 ppbv, and those obtained with the profiles retrieved from the NN, for the T1 and T3 intervals. Misfits in the P and R branches (1,230 and 1,350 cm^{−1}) are especially evident with the a-priori profiles, and corrected by the retrieval. A slight improvement can be also seen in the Q-branch (1,305 cm^{−1}), which is less evident because of the saturation. Overall residuals clearly improve within IASI noise (black) when using the profiles retrieved by the NN.

4. Conclusions

The work presented demonstrates the potential of a physics-constrained neural network approach for recovering vertical CH₄ profiles from satellite hyperspectral data. The results obtained show how the combination of physical constraints and machine learning techniques allows for robust estimates with uncertainties comparable to traditional approaches, while maintaining high computational efficiency. The analysis of the Nord Stream pipeline sabotage case study also showed the model's ability to capture significant variations in near-surface concentrations and to track the transport of the plume, consistently with independent studies. We note, however, that the metrics reported in Section 3.1 quantify the ability of the scheme to reproduce, through the forward model, the CAMS EGG4 fields used as training target, rather than the accuracy against the real atmosphere: the retrieval has not yet been validated against independent CH₄ measurements, and such a validation is deferred to the forthcoming study based on WRF-CHEM simulations. The models implemented in this phase will be further improved through fine-tuning or re-training of the network based on high-resolution atmospheric simulations, such as those produced by circulation models integrated by the chemical module Weather Research and

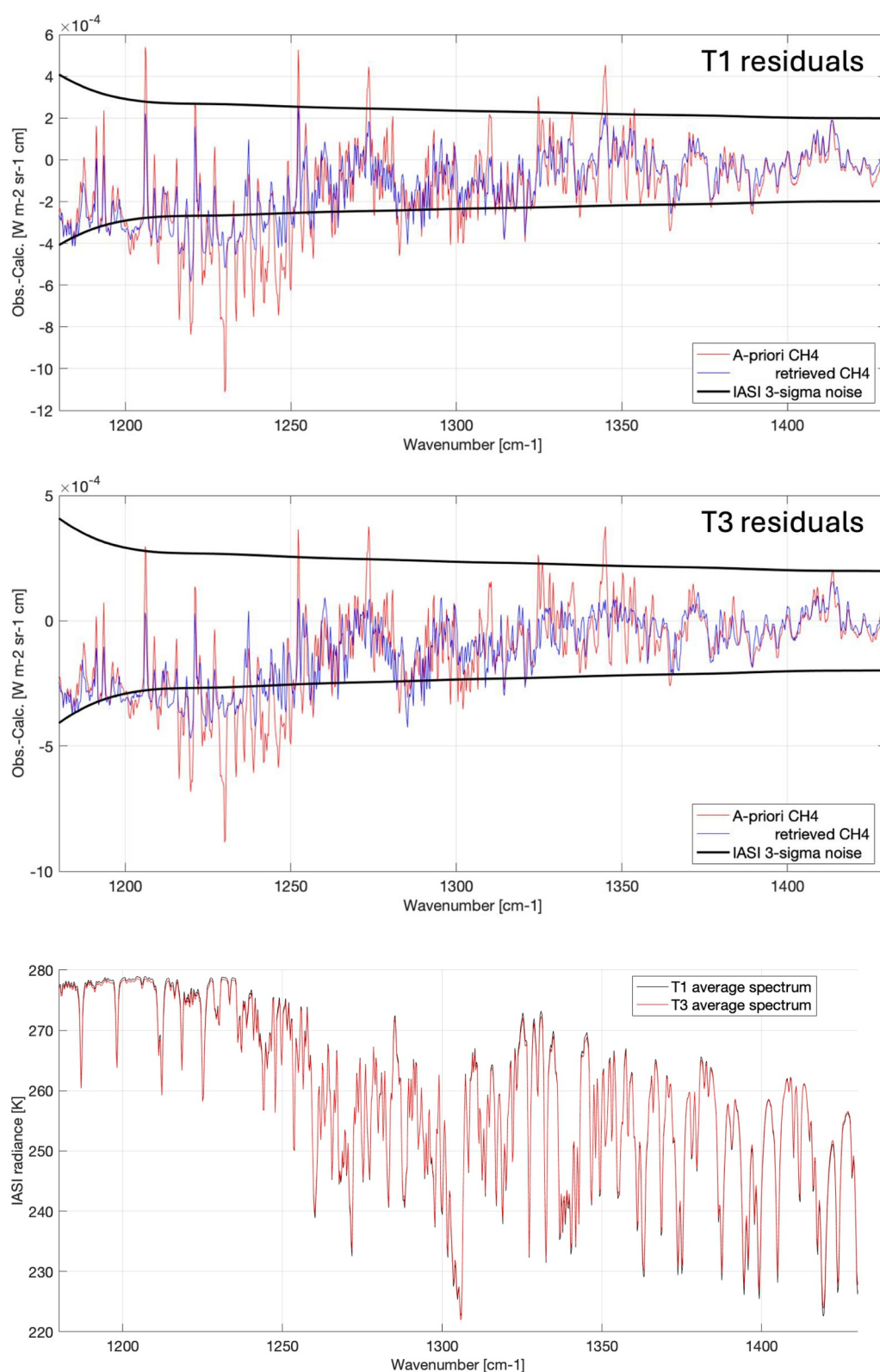


Figure 11. Top: Comparison between average residuals between IASI data and simulated spectra for the T1 interval, using a-priori profiles (red) and network-retrieved profiles (blue), compared with the IASI noise (black, solid). Middle: same as top, but for the T3 interval. Bottom: average IASI spectra for T1 and T3.

Forecasting - Chemistry (WRF-CHEM). This should enhance the model's ability to discriminate weak signals and reduce uncertainties in the lower layers, where spectral sensitivity is more limited. Furthermore, WRF-CHEM will provide profiles at a much higher spatial resolution, which should allow for the detailed capture of emission sources and higher and more realistic concentrations for network training.

Future developments will focus on two main areas: on the one hand, extending the approach to other greenhouse gases of primary importance for climate studies, such as CO₂; on the other hand, applying the method to more complex issues related to atmospheric chemistry in order to monitor gas concentration trends on a regional scale (Giosa et al., 2025). These developments could pave the way for wider use of physics-constrained neural networks in integrated greenhouse gas monitoring, combining satellite observations, numerical models, and in situ measurement networks. The integration of satellite observations with physics-constrained artificial intelligence approaches opens up new perspectives for improving our understanding of the processes that regulate the carbon cycle and the impact of greenhouse gases on the climate. Although still in an exploratory phase, this work clearly highlights how NN schemes integrating OE constraints can become an important alternative to traditional methods of atmospheric monitoring.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Availability Statement

The IASI Level 1C data that have been analyzed in this study are publicly available from the EUMETSAT data store at <https://data.eumetsat.int/data/map/EO:EUM:DAT:METOP:IASI:1C-ALL>. The neural-network models that have been developed in this work are publicly available on Zenodo at <https://zenodo.org/records/23401429> with the data utilized to run the Nord Stream Case Study (Giosa et al., 2026). The model that has been used to produce simulated data to train the network models is σ -IASI/F2N, whose latest version is available at a Zenodo repository (Masiello et al., 2026).

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