

Article

Assimilation of SO₂ TROPOMI Retrievals at the European Scale with EAKF Implemented in MINNI Through DART

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Abstract

Air quality modeling of sulfur dioxide (SO₂) concentrations remains challenging due to the high variability of both natural and anthropogenic emission sources, as well as the complexities associated with its multiphase chemistry. The data assimilation (DA) of satellite observations is a promising technique for constraining model uncertainties by combining the strengths of high-resolution and dense satellite retrievals with physical consistent model outputs. However, existing SO₂ DA applications have primarily focused on volcanic events while this study addresses them together with other emissions. The implementation of an SO₂ DA framework within the MINNI regional chemical transport model using an Ensemble Adjusted Kalman Filter (EAKF) via the DART framework is presented. The performances of the DA assimilation framework were tested using Sentinel-5P/TROPOMI SO₂-COBRA total column retrievals over continental Europe for August 2023. The filter constrained the ensemble variance to capture plumes from power plants and volcanic activity. The ensemble considered 20 members and perturbations of emissions and boundary conditions. On a monthly basis, the mean correction for the total column averaged over the domain was $2 \times 10^{-5} \text{ mol m}^{-2}$, with localized maximum adjustments reaching $3.3 \times 10^{-4} \text{ mol m}^{-2}$. At the surface level, domain-averaged corrections of concentrations reached up to $2.6 \mu\text{g m}^{-3}$. Despite current limitations related to ensemble size, static vertical localization, and the typical temporal fading of initial condition corrections, validation against in situ data confirmed the system's ability to transfer column information to near-surface levels. These results demonstrate the feasibility and added value of integrating mixed-source SO₂ satellite retrievals into regional air quality simulations, contributing to more accurate, observation-driven atmospheric monitoring.

Keywords: air quality modeling; satellite data assimilation; ensemble Kalman filter; sulphur dioxide; Sentinel-5P; TROPOMI



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1. Introduction

Air quality has improved over decades due to emission reduction policies; however, “Millions of years of healthy life are still lost each year due to pollution and at least 10% of

premature deaths in Europe are driven by exposure to polluted air, water and soil, noise and harmful chemicals” [1]. This assessment is based on current EU standards of the Ambient Air Quality Directive [2], which are less stringent than those established by the new Directive [3], leading thus to an under-estimation of pollution impacts on public health.

In addition to implementing further long-term emission reduction strategies, air quality managers can use air pollution forecasts to protect human health during high pollution episodes, allowing individuals and governments to take proactive measures to reduce exposure. The air quality modeling systems used for forecasting are continuously improved, with one key approach being the assimilation of measurements/observations. In particular, the assimilation of satellite retrievals of atmospheric constituents is an ongoing process as new satellite data becomes continuously available. This study makes use of data provided by the European Commission’s Sentinel-5P (S5P) satellite, TROPOMI instrument, that is part of the Copernicus earth observations (EO) program and delivers real-time data of mid-day concentrations for several atmospheric constituents at unprecedented resolutions ($5.5 \text{ km} \times 3.5 \text{ km}$) every day since April 2018 over Europe [4–6]. The atmospheric simulations combined with satellite observations through data assimilation (DA) techniques [7–9] improve the forecast of air parameters. Since the 1990s, many applications have shown the advantages of using data assimilation for chemical constituents of the atmosphere to improve both short-term air quality forecasting and long-term emission inventory accuracy, as well as to produce the reanalysis of past atmospheric composition [10–14].

Ongoing challenges in air quality modeling are mostly due to the complexity and lack of knowledge of the governing processes such as the strong coupling across scales, the large uncertainties in the pollution emission inventories and of physical and chemical interactions between pollutants, between atmosphere and surface, etc. [15]. The DA of satellite observations for pollutants has assisted in reducing simulations’ biases, and it has steadily transitioned from case studies applications to operational systems [16–19].

Among all pollutants, assimilating sulphur dioxide (SO_2) data is particularly important as it is emitted by industrial and volcanoes point sources that have high uncertainties in emission inventories and have significant health impacts from its rapid conversion to sulphate aerosols, which were proven to have direct adverse effects on respiratory, cardiovascular, and nervous systems [20–22]. In addition to these short-term air pollution effects, sulphate aerosols have impacts on long-term climate forcing by modifying directly and indirectly, through clouds, the radiative balance of Earth [23,24]. Moreover, on plants, the presence of SO_2 in the air inhibits photosynthesis and reduces crop yield [25], as well as it is a major contributor to acid rain, which damages forests, depletes soil nutrients and acidifies water bodies, and causes harm to both aquatic and terrestrial ecosystems [26].

However, to date, few applications of satellite SO_2 data assimilation are documented. Flemming & Inness [17] is the reference paper of the SO_2 DA performed by ECMWF/CAMS, which is a unique case of a global Near Real Time (NRT) forecasting and reanalysis system that assimilates SO_2 retrievals (GOME-2 [27] and TROPOMI) operationally. Bacles et al. [19] describes the SO_2 DA performed by Météo-France at regional scale for the MOCAGE system, where IASI [28] and TROPOMI SO_2 retrievals are routinely ingested for volcanic plume forecasts to support the Toulouse’s volcanic ash advisory center (VAAC).

Both studies employ variational assimilation methods, 4D-Var [29] and 3D-Var [8], respectively, and they focus exclusively on volcanic SO_2 , to improve NRT forecasts of volcanic plumes and better reproduce their effects on both weather and climate. However, they do not address retrievals from other input sources and, to date, no other study has reported the application of data assimilation to both background and volcanic SO_2 . Since SO_2 has weaker UV absorption features, compared to molecules such as O_3 [30], and not-volcanic SO_2 has low atmospheric concentrations, satellite SO_2 retrievals for background conditions

exhibit a low signal-to-noise ratio, making its assimilation complicated. In addition, since SO₂ sources (e.g., volcanic eruptions, power plants, smelters) emit at different altitudes and produce plumes with variable heights, errors in the assumed emission profiles can lead to substantial biases in the retrieved SO₂ columns [31,32] that risk to deteriorate the model first-guess if assimilated. Notwithstanding this complexity, this study demonstrates the potential and illustrates limitations of assimilating satellite SO₂ observations from various sources to meaningfully inform model predictions.

In this work, TROPOMI/S5P SO₂-COBRA Vertical Column Densities (VCDs) for August 2023 are assimilated in the MINNI air quality modeling system [33] over continental Europe. The DA approach used is the Ensemble Adjusted Kalman Filter (EAKF), assisted by NCAR's Data Assimilation Research Testbed tool (DART; [34–36]). This period includes eruptions that caused the closure of the Catania airport (Italy), that stands about 50 km south of the Etna volcano—on August 13, 14 and 15.

The article is organized as follows: Sections 2.1 and 2.2 describe the satellite data product and the air quality modeling system, respectively. The implementation of the assimilation system and its setup are described in Section 2.3. The results are presented in Section 3, whereas Sections 4 and 5 provide discussion and conclusions, respectively.

2. Materials and Methods

2.1. Sentinel-5P/TROPOMI SO₂-COBRA

The Sentinel-5P/TROPOMI COBRA Sulphur Dioxide [L2__SO2CBR_] total vertical column densities reprocessed (RPRO) products are assimilated (collection 03, version 2.00.01, [37,38]). The Covariance-Based Retrieval Algorithm (COBRA) SO₂ dataset used in this study was chosen as it delivers SO₂ columns with markedly lower noise and bias than the previous operational algorithm [39]. The Sentinel-5P satellite-carrying TROPOMI instrument has a sun-synchronous orbit, passing over the same spot on Earth at approximately the same local solar time each day. This, together with its relatively high spatial resolution (3.5 km × 5.5 km), allows for consistent daily SO₂ atmospheric data that distinguishes small-scale plumes of both volcanic and human-caused emissions previously undetectable from space. For the remainder of this study, the terms 'column', 'total column', and 'vertical column density' (VCD) are used interchangeably to refer to the column-integrated SO₂ concentration.

TROPOMI Sulphur Dioxide vertical columns, measured in the ultraviolet part of the electromagnetic spectrum, are calculated from the satellite slant columns using an a priori vertical profile [40]. Given the unpredictability of volcanic eruptions, four different data products are routinely delivered from a single slant column observation, using four different a priori vertical profiles. The data product used for this study is computed using the vertical profiles of the Chemical Transport Model (CTM) TM5 [41]. The TM5 simulation includes anthropogenic and biogenic SO₂ emissions [42] and consequently the vertical profiles reflect this diversity. Conversely, the three other products delivered by the Copernicus Operational center cover possible volcanic scenarios. Each uses a hypothetical SO₂ volcanic profile of plumes located either in the boundary layer (~1 km), in the mid-troposphere (~7 km), or in the stratosphere (~15 km). Both Flemming & Inness [17] and Bacles et al. [19] have used the mid-troposphere product for occasions in which a volcanic eruption occurred. As stated in the introduction, to date no other publications have addressed within a DA framework the SO₂ data retrieved with the general TM5 a priori vertical profile.

To remove cloud-covered scenes, partially snow/ice-covered areas, errors, and problematic retrievals, only data with quality flags above 0.75 are used. Moreover, very-low and noisy VCDs, under a threshold of $8.3 \times 10^{-6} \text{ mol m}^{-2}$, were removed, thereby excluding observations subject to greater measurement uncertainty, consistently with the filtering

approaches adopted for other TROPOMI species, such as NO₂, in previous studies [43]. The error estimates given by the data provider were used as observation errors. As shown in Section S1 (Equation (S5)), they are key components in the analysis to estimate computation, as it states the uncertainties behind the data. In this study, no bias correction nor perturbation were applied to the observations, following the high-quality flags considered. Data availability over Europe varied significantly throughout the day, with consistently sparse observations between 6:00 and 09:00 UTC. Consequently, these orbits were excluded from the assimilation process to lower the computational burden.

2.2. MINNI

MINNI air quality modeling system [33] is based on FARM [44–46], a 3D offline Eulerian CTM, that accounts for the transport (advection and turbulent diffusion), chemistry (gas phase, with flexible mechanism configuration, and aerosol module) and dry/wet removal of atmospheric pollutants. Gas phase chemistry is simulated using the SAPRC-99 [47] mechanism, while aerosol dynamics is modeled by means of AERO3 module [48,49]. Secondary inorganic and organic aerosol production is parameterized by ISORROPIA [50,51] and SORGAM [52], respectively. Wet removal processes are implemented following the EMEP approach [53], whereas dry deposition follows the resistance model of [54]. Biogenic emissions are produced using the MEGAN v.2.04 model [55], sea salt emissions are parameterized following [56] and dust resuspension and erosion are modeled as in [57].

The MINNI setup is the same used within the CAMS Regional Ensemble Forecast [33]. Meteorological data were provided by the ECMWF Integrated Forecasting System (IFS) with 10 km horizontal resolution over the European domain, using 17 terrain following vertical levels from 20 m to 11,790 m above the ground. The anthropogenic emissions were based on CAMS-REG_v.6.1.1 emission inventory for the year 2022 with time profiles TEMPO_v3.2 and chemical boundary conditions were provided by the ECMWF global C-IFS model reanalysis. This simulation, without assimilation, is hereafter referred to as the basecase.

2.3. DART

The Data Assimilation Research Testbed (DART; [34,36]) has been chosen to implement an assimilation algorithm in the MINNI air quality modeling system since it is an open-source system developed to enable ensemble-based data assimilation across various earth system numerical models. It implements several assimilation techniques; Ensemble Kalman Filter (EnKF; [58]) is one of them and it consists of a sequential technique that updates model forecasts with observations to obtain an improved estimate of the system state and the analysis. In each assimilation cycle, the analysis is then used to restart the model before propagating the simulation forward in time. Unlike variational approaches (3D-VAR [8], 4D-VAR [29], the EnKF method avoids the need for adjoint models and instead represents forecast uncertainty through an ensemble of simulations [59]. Each member of the ensemble evolves independently under model dynamics with stochastic perturbations that represent model errors. The ensemble statistics provide a flow-dependent approximation of the forecast error covariance, allowing the analysis to converge toward the optimal Kalman solution as the ensemble size increases [60].

The DART version implemented in MINNI uses the Ensemble Adjustment Kalman Filter (EAKF; [61]), a deterministic (no perturbation on observations), square-root ensemble filter formulation that preserves both the analysis mean and variance according to Kalman filter theory. To counteract sampling errors arising from small ensemble sizes, DART applies covariance localization and inflation techniques, which stabilize the filter and prevent the collapse of the ensemble spread. Details of the EAKF implementation, following Barré

et al. [62], are provided in the Supplementary Materials (Section S1) along with localization and inflation definitions.

MINNI-DART Coupling Settings

Total column retrievals TROPOMI/S5P SO₂-COBRA were used without modifying the Air Mass Factors (AMFs) and without applying superobbing-to-model-grid strategies. Superobbing, i.e., averaging multiple nearby observations into a single representative value, may lead to a loss of fine-scale features and an under-estimation of variance, thereby further affecting the representativeness error. Due to computational constraints, in CTM applications, the ensemble is typically constrained to 15–30 members [63]. In this study, 20 CTM simulations were run in parallel. Each hour of simulation uses initial conditions (ICs) from the previous hour. The hourly ICs are updated through assimilation whenever the acquisition time of satellite observation is half an hour before or half an hour after. The time at the beginning of the overpass is used to identify all pixels acquired within that orbit. DA is performed for all ensemble members, and the best guess of the true variable state is assumed to be the analysis ensemble mean. Neither the underlying emissions nor other variables or parameters are targeted in the assimilation, only the total column. The analysis is univariate, assuming that there are no cross-correlations between SO₂ background errors and other chemical species. In DART, the forward operator is user-defined, and it was built referring to the observation operator routines provided by the Copernicus Satellite Operator (from TNO, The Hague, Netherlands; <https://ci.tno.nl/gitlab/cams/cso>, last accessed on 16 May 2025), following DART's templates. Table 1 provides a summary of the MINNI-DART parameters used in this study. The following paragraph presents DART parameters along with the rationale for their selection. Details on the ensemble construction methodology can be found in Supplementary Materials in Section S2, whereas Section S4 provides computational details and timing information to document the implementation and assess the computational cost of the approach.

Table 1. An overview of the MINNI-DART implementation.

Parameter	Description
Model	AMS-MINNI with FARM CTM
Vertical levels	17 levels: 20–11,790 m
Horizontal resolution	0.10° × 0.15°
Observations assimilated	Level-2 TROPOMI/S5P SO ₂ -COBRA vertical column densities (/PRODUCT/sulfurdioxide_total_vertical_column[])
Assimilation period	August 2023
Assimilation tool	DART
Filter method	Ensemble Adjustment Kalman Filter (EAKF)
Ensemble size	20 Members
Outlier threshold	40 standard deviations
Ensemble spread management	Restarted prior inflation
Inflation settings for each restart (following DART's documentation [64])	$\mu = 1.05$ (adim) $\sigma = 0.6$ (adim)
Localization Function	Gaspari–Cohn
Cutoff Distance/Horizontal Influence	0.01 radiant
Vertical Influence	None
Localization threshold	20 observations
Cutoff floor	0.001 radiant

Since the size of the ensemble is minimum, an adaptive spatially and temporally varying inflation is applied to the prior state [65]. To avoid filter divergence caused by uncontrolled inflation, the inflation process is restarted after each assimilation cycle. Each inflated state is built from a common state, independent of the inflation fields from the previous time-step. A probability density function governs the initial state characterization; its mean and standard deviation were defined following DART's suggested option.

Limiting the impact of an observation on a state variable is called localization or tapering. The Gaspari–Cohn (GC; [66]) function, largely used in data assimilation algorithms, was chosen for this purpose. This function effectively limits the analysis (state) increments within a local volume around the location of the observation. In this work, the localization is used only on the horizontal, whereas on the vertical no-dampening effect is introduced. The vertical characterizations are inherited from the background error covariance matrix. The vertical localization is turned off to prevent misplacing corrections, because the assimilation includes mixed-type satellite SO₂ observations and the current localization scheme cannot handle this diversity. This decision is based on the need to avoid tailoring the implementation to a single emission source, as doing so could worsen the assimilation performance for the others. In the Supplementary Materials (Section S3.3), a run characterized by a vertical localization suited for volcanic eruptions (Figures S3 and S5) is shown, along with an equivalent run without vertical localization (Figures S2 and S4). When activated, corrections are applied in the mid-troposphere, which is appropriate for volcanic SO₂, but unrealistic for SO₂ plumes in regions dominated by strong anthropogenic emissions. The GC function has a Gaussian shape with values starting from one (over the observation location) and tapering off isotropically to zero with increasing distance from the observation location. The half-width parameter, the cutoff, defines the localization area; the vertical localization is excluded by setting a very high cutoff for the vertical axis.

An adaptive localization threshold of 20 observations is used to avoid filter divergence without discharging precious information. In fact, the filter may return unrealistic analysis when within the localization influence area too many observations are present [67]. This behavior is exacerbated by the sampling noise caused by insufficient ensemble size. To control this behavior, the cutoff and the number of off-diagonal terms in the model error covariance matrix are reduced to match the adaptive localization threshold. The cutoff floor defines the lower bound below which the cutoff distance cannot be reduced. The chosen adaptive localization threshold resulted in 0.012% of pixels being discarded after entering DART.

When the mean of the ensemble members is too far from the observation value, the assimilation of the observation might destabilize the model run. The outlier rejection threshold is introduced to avoid disruptive increments. However, since the emissions dataset does not include volcanic point sources, the model states and satellite retrievals do largely differ in the case of an eruption. Therefore, with respect DART's proposed setting, the outlier rejection threshold is relaxed to 40 standard deviations so that high SO₂ concentrations are not discarded.

To evaluate the background errors, a 20-member ensemble is generated following the perturbation methodology outlined in the Supplementary Materials (Section S2). The method was chosen because it preserves the underlying spatio-temporal structure of the atmospheric system and ensures a realistic variability in the generated perturbations. The background ensemble is created by perturbing both emissions and boundary conditions (BCs). On average, the ensemble returns the basecase emissions and BCs. Driven by a variety of emissions and BCs, each run contributes to sample the full error spectra towards a model error covariance matrix as realistic as possible.

For the present application, the perturbation parameters were chosen following preliminary sensitivity tests. The same values in percentage were used for perturbing both the

emission fluxes and the BCs. The perturbation bounds are currently defined heuristically to ensure numerical stability and a realistic system. Ideally, they should be grounded in the known uncertainties of the input parameters (e.g., emission inventories, boundary conditions), so that the ensemble's spread reflects physically meaningful error estimates rather than arbitrary numerical constraints. In this case, the calculated perturbations exceed the uncertainty rates typically reported in the literature for the chosen emission inventories and boundary conditions datasets [68,69]. While exceeding inventory-specific bounds, this inflation was adopted to induce sufficient ensemble spread and account for broader, unrepresented model errors. Results from these sensitivity analyses (not exhaustive) are reported in the Supplementary Materials (Section S3.4, Figures S6–S8) during the volcanic eruption. They show how different parameters' choices result in higher or lower assimilation. Given the sensitivity assessment, a correlation time τ of 24 h was chosen to account for the relatively long atmospheric lifetime of SO_2 before oxidation or deposition occurs [70]. Whereas a horizontal L_{hz} and a vertical L_{vz} correlation lengths of 200 km and 0.5 km were used, respectively, allowing emissions to be well-mixed over a significant spatial range before deposition [71,72]. Lastly, a spread σ_o of 16 was chosen, as lower values resulted in insufficient dispersion over the domain, limiting the effectiveness of corrections (not fully shown in Section S3.4). The parameters symbols refer to the mathematical description presented in Section S2.

Figure 1 illustrates the monthly averages of the ensemble mean and standard deviation for the perturbed input fields. The top row displays the statistics for the SO_2 emission ensemble at an altitude of 20 m. The mean field clearly highlights major shipping routes and localized point sources, including harbors, urban areas, and industrial hotspots, which constitute the primary sources of SO_2 within the domain. The corresponding spread map (top right) identifies the regions where the perturbation magnitude is largest. These high-variance areas, indicated in red, can be directly interpreted as the locations where the uncertainties in the emission inventories are most significant. By concentrating the spread in these areas, the ensemble effectively accounts for the lower confidence levels associated with localized and high-intensity emission sources. The bottom row of Figure 1 presents the ensemble statistics for the top boundary conditions (BCs). While the lateral BCs were also perturbed as part of the ensemble generation, only the top-level ensemble is visualized here to illustrate the uncertainty assigned to the vertical influx of SO_2 from the upper atmosphere.

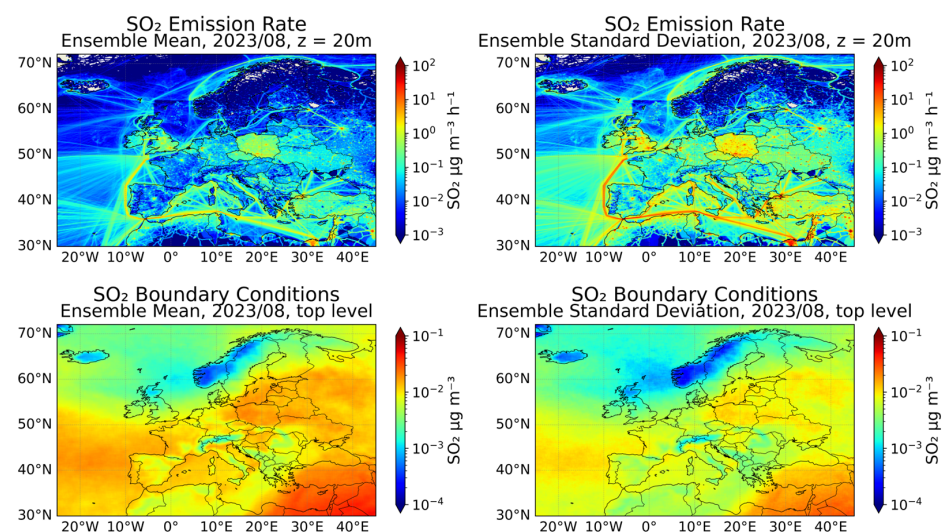


Figure 1. Ensemble mean (left) and spread (right) of perturbed SO_2 emission rates at 20 m (top) and of perturbed SO_2 concentrations at the top boundary level (bottom) for August 2023. Lateral BCs, also perturbed, are not shown.

3. Results

This section shows an extensive valuation of MINNI-DART implementation for August 2023 using TROPOMI/S5P SO₂-COBRA vertical column densities (VCDs). The results cover the assimilation cycle (Section S1), as well as the overall model performance in the period investigated. Firstly, the analysis focuses on the 2D observational space (Section 3.1), where the assimilation occurs, and compares the model states and the observations. To gain better insights into the effects of assimilating satellite observations, a focus on two subdomains characterized by different emission sources (Section 3.1.2) is presented in addition to the whole domain (Sections 3.1.1, 3.2 and 3.3). The analysis also investigates how the proposed assimilation influences the volumetric distribution of SO₂ across the atmospheric layers by showing SO₂ concentrations in the three-dimensional model space (Section 3.2). Finally, Section 3.3 shows MINNI-DART performances against independent ground-based measurements over the simulated domain.

3.1. Evaluation in 2D Observational Space

3.1.1. Full European Domain

The observed VCDs are compared to the modeled ones derived from the background (forecast) and the analysis fields (Figures 2 and 3). Since the assimilation in DART is performed on a satellite pixel basis as available in the TROPOMI Level 2 product and given that the pixels are irregularly spaced and vary between orbits, the comparison with modeled monthly means were carried out on the regular model grid, defined in Table 1. A first-order conservative gridding approach that preserves the area-integrated value, usually used for atmospheric modeling applications, was used to remap observed VCD. This remapping is applied only for the analysis of the monthly mean fields and does not affect the observation set assimilated by DART. Keeping the individual TROPOMI pixels preserves the spatial information and observation density of the original retrievals, while potentially introducing representativeness errors [73] due to the difference between the TROPOMI and MINNI spatial resolutions.

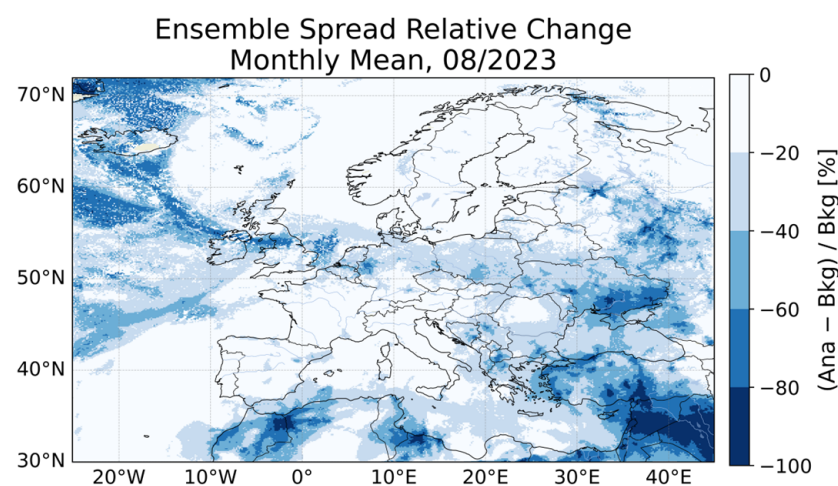


Figure 2. Percentage differences in the monthly mean ensemble spread between the analysis and the background fields with respect to background fields. Negative values indicate a reduction in spread following the assimilation of TROPOMI SO₂ observations. As expected, no positive values were registered.

Before comparing model VCDs to observations, it is necessary to evaluate the relative change in ensemble spread as a diagnostic of the filter performance. A fundamental consistency check for any ensemble-based assimilation system is the reduction in spread

from the background (Bkg) to the analysis (Ana). A properly functioning filter should yield a contraction of the ensemble's variance after the assimilation, as a negative change indicates that the analysis ensemble is more constrained—and thus less uncertain—than the background ensemble [74]. Figure 2 illustrates this relative change, expressed as a percentage. It shows uniformly negative values across the entire domain. This spatially coherent contraction of the spread demonstrates that the filter operated as expected, effectively absorbing the information from TROPOMI/S5P SO₂ observations and significantly constraining the model's uncertainty.

Monthly Mean Total Column SO₂, 08/2023, MINNI-DART

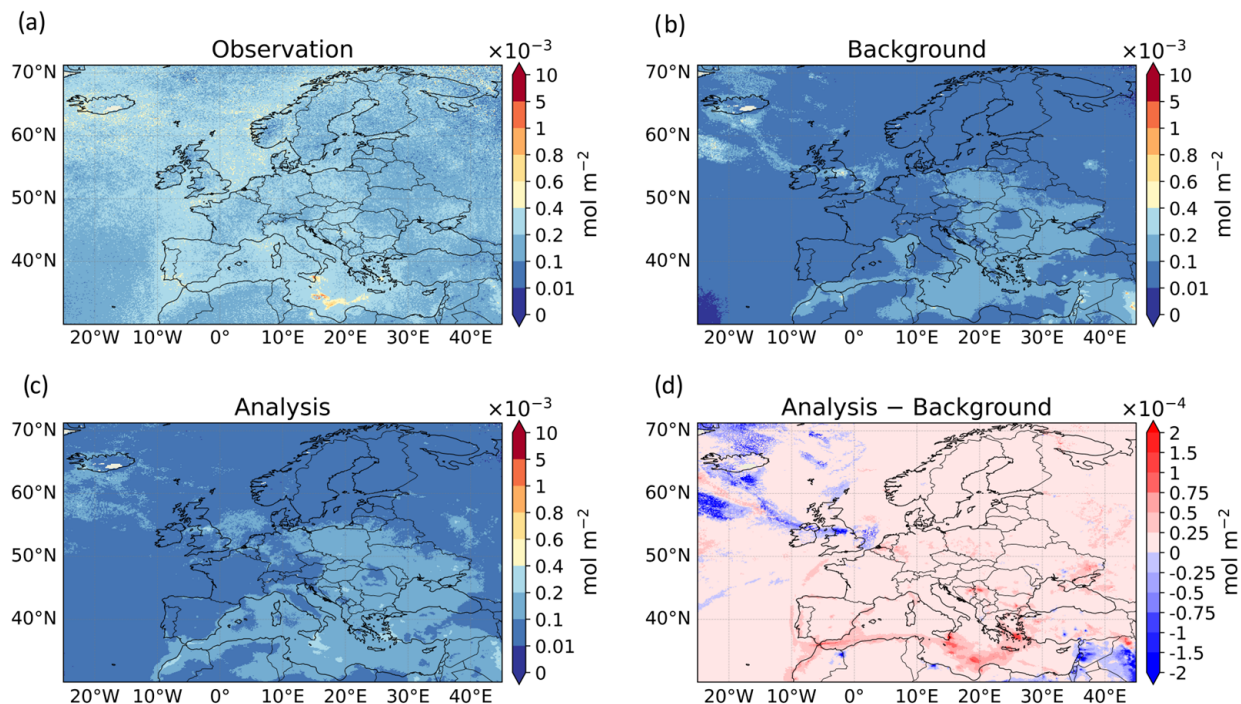


Figure 3. Monthly mean SO₂ vertical column densities on August 2023 as (a) retrieved by TROPOMI/S5P (observation), (b) simulated by MINNI before assimilation (background), (c) simulated by MINNI after the assimilation (analysis) and (d) the difference between the analysis and the background.

The impact of data assimilation on the SO₂ total column densities is illustrated in Figure 3, which presents the monthly mean fields for August 2023. Averages are computed on assimilation time. The comparison includes the TROPOMI observations (a), the model background (b), the resulting analysis (c), and the spatial distribution of the assimilation corrections (analysis—background, d). These maps allow the identification of structural model–observation discrepancies (Figure 3a,b) and pinpoint the geographical areas where the assimilation process introduced the largest corrections (Figure 3a,c,d).

Figure 3 shows that the MINNI background simulations (Figure 3b) are lower with respect to TROPOMI observations (Figure 3a), which is a finding further upheld by the domain-averaged time series presented in Figure 4. Assimilation reduces this gap, moving the analysis closer to observed values steadily throughout the month. Figure 4 shows how the correction on average over the full domain is constant in time, with very few day-specific trends (i.e., 19, 29 August): changes in the observed concentrations do not always correlate with changes in the domain-average shift introduced by the assimilation. While the domain-average correction of the ensemble mean is approximately $2 \times 10^{-5} \text{ mol m}^{-2}$ (Figure 4, red minus blue line) each day, localized monthly positive increments exceed

$2 \times 10^{-4} \text{ mol m}^{-2}$ on average in areas where local SO_2 sources are not well captured in the emission inventory (Figure 3d). This moderate behavior reflects/hints on the conservative tuning of the assimilation filter, which is designed to balance prior model information with observational uncertainty. By avoiding over-fitting to transient satellite noise, the system ensures that the analysis remains physically consistent while still effectively correcting broad-scale systematic errors.

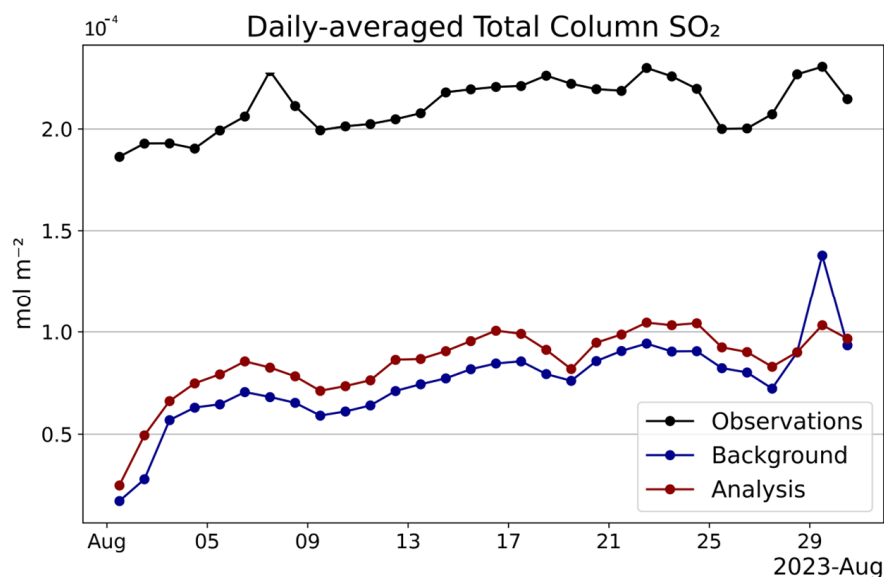


Figure 4. Daily averages of SO_2 total columns over the entire MINNI domain for Observations (TROPOMI/S5P, black), Background (simulated by MINNI before assimilation, blue) and Analysis (simulated by MINNI after the assimilation, red).

Although the assimilation generally leads to an increase in simulated VCDs, the corrections are not always positive. Significant negative corrections are also observed over the Atlantic Ocean on the Northwest corner of the domain and over parts of the Middle East, including Saudi Arabia and Iraq (blue areas in Figure 3d). In these regions, the background likely overestimates emission fluxes and/or BCs. Since these high concentrations are not confirmed by the satellite overpasses, the DA reduces the excess SO_2 before advancing the simulation in time.

The positive increments (red areas in Figure 3d) are concentrated along major Mediterranean shipping corridors and around Mount Etna (reflecting detected volcanic activities). Localized VCDs increase areas are also evident in the Balkans (Southeastern Europe), such as near Šabac (Serbia), suggesting a systematic under-estimation of the anthropogenic SO_2 emissions employed. Positive increments are also observed in Greece, where severe wildfires occurred in August 2023 [75]. Similar wildfire events in Greece have previously been associated with enhanced SO_2 concentrations, as documented during the August 2021 wildfires [76].

The capability of the DA system to compensate for unknown or underestimated sources, particularly for episodic events like volcanic eruptions, is examined in further detail in the following subsections (Section 3.1.2).

The frequency distribution of SO_2 VCD values for August 2023 over the MINNI domain is presented in Figure 5. The histograms represent the relative frequency (%) of the TROPOMI-retrieved observations (gray), the background (blue), and the analysis (red) fields across logarithmically spaced bins. The observed SO_2 columns display a broader distribution with higher median compared to the model fields (that is the model negative bias already introduced above) with a maximum frequency around $1 \times 10^{-4} \text{ mol m}^{-2}$.

Following the assimilation, the analysis exhibits a consistent rightward shift relative to the background ($\sim 25\text{--}30\%$ higher median), indicating a slight adjustment particularly within the 5×10^{-5} and the $8 \times 10^{-5} \text{ mol m}^{-2}$ range. However, this shift does not lead to a full alignment of the modeled and observed distributions; specifically, for VCDs exceeding $4 \times 10^{-4} \text{ mol m}^{-2}$, the frequency of the analysis values remains significantly lower than the observations. This persistent discrepancy in the high-value tail suggests that the DA system applies a cautious correction maintaining the overall features of the simulation. This ensures the mitigation of systematic biases while preventing over-fitting to high-intensity satellite peaks, which may be influenced by transient noise or representativeness errors. However, the ability of the system to deal with localized extremes, either from unknown or underestimated sources, is presented in the following section.

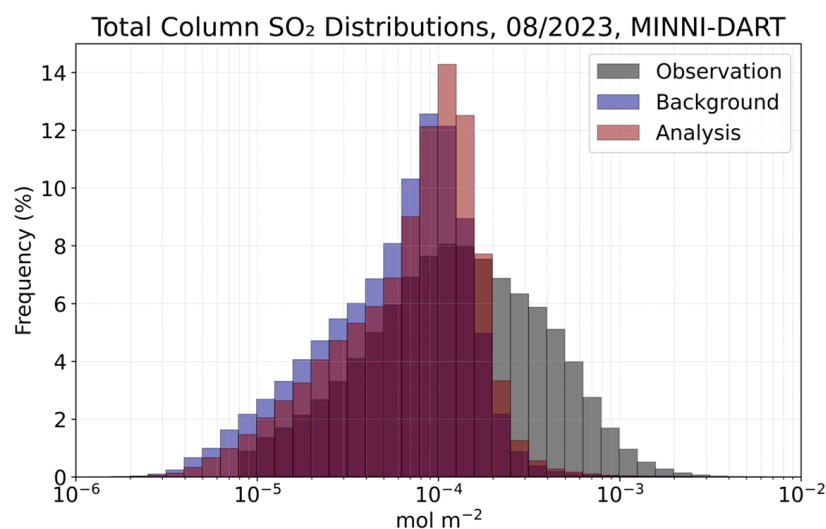


Figure 5. Distribution of SO₂ vertical column densities for August 2023 over the whole spatial domain for Observations (TROPOMI/S5P, black), Background (simulated by MINNI before assimilation, blue) and Analysis (simulated by MINNI after the assimilation, red).

3.1.2. Subdomains

The two areas investigated here, Mount Etna and Šabac, were selected because they are among the regions most influenced by the assimilation (Figure 3d) and they are characterized by distinct emission sources. The detailed geographical location and boundaries of the two subdomains are illustrated in Figure 6.

Figure 7 shows the monthly SO₂ emission rates computed as described in Section 2.2 over the two subdomains. The first subdomain includes major natural SO₂ emissions originating from Mount Etna's volcanic activity, covering the central Mediterranean Sea, off the coasts of Sicily (Italy), Malta, Tunisia and Libya. In addition to standard outgassing activity, between 13 and 15 August, an eruptive event caused the closure of the Catania airport (Italy), that is located about 50 km south of the Etna volcano. However, as volcanic eruptions are absent from the emission inventory used, the monthly SO₂ emission rates shown in Figure 7 highlight only anthropogenic SO₂ sources, primarily related to major shipping routes and harbor activities. This subdomain was chosen to examine in detail the effect of assimilating volcanic plumes encoded in the data product that used the TM5 a priori vertical profile in the framework of a broader, not source-related assimilation, and in cases where no prior volcanic SO₂ emissions are included in the model.

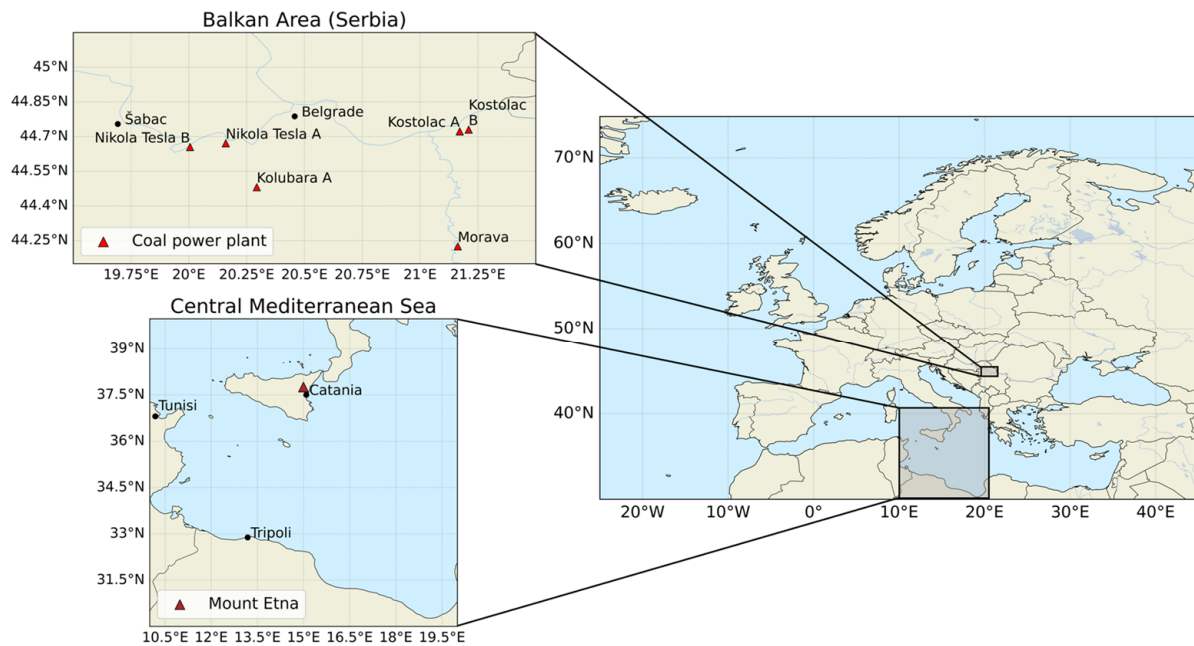


Figure 6. Geographical location of the two subdomains analyzed in Section 3.1.2.

SO₂ Emission Rate, Monthly Mean, $z = 20$ m, 08/2023

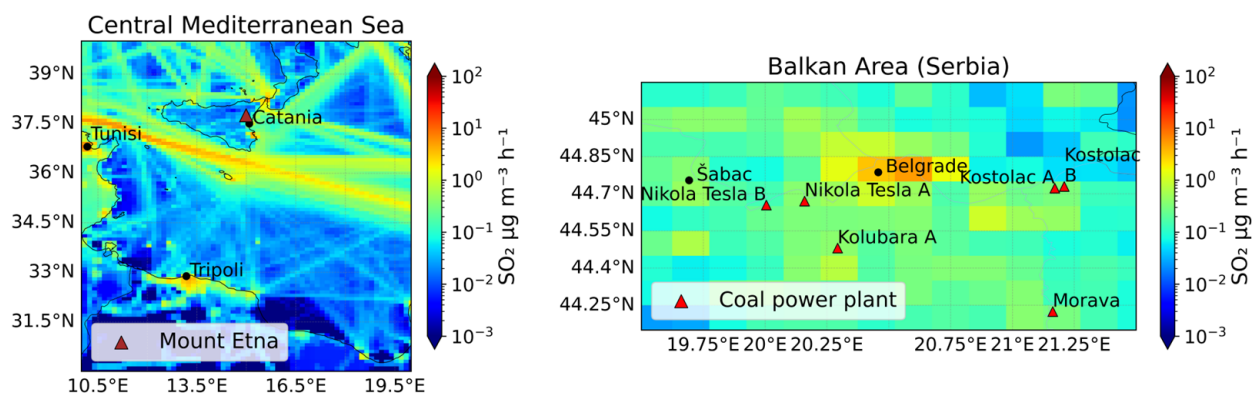


Figure 7. Monthly average SO₂ emission rates at ground level of the south-central Mediterranean Sea (left), and of an area in the Balkan region high in SO₂ concentrations and coal power plant density (right).

The second subdomain targets a region with intensive anthropogenic SO₂ emissions in the Balkan region of Eastern Europe. This area includes six of Serbia's eight largest coal-fired power plants (PPs) in terms of power generation capacity. The monthly mean SO₂ emission rates for this subdomain are presented in Figure 7. Table S1 lists the power plants in order of nominal intensity. In 2024, Serbian PPs emitted 4.6 times more SO₂ than allowed under the EU's National Emissions Reduction Plan, despite these being the lowest values since the Large Combustion Plants (LCP) Directive entered into force in 2001 [77]. Satellite data assimilation can help in evaluating the accuracy of such reported data by comparing satellite observations with model equivalents from simulations informed by the reported data. This region was selected among several areas with high concentrations of large coal PPs because the corrections introduced by the assimilation are significant. Please note that an inverse modeling framework is required to infer the correct emission fluxes, which exceeds the scope of this study.

Similarly to Figure 3, Figure 8 shows the monthly mean SO₂ vertical column densities as observed by TROPOMI (top) and simulated by MINNI before (middle) and after (bottom)

the assimilation process. Here, SO₂ plumes that were either misrepresented (Šabac, right column) or entirely absent (Mount Etna, left column) in the background simulation are successfully reconstructed or adjusted by the assimilation.

Monthly Mean Total Column SO₂, 08/2023, MINNI-DART

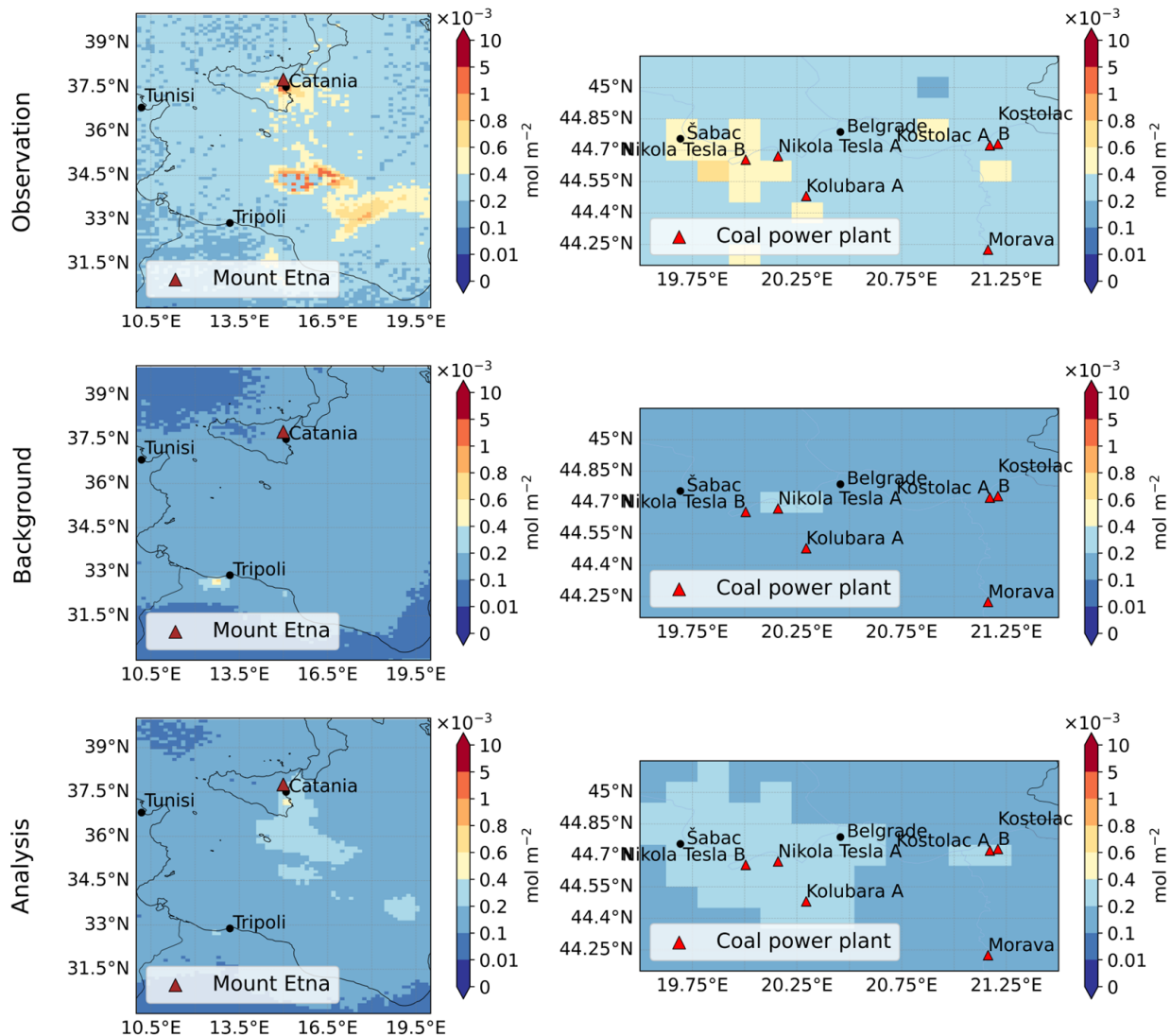


Figure 8. Monthly averages of SO₂ satellite-retrieved observation (**top**), background (**middle**) and analysis (**bottom**) fields, for the two subdomains: the south-central Mediterranean Sea (**left**) and the Belgrade (Serbia) area in the Balkan region of Eastern Europe (**right**).

In the central Mediterranean subdomain (Figure 8 left), the filter effectively increases the near-zero SO₂ concentrations over the open sea, recovering from the satellite observations of the volcanic plume emitted from Mount Etna. A localized peak is observed directly south of Catania; this feature is particularly instructive regarding the filter's performance. Since volcanic emissions were not included as point sources and therefore were not perturbed, the background ensemble variance in this region was initially low. When the ensemble spread is too low compared to the innovation (i.e., the discrepancy between the ensemble means and the actual observation) size, as it was in this case, the ensemble is over-confident, and it tends to ignore observations. In this context, covariance inflation plays a vital role, ensuring that the filter remains responsive to observational updates.

However, this was not the only mechanism at play. Due to simultaneous ensemble perturbations on ship emissions, harbor activities, and boundary conditions, the local ensemble covariance was effectively increased. This concurring covariance likely facilitated corrections in this region, as the Kalman Filter applies the largest increments where the background errors—represented by the ensemble spread—are the largest. Specifically, south of Catania, the high density of anthropogenic point sources (as shown in Figure 7, left) coupled with consistent wind directions ensured a region of high ensemble variance and satellite-observed plumes. Consequently, the assimilation was able to leverage this existing error structure to map the volcanic innovations onto the model state. This process may have occurred elsewhere along the plume-impacted area due to overlapping anthropogenic sources. However, such coincidences are not guaranteed; in their absence, the assimilation might have been less responsive to the eruption, underscoring the importance of a comprehensive error characterization for episodic events. Additionally, a high positive model bias over the coast of Libya, near Tripoli, is successfully reduced, correcting an outdated signal in the emission base inventory.

In the Balkan subdomain panels (Figure 8, right column), large increments are visible throughout the domain, bringing the analysis into strong agreement with observations. Specifically, the DA system corrects for the misrepresented emissions from the many coal-burning PPs of the area.

To supplement the regional analysis, two specific point locations—Catania (Italy) and Šabac (Serbia)—were selected to examine the temporal evolution of SO₂ in greater detail (Figure 9). By focusing on these discrete points, the analysis avoids the potential smoothing or ‘distortions’ inherent in area-averaged statistics, allowing for a more precise evaluation of the assimilation’s impact near high-intensity sources.

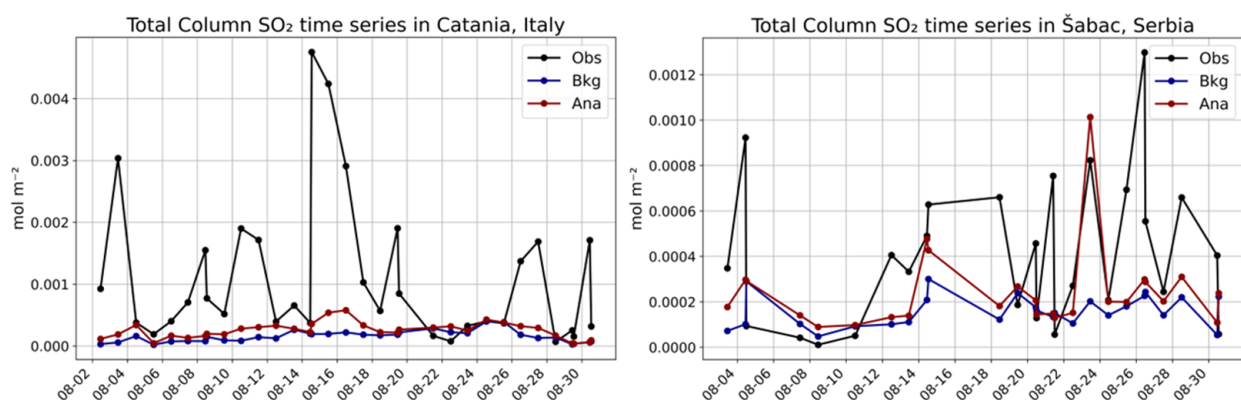


Figure 9. Comparison between the observed (black), the background (blue) and the analysis (red). SO₂ total vertical columns for each assimilation time in Catania (Italy) (left) and in Šabac (right). Mean values for each orbit are shown; background and analysis fields are filtered to match observations in number.

Both time series (Figure 9) display a consistent coupling between observations and analysis, alternating large positive increments with smaller ones depending on the level of confidence assigned to each observation and model first-guess. Figure 9 shows the values of the pixel closest to each city location for each assimilated orbit. Between 13 and 15 August, an eruption occurred, as reported by INGV [78,79]. SO₂ total columns as observed by TROPOMI over the city of Catania are shown in black on the bottom-left panel. A poor model first-guess, in blue, follows the omission of volcanic eruptions in the emission inventory. As shown in red, the analysis diverges from the background to capture the volcanic peak. While the time series in Figure 9 demonstrates a significant improvement in the analysis’s ability to track the temporal variability of SO₂, the observed peaks remain

consistently higher than the modeled values. This discrepancy is primarily attributed to the conservative weighting of satellite observations within the filter, which prioritizes the mitigation of systematic model biases over the assimilation of extreme, localized spikes.

3.2. Evaluation in Model Space

This section shows how the assimilation scheme acts at different heights by comparing the analysis with the basecase run. As mentioned in Section MINNI-DART Coupling Settings, the emissions were perturbed in the same way at all levels and the perturbations are proportional to emission levels. Figure 10 shows the differences in monthly mean SO₂ concentration between the analysis and the basecase simulation across five vertical levels (20 m, 125 m, 975 m, 2580 m, and 11,790 m) for August 2023. The basecase simulation, which excludes data assimilation, consists of a single simulation informed by unperturbed boundary conditions and emissions. The domain-averaged results of this simulation is equivalent to those of the ensemble-free experiment used by the assimilation procedure.

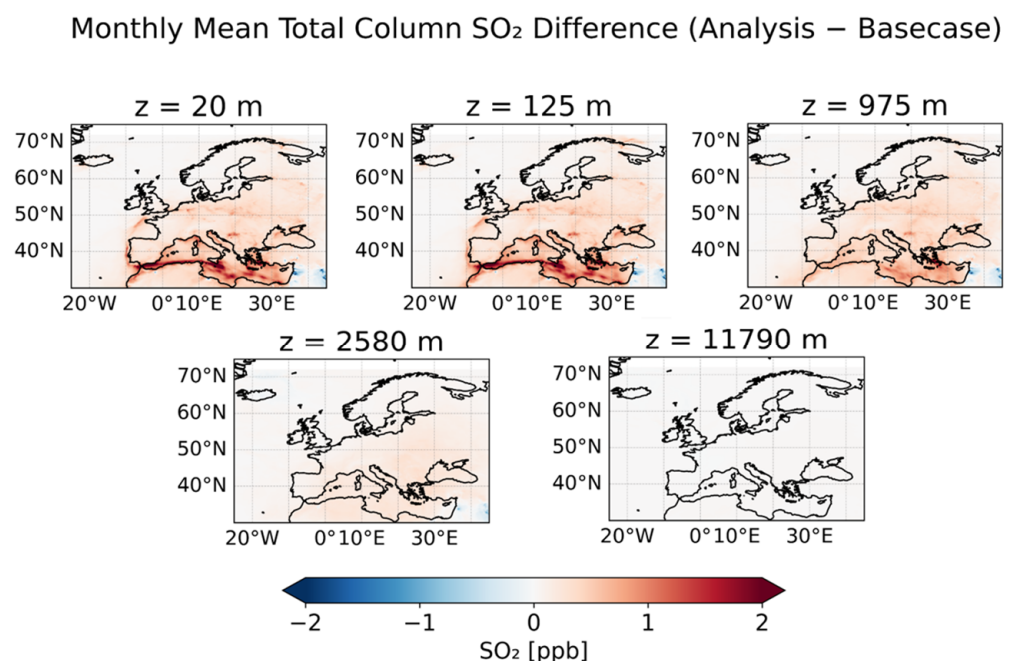


Figure 10. Differences in monthly means SO₂ concentrations between the ensemble mean analysis and the basecase simulation at five vertical levels.

In Figure 10, positive values (red), indicate an increase in SO₂ concentrations induced by the assimilation, whereas negative values (blue) correspond to decreases. It can be noted that the assimilation introduces substantial positive corrections on the level closest to the surface (20 m). At altitudes below 1 km, positive increments persist but exhibits reduced magnitude and spatial extent. This indicates that the assimilation's impact is mostly confined to the lower troposphere where the large majority of emitters are. Above 2.5 km, the differences are negligible. As already shown in Section 3.1, the largest increments are concentrated over industrial regions and shipping corridors, suggesting that the data assimilation system compensates for misestimated emissions. Since volcanic emissions are omitted and the vertical localization is turned off, the filter primarily allocates corrections to the lower troposphere, where the background SO₂ mass is most concentrated and the background variance is highest. Further considerations regarding these findings are deferred to Section 4.

3.3. Evaluation with Surface Measurements

The MINNI-DART experiment is evaluated against independent ground-based observations of the European Environment Agency (EEA) air quality network that measures surface SO_2 concentrations on an hourly basis (Figure 11). Only background stations are selected, since given the relatively coarse resolution of the model, only background SO_2 can be simulated. Background stations are those where pollution levels are representative of the average exposure of the general population or vegetation [3]. The comparison between model output (pink), basecase simulation (yellow) and measurements (green, only background stations are considered) reveals that the assimilation significantly reduces the negative bias characteristic of the basecase simulation, effectively shifting the analysis to align more closely with the observed mean values.

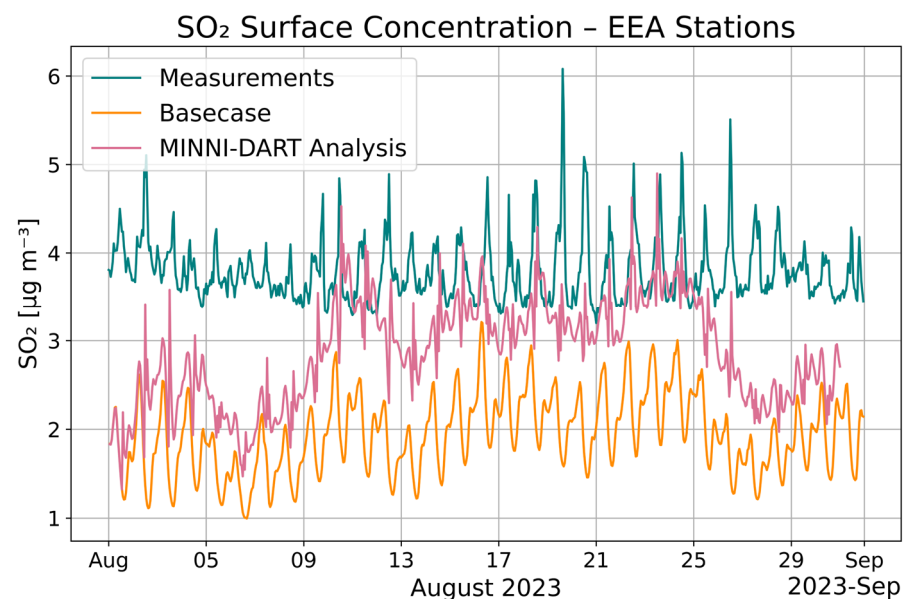


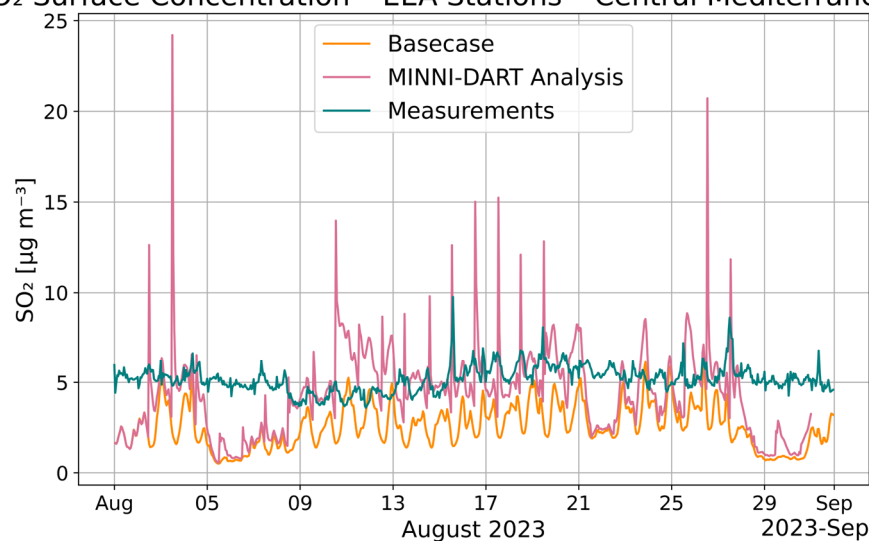
Figure 11. Time series of hourly SO_2 surface concentrations at EEA background monitoring stations (Measurements, in green) and simulated with MINNI (Basecase, in yellow) and MINNI-DART (Analysis, pink) during August 2023. Concentrations are expressed in $\mu\text{g m}^{-3}$ and are averaged over the whole domain.

The corrections were higher between 9 and 26 August. During this period, the ground-based observations exhibited a clear increase in the occurrence of more intense and sharply defined daytime peaks, with concentrations repeatedly reaching or exceeding $5.0 \mu\text{g m}^{-3}$, culminating in the maximum observed value of $6.2 \mu\text{g m}^{-3}$ on 19 August. This surge in concentration and variability was likely driven by a high-pressure system, which induced atmospheric stagnation [80]. Persistent summer high-pressure systems lead to very stable nocturnal conditions and diurnal convective conditions, with fully developed buoyancy-driven turbulence. These conditions increase the ensemble spread and the background error covariance, reducing the reliability of the model background with respect to observations and prompting DART to give more weight to satellite data when updating the background. Conversely, at the beginning and at end of the month (before the 9 and after the 26 August), with the onset of two troughs, atmospheric dispersion conditions were, on average, more neutral, leading to a reduced diurnal variation in concentrations. The model error is driven by fluctuations in emissions (and BCs), the effects of which are either amplified or dampened by the meteorological conditions governing the diurnal cycle. For a given level of fluctuation, conditions that favor a larger diurnal amplitude (i.e., 9–26 August) correspondingly increase the model error. Consequently, during periods where meteorology

dampens this amplitude (i.e., early and late August), the resulting surface concentration signals are much smaller. This is further exacerbated by the satellite's reduced sensitivity close to the ground. Facing these weak and noisy innovations, the filter applies only conservative increments to the base simulation. This cautious weighting is a critical feature of the DA system, as it prevents the propagation of observational noise into the analysis.

Figure 12 shows the same time series of SO_2 concentrations as Figure 11 but averaged over the monitoring stations within each subdomain (see Figure S1 for station locations). In the Balkan domain (Figure 12, lower panel), ground-based observations indicate very high surface SO_2 concentrations, whereas in the Central Mediterranean (Figure 12, upper panel) values remain lower, even during the volcanic activity on the 15, likely due to the fact that the volcanic plume is well above the surface and there is no impact on the measurements close to the volcano.

SO_2 Surface Concentration – EEA Stations – Central Mediterranean Sea



SO_2 Surface Concentration – EEA Stations – Balkan Area (Serbia)

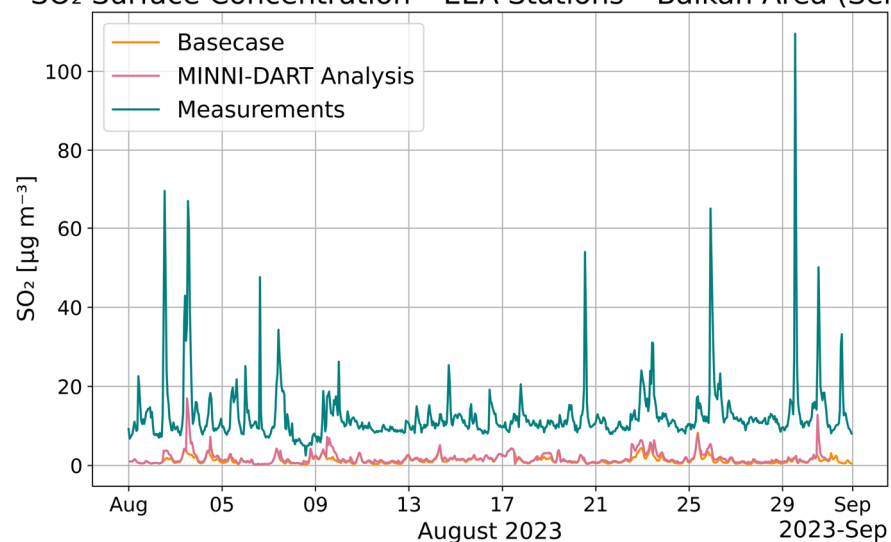


Figure 12. Time series of hourly SO_2 surface concentrations at EEA background monitoring stations (Measurements, in green) and simulated with MINNI (Basecase, in yellow) and MINNI-DART (Analysis, pink) during August 2023. The upper panel shows the central Mediterranean Sea subdomain, while the lower panel shows a portion of the Balkan region over Serbia. The locations of the stations in the two subdomains are shown in Figure S1. Concentrations are expressed in $\mu\text{g m}^{-3}$ and are averaged over the subdomains.

Although increments due to assimilation exist in both subdomains (noting the different panel scales), their agreement with observations differs substantially (Figure 12). In the Balkans, assimilation fails to reproduce both background and peaks of measured SO₂ concentrations, while in the Mediterranean domain, simulated peaks frequently exceed ground-based measurements. These discrepancies arise from the intrinsic differences between the observing systems. Satellite retrievals represent total column SO₂, whereas in situ measurements sample near-surface concentrations. Over the Balkans, a substantial fraction of near-surface SO₂ is therefore likely not captured by satellite observations, limited by their low sensitivity as we approach the ground. Conversely, in the Mediterranean domain, volcanic plumes detected aloft may be vertically misplaced toward the surface in the assimilation, resulting in overestimated surface concentrations relative to in situ observations. These shortcomings are further discussed in the following Section 4. The performances of the MINNI-DART analysis and basecase simulations against the EEA's air quality network observations were evaluated using the following statistical metrics: Root Mean Square Error (RMSE), Mean Bias (MB), Normalized Mean Bias (NMB), and Pearson correlation coefficient (R). Their values reported in Table 2 were first calculated locally over time for each individual monitoring station and, subsequently, averaged across all stations within each domain to provide domain-wide summary statistics. For all three domains, European, central Mediterranean and Balkan, the analysis has a better agreement with the stations than the basecase run. It can also be noted that RMSE and MB have higher values over the two subdomains than over the whole domain, likely attributable to higher SO₂ concentrations at stations in proximity of high-emission sources.

Table 2. Statistical evaluation metrics comparing the Basecase and the MINNI-DART analysis simulations against EEA's air quality network observations across the whole domain and specific subdomains (south-central Mediterranean Sea and Balkan) for August 2023. The metrics include Root Mean Square Error (RMSE), Mean Bias (MB), Normalized Mean Bias (NMB), and Pearson correlation coefficient (R). The last row shows the number of stations within the domain that had records in August 2023.

Metric	Domain					
	European Domain		Central Mediterranean Sea Subdomain		Balkan Subdomain	
	Basecase	Analysis	Basecase	Analysis	Basecase	Analysis
RMSE ($\mu\text{g m}^{-3}$)	1.95	1.16	2.94	2.64	13.82	13.20
MB ($\mu\text{g m}^{-3}$)	−1.85	−0.93	−2.63	−0.82	−11.37	−10.87
NMB (adim)	−48.70	−23.62	−50.61	−15.31	−89.38	−82.80
R (adim)	−0.16	−0.02	0.11	0.06	0.23	0.45
Stations count	849		31		2	

As generally expected for SO₂ during summer periods [81], the correlations (R) are low both for analysis and basecase. However, a significant improvement in R from 0.23 for basecase to 0.45 for analysis is observed in the Balkans domain under the influence of anthropogenic sources.

4. Discussion

The implementation of EAKF through DART in air quality modeling system MINNI and its configuration show that the assimilation process brings the modeled SO₂ fields closer to the observations. As a result, the analyses better reproduce the spatial and temporal variability seen in both satellite data (Figure 4, Figure 5 and Figure 9) and in situ measurements (Figure 11), supporting the overall reliability of the setup.

However, several methodological choices have influenced the behavior of the present DA application, and their potential impact is further discussed. Unlike previous studies [17,19], where the SO₂ background error covariance matrix was specifically designed to represent the volcanic eruptions—by deliberately maximizing errors in the mid-troposphere—this work employs a flow-dependent background error covariance matrixes derived from ensemble perturbations. These perturbations focus on anthropogenic emissions and boundary conditions, while volcanic emissions are neither included (consistent with standard reference MINNI runs) nor perturbed. Consequently, model uncertainty during volcanic events was likely sub-optimally represented. In some instances, volcanic plumes may have been successfully assimilated only due to its coincidence with ensemble perturbations from other sources (e.g., ship emissions), which artificially increased the local ensemble covariance and favored corrections in those regions, as highlighted by Figures 3, 10 and 12 upper panel. Introducing small point sources distributed throughout the entire volumetric grid and centered on Etna's main crater, with covariance inflation used to amplify the signal, may help overcome these shortcomings and will be tested in future analyses. Nevertheless, the use of inflation mitigated this drawback by increasing the ensemble variance, improving agreement with the expected innovations and reducing sensitivity to perturbations. For the same reason, the risk of underestimating the background error for not including the meteorological fields in the perturbation targets is anyhow limited. Moreover, given the complexity of perturbing the meteorological fields and subsequently applying a mass-conservation filter, it was preferred to postpone this aspect to a future implementation, especially considering that several previous studies did not include it either.

Further complications in the setup of the assimilation algorithm arise from the coexistence of observations related to distinct SO₂ source types—anthropogenic and volcanic—which exhibit markedly different temporal and vertical characteristics. This is particularly evident in the case of vertical localization. Since the DART version used here may also apply a fixed vertical localization (e.g., centered on the maximum value of the averaging kernel profiles [62], the height and damping parameters that control the placement of analysis increments cannot adapt dynamically to the nature of the observed concentrations. Therefore, tuning the system to better capture one source type can deteriorate the performance of the other. An explicative case is given in Figures S2–S5, where two runs, one without vertical localization and the other with a Gaspari–Cohn centered at 2580 m, are compared in the Etna and Balkan regions. When localization is imposed around a specific altitude, high concentration values areas are created, whereas without vertical localization the total column increment is distributed more uniformly across model levels, lowering the risk of significantly misplacing corrections to unrealistic plumes (Figure S2). However, as shown in Figure 12, the selected configuration still exhibits limitations, as ground-based observations are not fully reproduced in either subdomain. Future developments could address these issues by assimilating the four SO₂ retrieval products separately, each with ad hoc vertical localization parameters, or implementing adaptive localization methods to better represent the vertical structure of the analysis increments (e.g., following [82]). Another example in this direction was presented by [10], who successfully tested using additional near-real-time layer height data to adapt the positioning of increments in the model space. Overall, the adopted configuration proved to be an acceptable trade-off since it made “visible” Etna plume (Figures 3 and 8) and industrial plumes in the Balkan area (Figure 8).

The limited ensemble size due to high computational cost represented a major constraint of the present application. Running larger ensembles would significantly improve model uncertainty representation, but at the expense of computational feasibility. Another

important challenge lies in designing realistic perturbations, since overly simplistic representations of uncertainty can bias the assimilation outcome. Accurately representing model error structures is therefore crucial and not trivial. The sensitivity to emission perturbations shown in Section S3.4 gives an example of how assimilation is driven by the size of the perturbations. Emerging machine learning techniques for ensemble emulation or reduced-order modeling may help overcome these limitations by providing efficient ensemble surrogates [83–85].

Finally, since the assimilated observations occur within a limited time window when satellite data are available, over an area that covers a part of the simulated domain, and volcanic point sources are not known, the model correction tends to fade over time as SO₂ fields produced by CTM are highly driven by the underlying emission fields. As noted in previous works (e.g., [63]), emission rates represent more effective control variables than initial conditions for long-lived species such as SO₂. The upcoming hourly data stream over the whole of Europe from Sentinel-4 is expected to alleviate this limitation, allowing for more continuous IC corrections and better maintenance of assimilation gain over time. The benefits of assimilating observations across multiple time windows were also demonstrated by [19].

5. Conclusions

This modeling application integrates SO₂ satellite data with simulations produced by MINNI regional air quality modeling system, using the Ensemble Adjusted Kalman Filter (EAKF) through the DART assimilation framework. Sentinel-5P/TROPOMI SO₂-COBRA total column retrievals were assimilated for August 2023, without restricting the analysis to volcanic events, thus jointly addressing anthropogenic and volcanic contributions. The methodological setup and perturbation strategy were designed according to existing knowledge on satellite trace gases retrievals data assimilation in a CTM using an EAKF filter.

The MINNI-DART system successfully integrated satellite observations with the CTM fields, producing analysis states that were more consistent with both satellite and in situ measurements than the background simulations. On average, the correction of VCDs on the monthly means was of $2 \times 10^{-5} \text{ mol m}^{-2}$ (Figure 4), with peaks up to $3.3 \times 10^{-4} \text{ mol m}^{-2}$ (Figure 3). At the surface, domain-averaged corrections of SO₂ concentrations were as high as $2.6 \mu\text{g m}^{-3}$ (Figure 11). The ensemble variance consistently decreased after the assimilation, confirming the correct functioning of the EAKF filter. The application of the assimilation algorithm corrected both model over- (NW Atlantic Ocean and Middle East region) and under-estimations (along major shipping corridors, around the Etna Volcano, and where wildfires were reported (Greece)), (Figure 3d). The subdomain analyses over an area in Central Mediterranean and another in the Balkan region further demonstrate the EAKF filter's efficacy, showing that the system is highly responsive even to plumes not previously characterized in the background—such as those arising from misrepresented power plant emissions or unmodeled volcanic activity—while maintaining structural stability. Without over-fitting extreme, localized spikes, the system provides an improved reconstruction of SO₂ fields.

The analysis of the horizontal distribution of concentration at different vertical levels highlighted that assimilation increments were primarily confined to the lower troposphere, where most sources are located, while the impact decreased with altitude (Figure 10). The evaluation against EEA monitoring data confirmed that the assimilation significantly reduced persistent negative biases in the backgrounds, demonstrating its ability to produce more accurate and observation-consistent analyses (Figures 11 and 12) where emissions are not known, despite notable residual biases remaining in some regions. These findings

further indicate that this setup of the assimilation algorithm is able to transfer information from column observations toward near-surface concentrations, supporting thus the use of satellite data to improve air quality assessments at ground level.

Future developments will focus on addressing the limitations identified here such as ensemble size, fixed vertical localization, etc. In particular, including explicit volcanic sources, testing adaptive localization approaches, and extending the control variables to emission fluxes will be incorporated. The forthcoming Sentinel-4 mission, providing hourly SO₂ observations, will offer new opportunities to maintain assimilation updates more frequently and sustain corrections over time; nevertheless, the increased temporal frequency of satellite observations will not by itself overcome the limited sensitivity to near-surface SO₂. Future sensitivity analyses will further assess the system's robustness, and the impact of superobbing to reduce representative errors. Lastly, future work should further investigate the role of meteorological conditions in modulating the vertical and horizontal redistribution of SO₂ too, with particular attention to wind shear, low-level jets, and atmospheric turbulence, which may substantially influence SO₂ concentrations and transport under specific meteorological regimes [86].

In summary, to the authors' knowledge, this work provides the first demonstration of an operationally oriented SO₂ data assimilation framework for a regional CTM using a filtering approach and mixed-type SO₂ observations. The results confirm the feasibility and added value of integrating total column SO₂ retrievals into air quality forecasting chains, paving the way toward more accurate, observation-driven monitoring of atmospheric pollutants over Europe.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/atmos17090875/s1>. The Supplementary Materials include a mathematical description of the Ensemble adjustment Kalman filter and of the perturbations method, a map of the EEA monitoring stations in the sub-domains, a list of the 7 biggest coal-fired power plants in Serbia by nominal capacity, sensitivities to vertical localization and emission perturbations and computational details for the presented simulation. References [35,36,62,87,88] are cited in the supplementary file.

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Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The MINNI-DART implementation is hosted in a fork of the DART repository, available on Github at: <https://github.com/alessandroda/DART/tree/main> (accessed on 1 July 2026). In the same forked directory, a Pythonv3.10-based orchestrator script is also provided. It coordinates the running of both FARM CTM and DART executables, carrying on an online data assimilation as a sequence of well-defined steps that govern the following elements: the forecast model, the assimilation engine, and observations to be assimilated. The datasets and/or analysis

generated during the current study are not publicly available due to the high volume of data but are available from the corresponding author upon reasonable request.

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