



Technical note: Assessing CO₂ fluxes over Greece using CTDAS-WRF – A sensitivity analysis

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Abstract. CO₂ is a key driver of climate change and knowledge of its emissions is a prerequisite for climate mitigation. CO₂ emissions over Greece in 2019 are assessed using a regional atmospheric inversion framework based on the CarbonTracker Data Assimilation Shell coupled with the Weather Research and Forecasting model
25 (CTDAS-WRF). Ground-based CO₂ measurements from an urban background site (Thissio), a regional background site (Finokalia), and a high-altitude station (Helmos), together with satellite-derived column-averaged CO₂ observations from OCO-2, are assimilated to optimize CO₂ emissions by allowing 50% of uncertainty on CAMS anthropogenic CO₂ emissions. Biospheric and fire fluxes are kept fixed. Sensitivity tests of localization length and anthropogenic emissions uncertainty assess parameter choices and results robustness. For 600 km
30 localization, annual posterior anthropogenic emissions over Greece are 55.6 ± 1.9 Mt, close to CAMS prior (55.0 Mt) and below EDGAR (61.5 Mt) and Greece's reported total (65.7 Mt). The corresponding total CO₂ net flux over Greece is 118.3 ± 1.9 Mt. Since natural fluxes are not optimized, inferred adjustments cannot be attributed solely to anthropogenic emissions. Optimization reduces mean model bias by 80% and RMSE by about 40% at Thissio. This study demonstrates a CTDAS-WRF framework for regions with strong mesoscale variability and
35 complex airflow. Urban background observations strongly reduce emission uncertainty around Athens, while regional background observations inform long-range transport and have less effect on local emission inversions. Denser observation network is therefore needed covering major local emission sources. Future work should optimize anthropogenic emissions jointly with biospheric and fire fluxes.



40 1. Introduction

Carbon dioxide (CO₂) is the dominant greenhouse gas driving climate change and is responsible for most of the observed global warming since the industrial era (IPCC, 2023). CO₂ is emitted during combustion activities, including transportation, energy production, cement production, wood burning and open fires, as well as during land-use change that also involves burning. Overall, fossil fuel use and cement production account for about 90% of the anthropogenic CO₂ emissions, with the remaining 10% originating from land-use change, such as deforestation and forest degradation (Friedlingstein et al., 2025). CO₂ is removed from the atmosphere by the oceans and through photosynthesis by terrestrial vegetation. From the early 1960s to the late 2010s, the oceans absorbed an average of $25 \pm 2\%$ of total human-caused CO₂ emissions (Gruber et al., 2023). Nevertheless, due to progressive ocean acidification, the global ocean's capacity to absorb atmospheric CO₂ has declined by approximately 10 % since preindustrial times (Gruber et al., 2023). In addition, logging, permanent agriculture and wildfires intensified due to climate warming were identified as major drivers of forest loss and thereby they are able to change forests from a sink to a source of CO₂, permanent agriculture represents 35% of forest loss from 2001 to 2022 (Sims et al., 2025). Since the onset of the industrial era, atmospheric CO₂ concentrations have increased by 50%, rising from approximately 280 ppm before industrialization to over 420 ppm in 2023 (Friedlingstein et al., 2025). A complete elimination of CO₂ anthropogenic emissions would lead to a gradual decline in atmospheric CO₂ as land and ocean sinks continue to take up carbon, with the multi-model mean estimated decline reaching roughly 90 ppm over a century in idealized zero-emissions experiments (Intergovernmental Panel on Climate Change (IPCC), 2023). Long-term climate stabilization ultimately requires lowering atmospheric CO₂. Achieving this depends on accurately quantifying CO₂ emissions and their spatial and temporal variability to assess the effectiveness of mitigation efforts under the Paris Agreement.

Estimates of CO₂ fluxes rely on bottom-up and top-down approaches. Bottom-up approaches of anthropogenic CO₂ emissions combine activity data with emission factors (Andres et al., 2012; Greece (NIR), 2024). Although essential for inventory reporting, such methods can be subject to significant uncertainties at regional scales, especially in regions with complex energy systems or limited statistical reporting (Oda et al., 2019; Solazzo et al., 2021). Complementary to bottom-up inventory estimates, top-down approaches constrain surface fluxes by assimilating atmospheric CO₂ observations via transport models, which link observed concentrations with underlying emissions and uptake (Peters et al., 2005, 2007; Peylin et al., 2013). Atmospheric inversions have become essential tools for evaluating CO₂ budgets (Friedlingstein et al., 2025), providing means to verify reported emissions. Global inversion systems typically operate at coarse spatial resolutions, which limits their ability to resolve local emission sources and transport over complex terrain. The coupling of CarbonTracker Data Assimilation Shell (CTDAS) (van der Laan-Luijkx et al., 2017) with the Weather Research and Forecasting (WRF) model (Skamarock et al., 2019) presents an effective method for regional-scale inversions (Ioannidis et al., 2026; Reum et al., 2020). CTDAS employs an Ensemble Kalman filter to optimize surface fluxes without requiring an adjoint model (Peters et al., 2005). When coupled with WRF, the system benefits from high-resolution meteorological fields that can capture mesoscale processes that influence CO₂ transport. Regional inversions are highly useful for countries with complicated emission distributions and topography. Greece offers a strong test case for such an approach. The country's emission landscape is characterized by concentrated fossil-fuel combustion for power generation, industrial activity and transportation in urban centers such as Athens and Thessaloniki (Kanakidou et al., 2011) and seasonal variations linked to heating and tourism demand (Bougiatioti



80 et al., 2024; Vourdoubas, 2019). According to the national inventory report of Greece (Greece (NIR), 2024), the country's total greenhouse gas emissions (excluding Land-Use, Land-Use Change and Forestry, LULUCF) in 2019 were approximately 86 Mt CO₂ eq, with CO₂ contributing about 66 Mt in 2019. 60 Mt were attributed to fossil fuel combustion processes. Although national inventories provide detailed sectoral statistics, they often lack the spatial and temporal resolution that is necessary for emission monitoring. Furthermore, the meteorology and
85 physical geography of Greece make it difficult to accurately simulate atmospheric CO₂ transport. Mountainous terrain and with numerous islands create complex mesoscale circulation patterns. These features highlight the importance of using a model like WRF that can simulate detailed meteorological processes with high resolution, within an assimilation framework that adjusts emission fields based on observations. Most regional inversion studies in Europe have focused on central and western regions where dense observational networks exist (Broquet
90 et al., 2013; Monteil et al., 2020). The eastern Mediterranean, however, remains underrepresented in such analyses. Despite its strategic location (Lelieveld et al., 2002), high-resolution atmospheric inversion studies that consolidate observations and simulations in robust emission inventories over Greece remain scarce. Implementing an inversion system based on CTDAS-WRF for Greece is an opportunity to fill this gap, enabling a more accurate assessment of national CO₂ emissions in support of the clean carbon strategic development of the country.

95 For this study, we implemented and evaluated the CTDAS-WRF framework over Greece in the year 2019, developing a high-resolution inversion system configured to the region's meteorology and emissions. We assimilated both in situ and satellite-based CO₂ observations to optimize CO₂ emissions over Greece using anthropogenic emissions as state vector and evaluate their spatial and temporal consistency. We also performed sensitivity tests to determine the optimal set up for CO₂ inversions for Greece, specifically on the prior flux
100 uncertainty and the localization length used for the inversions. We assessed the performance of the optimized fluxes with respect to (only sparsely available) independent data. Finally, we compared the optimized (posterior) emissions with existing bottom-up inventories (CAMS, EDGAR) and the official national inventory to assess convergence and discrepancies. For simplicity, this study considered uncertainties only of anthropogenic emissions of CO₂. It provides a setup of the assimilation framework and a thorough testing of parameters used as
105 a basis for more complex setups that are necessary for accurate top-down estimates; most importantly; by taking into account the uncertainties of biological and fire CO₂ fluxes. This work contributes to the growing European effort toward observation-based emission verification in the region. It is in line with initiatives under the Copernicus program to develop a CO₂ Monitoring, Verification and Support (CO₂MVS) (Janssens-Maenhout et al., 2020) capacity that integrates satellite observations, in situ measurements, and atmospheric modeling to
110 quantify anthropogenic CO₂ emissions at regional and national scales, with the long-term objective of extending such capabilities to the local scale. Beyond the national context, this study acts as a prototype framework for applying CTDAS-WRF in other Mediterranean regions where there is strong mesoscale variability and limited observational coverage.

2. Methodology

115 2.1. Experiment setup

Simulations were performed with the CTDAS-WRF modeling framework from 18 December 2018 to 14 January 2020, with the periods outside 1 January–31 December 2019 treated as model spin-up and spin-down, respectively, and excluded from the analysis. The year 2019 was chosen as it is the first year with available CO₂ monitoring in



Athens (Bougiatioti et al., 2024). Two main experiments were conducted in this study. First, a prior simulation was performed using prior surface CO₂ fluxes without any data assimilation. Second, a posterior simulation was carried out in which anthropogenic CO₂ emissions were optimized using CTDAS. The posterior simulation assimilates both ground-based in situ CO₂ measurements and satellite-derived column-averaged CO₂ observations within a single inversion framework. The inversion targets anthropogenic CO₂ emissions only, which are adjusted through multiplicative scaling factors applied to the prior inventory. These scaling factors are estimated on a weekly time scale and defined at the level of Greek administrative prefectures, allowing for spatially differentiated emission adjustments within the country. Biogenic CO₂ fluxes, biomass burning emissions, and lateral boundary CO₂ concentrations are not optimized in the inversion. Therefore, the results of the performed inversions have to be viewed as optimization of the total CO₂ emissions over Greece. Sensitivity studies have been performed to investigate the impact of uncertainty of the prior anthropogenic emissions as well as of the assimilation window on the posterior emissions (Table 1; Test 1 to 4). Furthermore, tests were performed to determine the optimal localization length, i.e. the length scale up to which an assimilated observation can influence the state vector (Thanwerdas et al., 2025). Localization lengths for observations of 180 km, 400 km and 600 km have been tested (Table 1).

Table 1. Simulations performed for this study in addition to the forward simulation, using the prior emissions. All simulations started in December 2018. The simulated period of December was used as a spin-up as mentioned above. The prior emissions for January 2019 are 5.53 Mt

Sensitivity Tests	Uncertainty in anthropogenic emissions	Assimilation Window	Localization Length	January 2019 posteriori emissions
Test 1 (1 month run)	50%	3 weeks	180 km	6.07 Mt
Test 2 (1 month run)	10%	2 weeks	180 km	6.73 Mt
Test 3 (1 month run)	25%	2 weeks	180 km	6.56 Mt
Test 4 (1 month run)	100%	2 weeks	180 km	6.20 Mt
Test 5 (base)	50%	2 weeks	180 km	6.12 Mt
Test 6	50%	2 weeks	400 km	5.96 Mt
Test 7 (reference)	50%	2 weeks	600 km	5.93 Mt

2.2. WRF modeling system

The Weather Research and Forecasting model coupled with Chemistry (WRF-Chem v4.2) (Skamarock et al., 2019) and particularly the greenhouse gas configuration (WRF-GHG) (Beck et al., 2011), which treats trace gases as passive tracers. The outer domain (D01) spans the eastern Mediterranean at a horizontal resolution of 36 km × 36 km, capturing major transport pathways from the Balkans, North Africa, and the Middle East (Figure 1a). The inner domain focuses on Greece with a finer 12 km × 12 km horizontal grid. A one-way nested domain configuration was employed for this study. The model uses 35 vertical layers defined on WRF's hybrid terrain-following sigma–pressure coordinate, extending from the surface to 50 hPa. The physical parameterizations used for the simulations are summarized in supplementary Table S1.

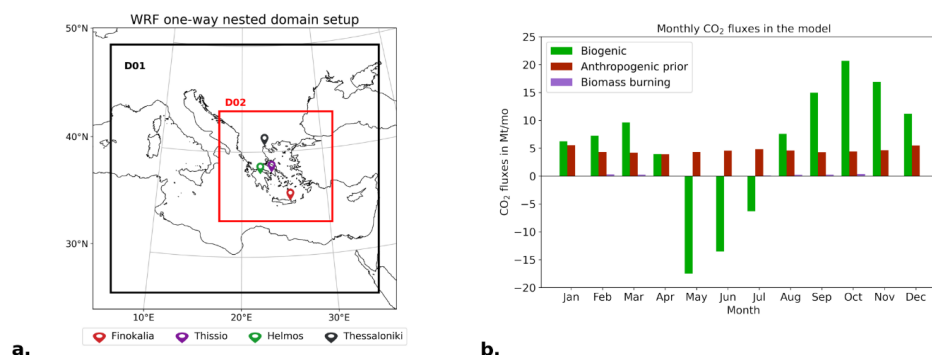


Figure 1: (a) WRF domain setup, with horizontal resolutions of 36km x 36km (D01) and 12km x 12km (D02). Colored markers indicate the locations of the ground-based CO₂ observation sites Finokalia (Red), Helmos (Green), and Thissio (Purple) used for the inversions and the FTIR site at Thessaloniki (black) used for independent model evaluation (b) Monthly mean CO₂ emissions over Greece in the model.

The meteorological initial and boundary conditions were derived from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA5 reanalysis dataset, which provides global data at a $0.25^\circ \times 0.25^\circ$ resolution (Hersbach et al., 2023). To enhance simulation accuracy, spectral nudging to ERA5 meteorology was applied hourly to the parent domain, as it has been shown to outperform grid nudging in previous studies (Zittis et al., 2018). No nudging was applied to the finer domain to retain the detailed spatial variability afforded by its higher resolution. Geographical input datasets except land use and topography were obtained from the NCAR repository (https://www2.mmm.ucar.edu/wrf/users/download/get_sources_wps_geog.html; last access 2.12.2025). Land use information was taken from the CORINE Land Cover 2018 dataset, available at 100 m resolution (Copernicus Land Monitoring Service, 2020), while topography was represented using the European Digital Elevation Model (EU-DEM) with a 30 m spatial resolution (Copernicus DEM, 2022). Chemical initial and boundary conditions were provided by the optimized Copernicus Atmosphere Monitoring Service (CAMS) product (v20r2), available at $3.75^\circ \times 1.9^\circ$ spatial and 3-hourly temporal resolution (Chevallier et al., 2010; Copernicus Atmosphere Monitoring Service, 2020). These fields were interpolated to a regular $1^\circ \times 1^\circ$ grid and subsequently interpolated onto the WRF grid to provide the initial and lateral boundary conditions. Biomass burning emissions were taken from the Fire INventory from NCAR (FINN) version 2.5, which integrates active fire detections from MODIS and VIIRS with land cover data, emission factors, and fuel load estimates to produce daily high-resolution (1 km) biomass burning emissions (Wiedinmyer et al., 2023). Biogenic emissions were simulated online using the Vegetation Photosynthesis and Respiration Model (VPRM; (Mahadevan et al., 2008)). VPRM uses satellite-derived inputs from MODIS, specifically the Enhanced Vegetation Index (EVI) and the Land Surface Water Index (LSWI), provided at 500 m spatial resolution and updated every eight days. Note that recently, (Glauch et al., 2025) used the higher resolution Sentinel-2 instead of MODIS in VPRM and found significant improvements in the estimation of the European carbon budget. These changes will be considered in future studies. Meteorological variables such as temperature and Photosynthetically Active Radiation (PAR) were obtained directly from WRF



calculations. Through this method, VPRM estimates the exchange of CO₂ between vegetation and the atmosphere, commonly referred to as the Net Ecosystem Exchange (NEE). Anthropogenic emissions were taken from the CAMS dataset (version 5.3; Granier et al., 2019), which provides monthly emission estimates at a spatial resolution of 0.1° × 0.1° for 20 individual source sectors and their combined total. In this study, we utilized the aggregated variable representing the total anthropogenic emissions across all sectors. The monthly variation of the prior biogenic, biomass burning and anthropogenic emissions of CO₂ in the model in 2019 are given in Figure 1b. Negative fluxes (from May to July) indicate a net CO₂ uptake by vegetation.

2.3. CarbonTracker Data Assimilation Shell

CTDAS is an ensemble-based inverse modeling system designed to estimate surface fluxes or emissions of atmospheric trace gases by assimilating atmospheric concentration observations. In this study, CTDAS was used to optimize CO₂ emissions by integrating both satellite observations from OCO-2 and ground-based in situ measurements. The system applies an ensemble Kalman smoother with a fixed-lag assimilation window (van der Laan-Luijkx et al., 2017; Peters et al., 2005), adjusting prior emission estimates to reduce discrepancies between modeled and observed CO₂ concentrations. The state vector x represents scaling factors applied to the prior anthropogenic CO₂ emissions, and the optimization process minimizes the following cost function:

$$J(x) = (y^o - H(x))^T R^{-1} (y^o - H(x)) + (x - x^b)^T P^{b-1} (x - x^b) \quad (1)$$

Here, y^o denotes the observed CO₂ mole fractions, with their uncertainties represented by the model-data-mismatch covariance matrix R . The observation operator H projects the model state into the observation space through the atmospheric transport model, which in this study is WRF-Chem. The prior state vector x^b represents the initial emission estimates, and its uncertainty is characterized by the prior error covariance matrix P^b . Minimizing this cost function gives the posterior estimate of the state vector (x^a),

$$x^a = x^b + K(y - H(x^b)) \quad (2)$$

With posterior error covariance (P^a):

$$P^a = (I - KH)P^b \quad (3)$$

Where I is the identity matrix and K is the Kalman Gain:

$$K = (P^b H^T)(H P^b H^T + R)^{-1} \quad (4)$$

The Kalman gain determines how strongly the prior state is drawn towards the observations, balancing the prior uncertainty P^b against the model-data-mismatch uncertainty R . In the ensemble system used here, P^b and the products $P^b H^T$ and $H P^b H^T$ are not calculated explicitly but are estimated from the ensemble. The assimilation system employs an ensemble of 150 members, one representing the mean and the other 149 using perturbed versions of the prior emissions, sampled from their probability distribution defined by mean and covariance matrix, to generate an ensemble of 150 forward CO₂ simulations. In this study, we used a temporal resolution of one week for calculating changes in the state vector and a two-week assimilation window. The assimilation window defines the temporal period over which observations influence the state vector and must be long enough to accurately represent CO₂ dispersion across the study region. Note that a sensitivity simulation with a three-



weeks assimilation window has shown no significant difference in the optimized emissions (difference < 1%) over one month of inversion (Simulations Test 1 and Test 5 in Table 1). Through this procedure, CTDAS estimates weekly scaling factors for anthropogenic emissions by assimilating both ground-based and satellite observations as discussed in the following sections. As shown in Table 1, most inversion simulations performed here assume a
 215 50% uncertainty for anthropogenic emissions, while all other sources were treated as with full confidence. Simulations Test 2, Test 3, Test 4 and Test 5 (Table 1) investigated the impact of the prior uncertainty on the inversions.

Our setup derives a scaling factor for each prefecture of Greece (Figure 2). The prior error covariance matrix P^b
 220 is diagonal. Each region is assigned an independent prior uncertainty (50 % in the base case, Table 1), with no prior correlations imposed between prefectures. During each assimilation cycle, the Kalman gain matrix (Eq. 4) determines how much the scaling factors are updated based on the information provided by the observations. In the ensemble Kalman smoother framework, this update is applied both at the current assimilation time and, through the fixed-lag configuration, also influences the previous time step within the window (of two-weeks
 225 except Test 1). To prevent an unrealistic influence of distant observations caused by the finite ensemble size (150 members), a spatial localization is applied to the Kalman gain matrix. The localization is implemented using a Gaussian function of distance between each observation location and the region centroid associated with each element of the state vector (i.e., the prefecture-specific emission scaling factors). The centroid of each prefecture is defined as the mean latitude and longitude of all model grid cells belonging to that prefecture. The localization
 230 coefficient was given by

$$a(d) = \exp\left(-\left(\frac{d}{L}\right)^2\right) \quad (5)$$

where d denotes the distance between the observation and the prefecture centroid, and L is the localization length scale. Different localization length values have been tested ranging from 180 to 600 km and the value of 600 km was chosen as the reference (see discussion section 3.3) for this study (Table 1). The initial choice of 180 km was
 235 guided by the spatial structure of the state vector, in which each prefecture is treated as a region of fully correlated prior emission scaling factors. Taking the characteristic size of these regions as the correlation length ($L_{corr} \approx 62\text{km}$, area-weighted; see Supplement S2 for the derivation and per-region values), we follow the approach used for CO_2 flux estimation by Peters et al. (2005), who set the localization length to three times the covariance scale. This $\times 3$ rule yields localization lengths of approximately 163–220 km across the different radius definitions and
 240 shape conventions (Table S2) which supports the choice of 180 km localization length. We note however, that the validity of the $\times 3$ rule for a domain of this size is not established. The physical connection of fluxes to atmospheric concentrations via transport may require a longer localization length, which is the motivation for the 400 km and 600 km sensitivity tests reported below. The localization coefficients were applied multiplicatively to the Kalman gain, reducing the influence of observations on distant prefectures while preserving strong updates for nearby
 245 regions.



Regions with different scaling factors

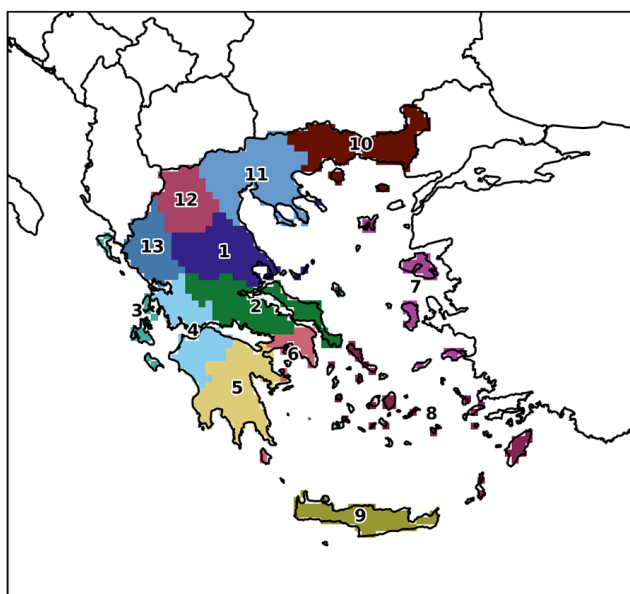


Figure 2: Greek prefectures, each assigned an individual scaling factor for its respective emissions: 1. Thessaly, 2. Continental Greece, 3. Ionian Islands, 4. Western Greece, 5. Peloponnese, 6. Attica, 7. North Aegean, 8. South Aegean, 9. Crete, 10. East Macedonia and Thrace, 11. Central Macedonia, 12. West Macedonia, 13. Epirus

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2.4. Posterior uncertainty reduction calculation

To assess how efficiently the available observations constrain the emission estimates, we quantify the posterior uncertainty reduction (UR). In the inversion, emissions are allowed to increase or decrease relative to their prior values through multiplicative adjustment factors, represented through an ensemble of possible emission states.

255 Uncertainty reduction is quantified as the fractional decrease of uncertainty after the assimilation:

$$UR = 1 - \frac{\sigma_{\text{posterior}}}{\sigma_{\text{prior}}} \quad (6)$$

where σ_{prior} and $\sigma_{\text{posterior}}$ denote the ensemble standard deviation of the emission adjustment factors before and after assimilation, respectively.

260 2.5. CO₂ ground observations -assimilated data

Hourly CO₂ measurements from all monitoring sites with continuous observations across Greece, representing its diverse atmospheric environments were used. The dataset includes observations from the Finokalia Atmospheric Observatory, Thissio and Helmos. Finokalia is operated by the University of Crete, located approximately 50 km east of Heraklion (35°20' N, 25°40' E, 250 m a.s.l.). This site has been extensively used to characterize the regional background atmospheric composition of the Eastern Mediterranean (Mihalopoulos et al., 1997; Kouvarakis et al.,

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2002; Vrekoussis et al., 2007; Kalivitis et al., 2019). Continuous in situ CO₂ measurements began in 2014 using a Picarro G2401 analyzer (Gialesakis et al., 2023). Thissio station is located at the premises of the National Observatory of Athens (NOA) (37.97° N, 23.72° E, 105 m a.s.l.), on “Nymphs Hill” in central Athens in a moderately populated area, limiting the influence of direct local emissions (Stavroulas et al., 2019). Its elevated position overlooking the city center allows it to effectively capture urban background air quality conditions, acting as a receptor point for aerosols and gases transported through local dispersion and circulation. Continuous urban background CO₂ observations have been performed at Thissio station since January 2019, using a Picarro G2401 instrument, following the same calibration protocol as at Finokalia (Bougiatioti et al., 2024; Dimitriou et al., 2021). Finally, the Helmos Hellenic Atmospheric Aerosol and Climate Change station (HAC²) station (37.98° N, 22.19° E, 2314 m a.s.l.), operated by the National Center of Scientific Research ‘Demokritos’ is the only permanent high-altitude facility for atmospheric research in Greece. It is also one of the least affected by boundary layer high-altitude stations in Europe (Collaud Coen et al., 2018), making it an excellent site for sampling free-tropospheric (FT) air masses. Continuous greenhouse gas monitoring has been conducted at Helmos since 2016 also using a Picarro G2401 analyzer (Bougiatioti et al., 2024). All available hourly observations were assimilated, with no filtering by time of day. Thus, the setup prioritizes the use of maximal available information from the sparse observation network, despite the risk of transport model inaccuracies that may degrade the results (Geels et al., 2007). Modeled CO₂ concentrations were sampled from the WRF output at the exact times and locations of the observations from these monitoring sites: Finokalia, Thissio and Helmos. Each site was assigned to the corresponding WRF grid cell in the finest nested domain encompassing its coordinates. Horizontally, the value from the nearest model grid point was selected, while vertically, the model level whose geometric height above sea level (ASL) was closest to the inlet elevation was used to represent the in-situ measurement. In total, 8,793, 8,602 and 6,690 hourly observations from Finokalia, Thissio and Helmos, respectively, were assimilated (24,085 data in total). For ground-based CO₂ observations, the total observation uncertainty was calculated as the quadrature sum of the instrumental measurement error and a site-dependent representation error reflecting local atmospheric variability.

$$\sigma_{obs} = \sqrt{\sigma_{measurement}^2 + \sigma_{rep}^2} \quad (7)$$

The representation error was estimated by the prior model-data residuals, with the instrumental measurement error removed in quadrature, and taken as the MAD (Median Absolute Deviation) scaled standard deviation ($1.4826 \times \text{MAD}$) of the resulting per-observation values: 1.48 ppm at Finokalia, 5.21 ppm at Thissio and 1.08 ppm at Helmos.

2.6. CO₂ satellite observations – assimilated data

To supplement the ground-based network and provide broader spatial coverage, we incorporated satellite-based CO₂ observations from the Orbiting Carbon Observatory-2 (OCO-2) mission. Launched by NASA in July 2014, OCO-2, provides global measurements (from 85°N to 85°S) of column-averaged dry-air mole fractions of CO₂ (XCO₂) with high precision and spatial resolution (Crisp et al., 2017). The satellite operates in a near-polar sun-synchronous orbit at an altitude of approximately 705 km, with a local equator crossing time of about 13:30 local time. OCO-2 carries three high-resolution grating spectrometers that measure reflected solar radiation in the near-infrared bands centered at 0.76 μm (O₂ A-band), 1.61 μm, and 2.06 μm, which are used to retrieve XCO₂ at a



nominal footprint of $1.29 \times 2.25 \text{ km}^2$. In this study, we used bias-corrected Level 2 XCO₂ retrievals (v10r),
 305 obtained from the NASA Goddard Earth Sciences Data and Information Services Center (GES DISC;
<https://doi.org/10.5067/E4E140XDMPO2>, last access: 29/05/2021), filtered for high-quality soundings based on
 the standard OCO-2 quality flag (quality flag = 0). For each OCO-2 sounding within the model domain, the two
 instantaneous WRF output times bracketing the satellite overpass were identified, and model fields were linearly
 interpolated in time to match the exact observation time. The model grid cell corresponding to the geolocation of
 310 each sounding was used for horizontal sampling. Model CO₂ vertical profiles were then integrated over pressure
 using the retrieval's averaging kernel and pressure weighting function, ensuring that the simulated XCO₂ reflected
 the same vertical sensitivity as the satellite retrieval. When the retrieval pressure range exceeded the model
 pressure range, the model profile was extended above or below the model boundaries using constant mixing ratios
 to maintain vertical continuity. For OCO-2 column observations, the observation error used in the assimilation
 315 was defined as the quadrature sum of the retrieval's reported uncertainty and of an additional term representing
 model/representation errors. Specifically, for each sounding, we set

$$\sigma_{obs} = \sqrt{\sigma_{retrieval}^2 + \sigma_{model}^2} \quad (8)$$

where $\sigma_{retrieval}$ is the per-sounding uncertainty provided with OCO-2 product (mean value of 0.50 ppm across all
 quality-filtered soundings used in this study), and σ_{model} is an additive error term set to 3 ppm for all soundings,
 320 giving a total σ_{obs} of approximately 3.04 ppm. The magnitude of σ_{model} was informed by the total single-sounding
 XCO₂ error over land reported by Connor et al. (2016), who find aerosol interference, spectroscopy, and
 instrument calibration to be the dominant sources of variable error, combining with measurement and smoothing
 to give a total error of approximately 1.5–3.5 ppm. We note that σ_{model} is treated here as a single, fixed value
 representing the combined contribution of unresolved retrieval error, transport error and representation error,
 325 rather than a magnitude independently derived for the latter two components. Future sensitivity tests varying σ_{model}
 would help quantify its influence on the posterior flux estimates. Because of the small swath width (10 km), the
 spatial resolution of 3 km² and the 16-day cycle (Crisp et al., 2017), OCO-2 provides spatially sparse coverage
 over the study region, with large gaps between overpass tracks (Gialesakis et al., 2021). Over the full year in 2019,
 659,864 soundings from OCO-2 were assimilated. This annual total covers d01 and the coverage within any
 330 individual region of Greece is limited (Figure S1).

2.7. FTIR measurements – validation data

Column-averaged XCO₂ measurements from a ground-based FTIR spectrometer located in Thessaloniki, are used
 to evaluate the model simulations. The instrument is an EM27/SUN portable Fourier Transform spectrometer,
 335 operated on the roof of the Laboratory of Atmospheric Physics of the Aristotle University of Thessaloniki
 (40.629°N, 22.947°E, ~60 m a.s.l.). The EM27/SUN measures direct solar radiation in the near-infrared spectral
 range, from which column-averaged dry-air mole fractions of CO₂ (XCO₂) are retrieved using the PROFAST
 retrieval algorithm (Mermigkas et al., 2021). Unlike the in-situ and OCO-2 observations, the FTIR dataset is not
 assimilated in the inversion and therefore provides an independent assessment of the model performance, allowing
 340 us to evaluate whether the emission adjustments derived from the assimilation are consistent with an observation
 type and location not used to constrain the inversion.



3. Results and discussion

First, we present the sensitivity of the model results to the various parameters used to set up the modeling framework, in order to test our choices of the optimal modeling set up. Next, we analyze the reference/optimal
 345 inversion output.

3.1 Impact of assimilation window

To investigate how the length of the assimilation window affects the inversion, we performed two simulations, one with a two-week window (test 5) and one with a three-week window (test 1) (Table 1). A back-trajectory climatology for Greece shows that long-range transport of air masses from the major anthropogenic source regions
 350 of western and northern Europe reaches the country within about four days (Katsoulis, 1999), and emissions originating within the domain influence the measurement sites on even shorter timescales. A comparable inversion over the whole of Europe using CTDAS adopted a 20-day window (Steiner et al., 2024). Given our smaller domain, we tested a 3-week configuration (test 1) against the 2-week case (test 5) to verify that the window was not too short. In the fixed-lag smoother, each weekly flux is re-estimated in successive cycles as it ages through
 355 the lag window, so we can quantify how much each additional week of constraint refines the estimate. Following the diagnostic of Peters et al. (2005), for every week we computed the squared correlation (r^2) between its estimate at each weekly lag and its final posterior value. In the 2-week (test 5) simulation the first estimate of the weekly fluxes already explains 95% of the variance of the final value, and in the 3-week configuration the first estimate explains 92%, rising to 95% after the second week. In both configurations, therefore, the bulk of each week's flux
 360 estimate is fixed at first exposure to the observations, with the later lags adding only a small remainder. This diagnostic is defined relative to each window's own final estimate and so does not by itself establish convergence to a window-independent value. The close agreement between the two cases provides more direct confirmation. The two configurations perform very similarly against the surface observations (Table S3). At Thissio, the 2-week simulation performs slightly better, with RMSE of 9.96 ppm and R^2 of 0.715 compared to 10.85 ppm and
 365 0.667 for the 3-week simulation, and a smaller mean bias (-2.33 vs -3.54 ppm). At Finokalia and Helmos, the differences are negligible, with nearly identical RMSE (1.15 vs 1.16 ppm and 1.08 vs 1.08 ppm respectively), R^2 and mean bias values for both simulations. The optimized January 2019 CO₂ emissions over Greece are also very similar between the two simulations, with 6.12 Mt for the 2-week and 6.07 Mt for the 3-week assimilation window (Table 1). Based on these results, no meaningful advantage was found in extending the assimilation window to 3
 370 weeks and given that a shorter window also reduces computational cost, the 2-week configuration has been retained for further simulations.

3.2 Impact of prior uncertainty

The prior uncertainty directly controls how strongly the assimilation system can correct the emission estimates. A larger uncertainty allows larger adjustments, while a tighter constraint keeps the posterior closer to the prior. To
 375 investigate this sensitivity, four simulations were performed with prior uncertainties of anthropogenic emissions of 10%, 25%, 50% and 100% (Test 2, Test 3, Test 4 and Test 5, respectively). The model statistics at the three stations for January 2019 are summarized in Table S4. At Thissio, the choice of prior uncertainty has a noticeable impact on the results. All posterior simulations substantially reduce the prior RMSE (from 19.58 ppm to 9.96) and mean bias (from -14.0 ppm to -1.70), confirming that the inversion successfully corrects the underestimation in
 380 the prior emissions over Athens. The simulations assuming 25% and 50% uncertainty in anthropogenic emissions



yield the best performance, with RMSE of 10.26 and 9.96 ppm and R^2 of 0.72 and 0.71, respectively, while the 100% configuration performs comparably (RMSE = 10.14), and the most restrictive 10% configuration performs the worst (RMSE = 11.22). This suggests that allowing little freedom prevents the system from fully correcting the prior bias, while more freedom does not lead to further improvement of RMSE. Mean bias, in contrast, continues to improve monotonically with increasing prior uncertainty, from -4.52 ppm at 10% to -2.87 ppm at 25%, -2.33 ppm at 50% and -1.70 at 100%. At Finokalia, all posterior simulations slightly improve upon the prior (RMSE = 1.20 ppm, R^2 = 0.70), with the 25% and 50% configurations performing slightly better (RMSE = 1.15 ppm, R^2 = 0.76), though differences across uncertainty levels are negligible. At Helmos, the posterior simulations show a slight degradation relative to the prior across all uncertainty levels, with RMSE increasing from 1.03 to 1.08–1.09 ppm and R^2 decreasing from 0.52 to 0.47–0.48. The sensitivity of the posterior emission scaling factors to the assumed prior uncertainty is further illustrated in Table S5 for January 2019. The sensitivity varies markedly across regions, reflecting differences in the magnitude of model–observation residuals at each station. In Athens, the scaling factors consistently exceed 1.0 across all uncertainty levels and all weeks, ranging from 1.08 to 2.78. This indicates a robust and stable upward correction of prior emissions regardless of the assumed prior uncertainty. At Helmos, the scaling factors vary from above to below 1.0 depending on the week and uncertainty level, ranging from 0.20 to 2.69, reflecting the greater variability in the model–observation differences at this mountain station across the simulated weeks and the freedom afforded by the looser prior uncertainties. At Finokalia, a similar pattern is observed, with scaling factors ranging from 0.07 to 2.82. These results collectively demonstrate that the choice of prior uncertainty has a smaller impact on well-constrained regions such as Athens than in regions where there are fewer observations to constrain the fluxes. Based on the station performance statistics, and particularly the best RMSE agreement at Thissio achieved with the 50% configuration, this value has been adopted for the base case simulation.

3.3. Impact of localization length and comparison to observations

The window and prior-uncertainty parameters selected above were determined using the 180 km localization length (test 5). In an ensemble Kalman smoother, the covariances between observations and state vector elements are estimated from a finite ensemble (here 150 members). Such estimates contain spurious correlations, especially between observations and distant prefectures, which can produce adjustments with no physical basis. Localization suppresses these by tapering the influence of each observation on a prefecture with distance. The appropriate localization length therefore balances two effects. Because CO_2 is long-lived, its enhancement caused by a local source can be carried over hundreds of kilometers within the assimilation window, which argues for a longer localization length. However, where orography and mesoscale circulation keep the signal close to its source, a long localization length erroneously allows observations to influence the emissions of distant prefectures that do not contribute to the measured signal, attributing noise to them. We therefore tested localization lengths of 180, 400 and 600 km for the entire year of 2019 (Figure S2). As expected, the largest uncertainty reductions are found in the greater Athens area, where the observations provide the strongest constraint on emissions. The spatial patterns are similar across the three simulations, with longer localization lengths giving consistently higher uncertainty reduction (Table S6). The domain mean uncertainty reduction increases with increasing localization length in essentially every region. The largest gains occur in regions only weakly constrained at 180 km, especially



at the Ionian islands (1.8% to 11.1%), Western Greece (10.2% to 18.7%), while the already well-constrained Attica region remains high throughout (83.6% to 87.1%). Longer localization therefore provides the expected stronger and spatially broader constraint. Table S7 summarizes the model performance against the surface in-situ measurements at the three stations. The posterior simulations reduce the RMSE at Thissio station from 14.51 ppm to around 8.5–8.7 ppm and improve R^2 from 0.55 to 0.77–0.78. At Finokalia and Helmos, the prior RMSE is much smaller (2.2 and 1.7 ppm, respectively), as are the differences between the prior and the posterior simulations, with marginal improvements across all localization lengths. No systematic relationship between localization length and station-level performance is apparent from these metrics alone. The evaluation against OCO-2 satellite retrievals and independent FTIR XCO₂ column measurements, shown in Figures 3a and 3b respectively, confirms this overall picture. FTIR statistics show a slight progressive improvement with increasing localization length, with RMSE decreasing from 1.70 ppm in the prior to 1.51 ppm for the 600 km simulation and R^2 increasing from 0.669 to 0.720, while the 180 km simulation performs comparably to the prior (RMSE 1.72 ppm, R^2 0.664). Against OCO-2, all simulations perform similarly, with RMSE values between 1.37 and 1.43 ppm and R^2 between 0.792 and 0.803, suggesting that the satellite retrievals do not have a strong impact on optimized emissions and do not clearly discriminate between the localization lengths tested, although a small improvement can be seen for the longest localization length. This limited sensitivity reflects the spatial distribution of the assimilated OCO-2 soundings. In 2019, a total of 659,864 soundings were assimilated across the d01 domain. However, only 119,490 (18.1%) fell within the d02 domain covering Greece, and these were further thinly distributed across the country's administrative regions (Figure S1). Because most of the assimilated soundings lie outside Greece, they exert limited influence on the regional state vector. The annual posterior total differs between the simulations (61.3 (±1.8) Mt at 180 km versus 55.0 (±1.8) at 400 km and 55.6 (±1.9) Mt at 600 km), but this difference in the annual sum does not reflect the magnitude of the underlying adjustments. In fact, the mean absolute monthly adjustment (the average over the twelve months of |posterior – prior|) increases with localization length, from 0.80 Mt for 180 km to 1.17 Mt for 400 km and to 1.45 Mt for 600 km (Table S8). Figure S3b, which depicts the monthly mean variation of the anthropogenic CO₂ emissions for the prior and the posterior for the three different localization lengths tested, reveals positive and negative adjustments to the prior emissions. It also shows that the results of the inversion for the shorter localization length of 180 km differ from the other posterior estimates providing smaller adjustments in March, September and October. At the longer localization lengths, the monthly adjustments are larger and of mixed sign, for example, strong positive corrections in summer (e.g. +2.9 Mt in July at 600 km) are offset by strong negative corrections in autumn (e.g. -2.5 Mt in September). Consequently, the posterior emissions show stronger seasonal variability than the prior emissions, which largely cancel out in the annual total, bringing it closer to the CAMS prior (55.0 Mt). The 180 km adjustments are smaller in magnitude but mostly positive, accumulating to a larger annual increase. The annual totals are therefore not a reliable indicator of the strength of the constraint, and do not by themselves favor a particular localization length. The clearest discriminating evidence comes from the independent FTIR data, against which the longer localization lengths perform marginally better, although the differences between configurations remain small. This seasonal structure of the adjustments is itself informative. The amplitude of the seasonal cycle of the optimized emissions grows with localization length (Table S8), and the monthly adjustments increasingly mirror the seasonal cycle of the net biospheric flux (Figure S3a). The correlation between the monthly anthropogenic adjustment and the net biospheric flux over Greece strengthens from $r = -0.17$ at 180 km to -0.40 at 400 km and -0.49 at 600 km. The



460 posterior minimum in September–October, when the inversion most strongly reduces the prior emissions at the
longer localization lengths, coincides with the autumn maximum in net biospheric CO₂ release. Since in the
present study, only the anthropogenic emissions carry uncertainty while the biospheric fluxes are assumed to be
accurate, this correlation suggests that part of the seasonal signal recovered at longer localization lengths is
biospheric in origin, although it is here attributed to the anthropogenic emissions, the only optimized sector. A
465 clean separation of the anthropogenic and biospheric seasonal signals would require the joint optimization of both
flux categories, which is work in progress. With an assimilation window of two weeks and a 50% uncertainty on
anthropogenic emissions, testing localization lengths (Tests 5 to 7, Table 1) shows that the 600 km configuration
(Test 7) provides the best agreement with the independent FTIR data, albeit by a small margin. These results
provide indications that the longer localization length is better suited for future work in this domain. Note however,
470 that the performance difference of the three test simulations (5 to 7) is very close, suggesting that data from
additional sites over Greece are needed to better constrain the emissions.

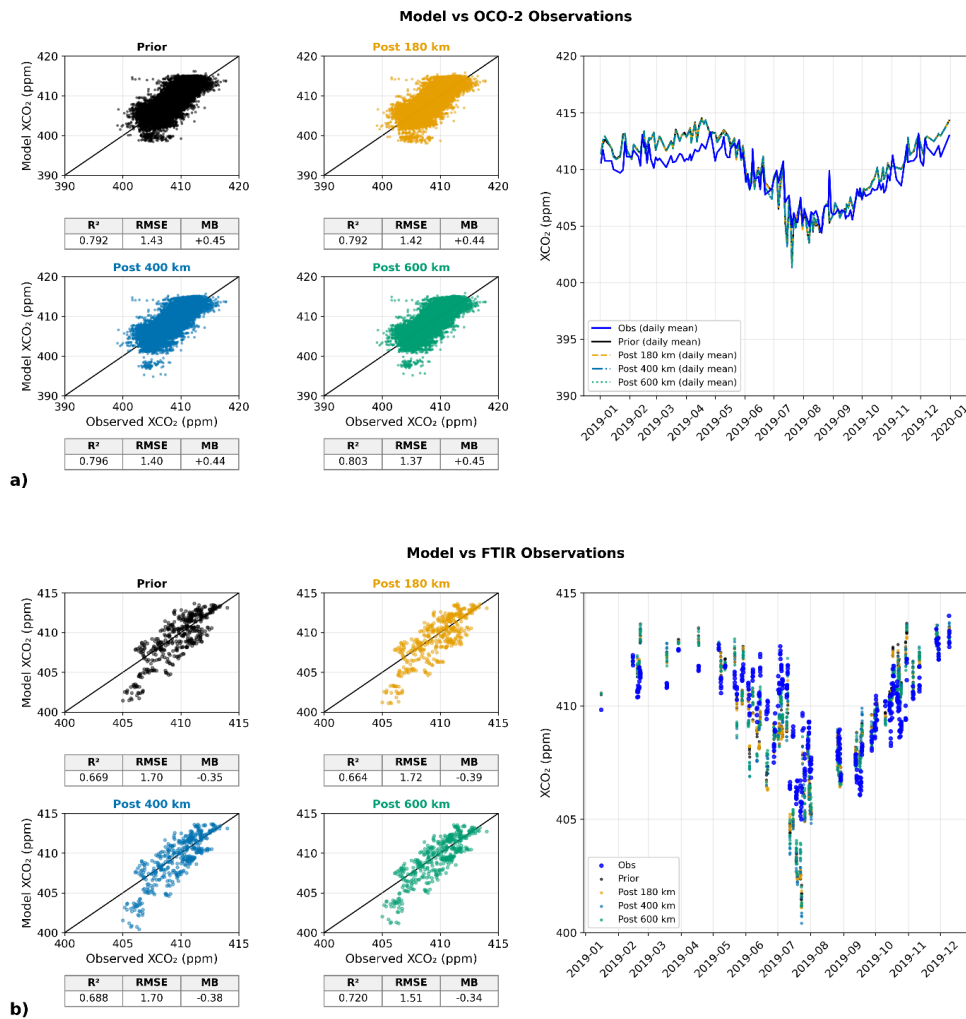


Figure 3: Simulated versus observed XCO₂ (ppm) over Greece for 2019, evaluated against (a) assimilated OCO-2 retrievals and (b) independent FTIR measurements at Thessaloniki. Left and middle panels: scatter plots of the collocated model versus the observed XCO₂ for the prior and posterior simulations (with localization lengths of 180, 400, and 600 km), showing the 1:1 line in black and the metrics (R^2 , RMSE, and mean bias) for all collocated soundings. Right panels: corresponding XCO₂ time series (observations in blue, simulations in other colors), shown as daily means for OCO-2 (a) and at individual measurement times for FTIR (b).

Guided by these results, we have chosen to use the 600 km localization length (Test 7) as the reference configuration here and compare the observed CO₂ concentrations at the three monitoring stations with WRF model results before and after optimization of the emissions. Figure 4a shows the daily mean observed CO₂ concentrations at the Thissio (urban), Finokalia (regional background), and Helmos (high-altitude background)



stations and compares them with the modeled concentrations using both the prior and the posterior emission
485 estimates for the whole of 2019 (Figure 4b).

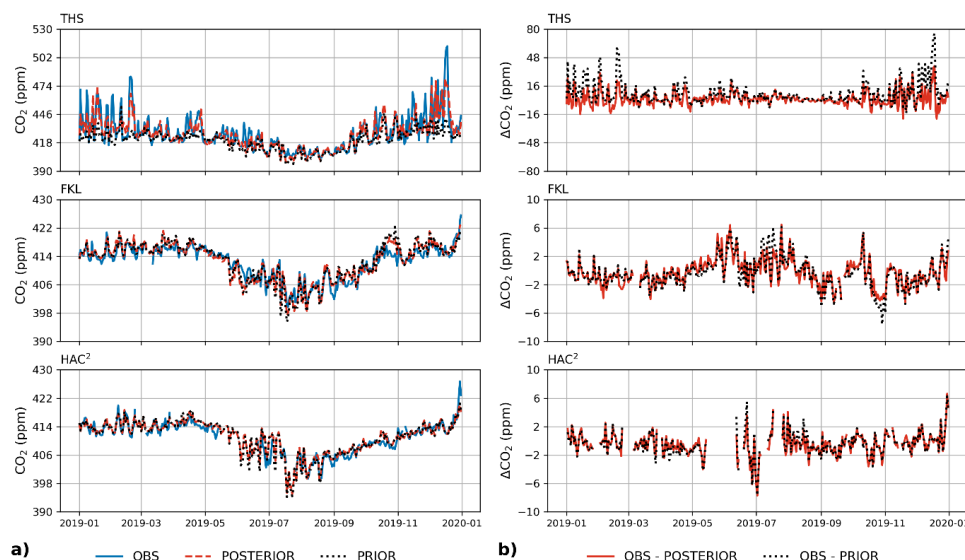


Figure 4: a) Daily average CO₂ concentrations at the Thissio (THS), Finokalia (FKL), and Helmos (HAC²)
stations in 2019. Observations (OBS; blue solid line) are compared with prior (black dotted line) and posterior
(Test 7; red dashed line) CTDAS-WRF simulations. b) Residuals between observed and simulated daily average
490 CO₂ concentrations (OBS – POSTERIOR, red solid line; OBS – PRIOR, black dotted line) at the THS, FKL,
and HAC² stations in 2019.

At Thissio station, the prior simulation showed a negative bias because it underestimates the magnitude of
observed peaks in the daily CO₂ timeseries. Assimilation of in situ and satellite observations leads to a substantial
improvement in the posterior simulation. Focusing on Figure 4b, significant improvements in Athens can be seen
495 from January to March and in November and December, while improvements at Finokalia can be seen during the
summer and in October. The model's performance at the Helmos station appears unaffected, both simulations
(prior and posterior) reproduce the observed day-to-day variability and seasonal cycle except in June, when the
discrepancy between model results and observations is reduced by about 2 ppm during a few days. These results
indicate that the inversion preserves free tropospheric CO₂ characteristics and does not introduce false large-scale
500 concentration offsets.

Overall, these results demonstrate that CO₂ inversions over regions with complex orography and strong mesoscale
variability, like those in the Mediterranean region, require careful tuning of the localization length. Longer
localization can strengthen and broaden observational constraints but it may also introduce cross-regional
transport and biospheric influences that are misattributed to anthropogenic emissions. Robust emission estimates
505 therefore depend on a balanced combination of appropriate localization, realistic mesoscale transport, and



sufficiently dense observations. These findings highlight the importance of using region-specific rather than universal localization lengths in mesoscale studies.

3.4. Quarterly patterns of CO₂ emission adjustments

To examine the spatial and temporal characteristics of the inversion results, we analyzed the anthropogenic CO₂ emissions adjustments through quarterly averages. The quarterly relative changes (%) in posterior anthropogenic CO₂ emissions for the reference (600 km) simulation with respect to the prior emission inventory are shown in Figure 5, with the corresponding results for all localization length sensitivity tests provided in Figure S4. Figure 5a shows the quarterly adjustments of the anthropogenic CO₂ emissions in Greece in 2019, grouped by the following months: January, February, March (JFM), April, May, June (AMJ), July, August, September (JAS) and October, November, December (OND). Across most of the year, the most prominent adjustments are found over the Athens metropolitan area, where the inversion yields positive corrections relative to the prior inventory, except during summer (JAS). These corrections are most distinct during the 1st quarter (JFM) and the last quarter (OND) of the year, when relative increases locally reach up to approximately 80-140% (Figure 5a). This behavior is consistent with the evaluation shown in Figure 4, which shows that the prior simulation substantially underestimates urban CO₂ enhancements during the cold months (December, January, February). During the 2nd quarter of the year (AMJ), the magnitude of the emission adjustments is low across most of the domain. In Athens, increases relative to the prior inventory persist during spring, although with reduced magnitude compared to winter. This suggests the prior emissions underestimate CO₂ sources in Athens beyond the cold months, with reduced intensity relative to winter. Note however that as discussed in section 2.1, these adjustments reflect total rather than anthropogenic CO₂ emissions. Outside of Attica, during the cold quarters most regions instead show reductions relative to the prior simulation, and this broad-scale pattern becomes more pronounced with increasing localization length (Figure S4). Potential inaccuracies in the simulation of the boundary layer height by the model could contribute to such discrepancies (Zhong et al., 2026), especially with our choice to assimilate data from all hours of the day in order to investigate the maximum information content of the sparse observation network.

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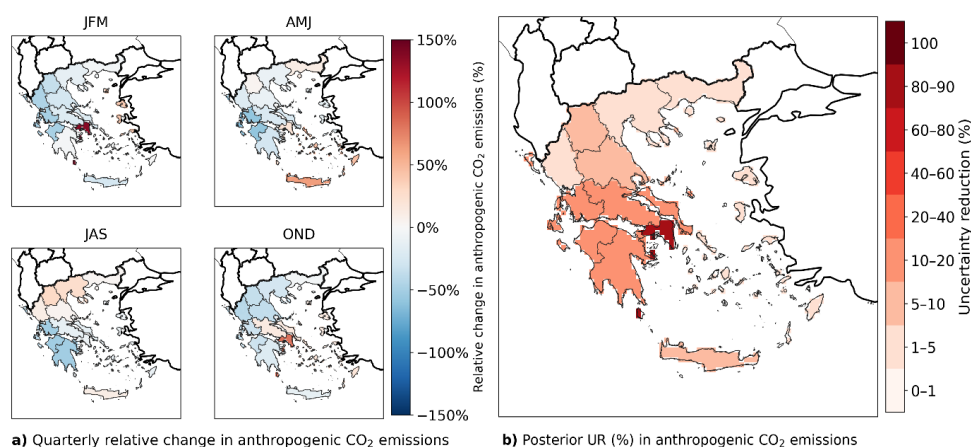


Figure 5: a) Quarterly mean relative change (%) in anthropogenic CO₂ emissions over Greece for 2019, inferred from the inversion with respect to prior emission inventory. Results are shown for JFM, AMJ, JAS, and OND and for the reference case (600 km). b) Posterior uncertainty reduction (%) of anthropogenic CO₂ emissions at the prefecture level, averaged over assimilation cycles.

Significant increases were calculated in Attica in most quarters and were more pronounced in JFM and OND when energy demand for heating is high, including domestic burning. However, during that period the biogenic flux of CO₂ is also high (Figure 1b and Figure S3). Although biogenic fluxes and fire emissions are treated independently in the model configuration, uncertainties in their magnitude, which are not considered in this study, can influence modeled CO₂ concentrations. As a result, the inversion may partially compensate for such mismatches through adjustments to anthropogenic emissions. Therefore, the calculated emission adjustments reflect changes in total CO₂ emissions, rather than anthropogenic emissions alone. During the 3rd quarter of the year (JAS), the emission adjustments are generally weaker and more spatially heterogeneous than during the cold months. Over Attica, the correction reverses sign in summer, shifting to a small reduction, in contrast to the strong positive corrections of the surrounding quarters. Furthermore, reductions in the emissions were calculated for Western Greece and the Peloponnese during most of the year.

3.5 Posterior uncertainty reduction

Figure 5b presents the prefecture pattern of posterior uncertainty reduction on annual fluxes calculated as explained in section 2.4. The results show a heterogeneous pattern, with large uncertainty reduction (exceeding 80%) in Athens, small reductions (10-20%) in the adjacent areas, while most other prefectures exhibit negligible reduction (< 10%). Although this reduction is affected by the choice of the localization length as shown in Supplementary Figure S2 and Table S6, the spatial pattern of the posterior uncertainty reduction is robust. It is dominated by the observation network and atmospheric transport. Observations in Thessio exert a pronounced, localized influence on nearby emission adjustment factors and therefore drive the largest uncertainty reductions. In contrast, measurements at Helmos, Finokalia and from satellites appear to have little influence on the results, as suggested by the smaller uncertainty reductions outside of Attica. Overall, the results indicate that, under the



current configuration, the inversion achieves its largest emission constraints over the Athens metropolitan area, where large and concentrated anthropogenic emissions lead to stronger and more distinct CO₂ enhancements. This demonstrates the capability of the inversion system to deliver regional emission optimization and suggests that national-scale constraints could be achieved with a denser and more spatially distributed observing network, in particular providing constraints to emissions in Central and Northern Greece.

3.6 Impact of the inversion on national CO₂ flux totals

To assess the impact of the inversion on total CO₂ fluxes over Greece, fluxes were spatially integrated to derive monthly and annual totals for the prior and posterior simulations. Figure S3b presents a comparison of the monthly anthropogenic emissions over Greece from the three posterior simulations (180, 400 and 600 km) with the prior (CAMS) emissions and the corresponding values from the Emissions Database for Global Atmospheric Research (EDGAR) inventory (<https://edgar.jrc.ec.europa.eu/>; last access 12.01.2026). The monthly and annual posterior anthropogenic CO₂ emissions for all simulations are summarized in Table S8. All three posterior simulations show pronounced month-to-month variability relative to the CAMS prior, but the amplitude of the seasonal adjustments increases with localization length. During the cold months (December–February), the total posterior emissions are higher than those of the CAMS prior in all configurations, with the largest positive adjustment in December. The 180 km simulation yields predominantly positive adjustments throughout the year, accumulating to an annual total above the prior. At the longer localization lengths, the monthly adjustments are larger but offsetting, so that they largely cancel in the annual sum (Sect. 3.3). The posterior emissions also differ from the EDGAR inventory in their seasonal distribution (Figure S3b). EDGAR exhibits a relatively weak seasonal cycle, whereas the posterior simulations, particularly at the longer localization lengths, show a much stronger seasonal amplitude. The posterior exceeds EDGAR during summer and in December, but falls below it during spring and autumn, so that the agreement with EDGAR varies considerably from month to month rather than holding uniformly across the year. Aggregating over the full year, the posterior anthropogenic CO₂ emissions for Greece are 61.3 ± 1.8 Mt at 180 km, 55.0 ± 1.8 Mt at 400 km and 55.6 ± 1.9 Mt at 600 km, compared to 55.0 Mt for the CAMS prior and 61.5 Mt for EDGAR. All three posterior estimates remain below the officially reported national anthropogenic CO₂ emissions of 65.7 Mt. Depending on the localization length, the annual total optimized emissions approach the EDGAR value (for localization length of 180km) or the CAMS prior emissions (for localization length of 600 km). As earlier noted, these adjustments cannot be attributed to anthropogenic emissions alone, since the biogenic and fire fluxes are not optimized. They have to be viewed as adjustments to the total emissions of CO₂ over Greece. For the reference simulation, the total CO₂ flux over Greece is 118.3 Mt yr⁻¹ (Figure 6), compared to 117.7 Mt yr⁻¹ for the prior and to 124 Mt yr⁻¹ for the 180 km localization length.

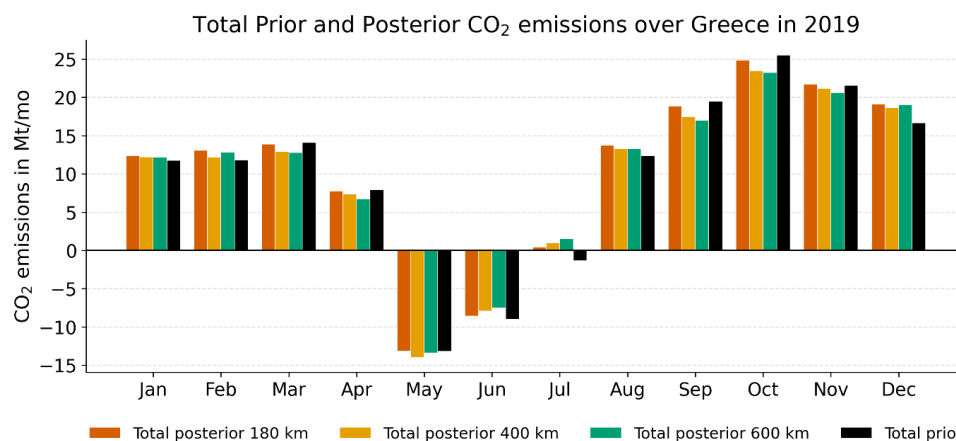


Figure 6: Total monthly CO₂ emissions over Greece in 2019, calculated as the sum of net vegetation flux, fire, and anthropogenic emissions. Shown are totals using the CAMS prior anthropogenic emissions (black) and the posterior anthropogenic emissions inferred from the inversion for the three localization lengths tested (180 km, orange; 400 km, yellow; 600 km, green).

4. Conclusions

Accurate, spatially resolved estimates of CO₂ emissions are needed to verify national inventories and support climate mitigation, yet high-resolution atmospheric inversion studies remain scarce for the eastern Mediterranean, a region characterized by complex orography and strong mesoscale circulation. We presented a regional inversion system coupling the CarbonTracker Data Assimilation Shell with the Weather Research and Forecasting model (CTDAS-WRF) to estimate CO₂ emissions over Greece for 2019. The atmospheric inversion framework was set-up, using complementary ground-based measurements from urban, regional background and high-altitude stations together with satellite observations from OCO-2. Sensitivity experiments demonstrated that the inversion is sensitive to the localization length and prior uncertainty of emissions, highlighting the need to tailor these parameters to the underlying transport regime. The system optimized CAMS anthropogenic CO₂ emissions in Greece at the prefecture level on a weekly timescale, while no uncertainty was attributed to all other sources of CO₂. Therefore, the model results reflect adjustments in the total emissions needed to explain the atmospheric observations rather than the anthropogenic emissions only. The optimized simulation shows a clearly improved agreement with observed CO₂ concentrations at the urban background Thissio station, where the mean model bias was reduced by 80% and the RMSE by about 40%. Independent FTIR observations in Thessaloniki also showed improved agreement with the optimized simulation, supporting the robustness of the inversion.

The spatial pattern of the optimized emission fluxes reveals a strong dependence on near-by observations and atmospheric transport. The largest uncertainty reduction occurs over Athens, where concentrated urban emissions create a clear and detectable atmospheric CO₂ signal. In contrast, Finokalia and Helmos stations, as well as satellite observations, have a smaller impact on regional surface emissions inversions. These stations mainly sample regional background air, which is largely influenced by long-range transport rather than by local or regional sources, and therefore provide only a weak constraint on Greek emissions. An upward correction of the prior



emission inventory over Athens is calculated for most of the year, most clearly expressed during winter, while during summer the correction reverses to a small reduction. Reductions of the prior emissions, accompanied by moderate uncertainty reductions, were calculated for the Peloponnese and Western Greece. Emission adjustments for the remaining prefectures are minor and do not significantly reduce emission uncertainty. These changes suggest substantial spatial and seasonal variability that is not fully captured by the prior inventory.

At the national scale, the annual optimized emissions vary with localization length, ranging from a value closer to EDGAR at 180 km to values close to the CAMS prior at 600 km, but remain below Greece's officially reported 2019 inventory of 65.7 Mt. For the reference configuration, the total CO₂ budget over Greece is 118.3 ± 1.9 Mt yr⁻¹, against 117.7 Mt yr⁻¹ for the prior. These annual adjustments to emissions should not be interpreted as direct estimates of anthropogenic emissions given that the biogenic and fire emissions were not optimized. The increase of the seasonal amplitude of optimized emissions with localization length, and the correlation between the monthly anthropogenic adjustment and the net biospheric flux strengthening (from $r = -0.17$ at 180 km to -0.49 at 600 km), indicate that part of the seasonal signal recovered at longer localization lengths is likely biospheric but is aliased onto the optimized anthropogenic sector.

These results have broader implications for atmospheric inversions in regions with complex orography and strong mesoscale variability, like the Mediterranean. In such environments, localization length cannot be treated as a universal parameter. Long localization length can improve the representation of long-range transport and broaden the observational constraint but may also transfer information between dynamically disconnected regions and increase the risk of attributing transport or biospheric variability to emissions. Conversely, overly short localization may underuse observations needed to constrain long-range CO₂ transport. The strong spatial variability in uncertainty reduction therefore emphasizes the importance of combining realistic mesoscale transport with dense and spatially distributed observations.

The results are subject to several limitations. First, the observational network is sparse: the strongest emission constraints are obtained close to Thissio, where large and spatially concentrated anthropogenic emissions generate intense, localized CO₂ enhancements that are well resolved by the observations. Expanded observational coverage would be needed for stronger emission constraints, especially near major CO₂ sources outside Athens. Second, the spatial coverage of OCO-2 data over Greece is limited. Third, biospheric and fire fluxes were kept fixed and the lateral boundary conditions were not optimized, so potential errors in these components may be attributed to anthropogenic emissions. Finally, all available ground-based observations were assimilated regardless of time of day, which increases the potential influence of errors in the transport model on the posterior fluxes (Geels et al., 2007). Future improvements may include selection of observations according to time of day: afternoon data at the lowland sites when the planetary boundary layer is well represented by the model and night-time data at the high-altitude Helmos station, when it samples free-tropospheric air rather than air transported up from below during the day (Munassar et al., 2023). Applying this inversion framework simultaneously to each one of the major category sources (biogenic flux and fires), together with optimization of the lateral boundary conditions, could also allow for a better understanding of the carbon cycle over Greece.

Overall, this study demonstrates the potential of regional atmospheric CTDAS-WRF inversions to improve CO₂ emission estimates, while showing that their success depends strongly on the interaction between observation



coverage and mesoscale transport. The results provide a basis for designing region-specific observing and inversion strategies in other Mediterranean and complex-terrain environments. Expanding observations near major emission sources and across dynamically distinct regions will be essential for achieving more robust and policy-relevant estimates of anthropogenic CO₂ emissions and for improving their use in monitoring progress
660 toward climate targets.

Code and data availability

CTDAS-WRF is available at <https://git.wur.nl/ctdas/CTDAS/-/tree/ctdas-wrf>. WRF is available from the official repository (<https://github.com/wrf-model/wrf>). Ground-based observational data and FTIR data are available
665 upon request from the data providers. The OCO-2 Level 2 Lite XCO₂ product (V10r) is available from the NASA GES DISC (<https://doi.org/10.5067/E4E140XDMPO2>).

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Competing interests

At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

Authorship contribution statement

NG performed all simulations, data analysis and wrote the initial manuscript, IE wrote the initial code for the
680 assimilation of the in-situ observations, FR did the coupling of CTDAS with WRF and wrote the code for the assimilation of the satellite observations, AB and NM provided CO₂ observations from Thissio, KE and MG from Helmos observatory, MM and DB provided the FTIR data from Thessaloniki. MK conceived the study and acquired the funding. MK, MV, FR and ND supervised the work and contributed to the manuscript writing. All authors commented on the manuscript.

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