



Validation of the CAMS regional services: concentrations above the surface

Status update for the period
December 2020 -February 2021

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Executive Summary

The Copernicus Atmosphere Monitoring Service (CAMS, <http://atmosphere.copernicus.eu>) is a component of the European Earth Observation programme Copernicus. The CAMS service consists of two major forecast and analysis systems. First, the CAMS global near-real time (NRT) service provides daily analyses and forecasts of reactive trace gases, greenhouse gases and aerosol concentrations, and is based on the ECMWF Integrated Forecast System (called CAMS-global in this document). Secondly, nine models in Europe, perform air quality forecasts and analyses on a daily basis, nested within CAMS-global. Based on these individual forecasts and analyses, an ensemble forecast of air quality over Europe is produced and disseminated by Météo-France (called ENSEMBLE or CAMS-regional below). The regional members use the global forecasting results as boundary conditions at the sides and top of the domain.

This document reports on two validation activities, namely

- an evaluation of the consistency between the global and regional modelling components of CAMS, focussing on the boundaries of the regional domain, and
- an evaluation of the regional ENSEMBLE and the nine individual models contributing to the ensemble with independent observations, focusing on the concentrations above the surface.

The current analysis includes ozone (O₃), nitrogen dioxide (NO₂), aerosol (PM₁₀/PM_{2.5}/AOD) and carbon monoxide (CO) forecasts covering the period up to February 2021.

The forecasts from the regional models were compared with the following set of observations:

- aerosol lidar observations from the EARLINET network;
- aerosol AOD observations from the AERONET network;
- IAGOS routine aircraft measurements of ozone and CO;
- ozone sonde profiles;
- MAX-DOAS NO₂ tropospheric columns;
- GOME-2/MetOp-A NO₂ satellite tropospheric column retrievals (IUP-UB v1.0 product);
- high-altitude ozone surface stations;
- CO and O₃ from GAW mountain stations; and
- CO observations from the MOPITT satellite instrument.

These observations are available to CAMS within one month after the observations were made.

This report is based on regional model data available for the months May 2016 to February 2021, with a focus on December 2020 – February 2021 (DJF2021). The report is updated every 3 months. The main results are summarised below, focusing on the performance of the regional ensemble. Detailed results, also for the individual models, are presented in sections 3 to 11, and each of these sections starts with a summary of the main results. Model specific findings are summarised in section 12. The last upgrade implemented in CAMS-global was on 6 October 2020, while the last upgrade for the CAMS regional models happened on 25 November 2020, implementing new anthropogenic emissions and making available an additional pollen species.

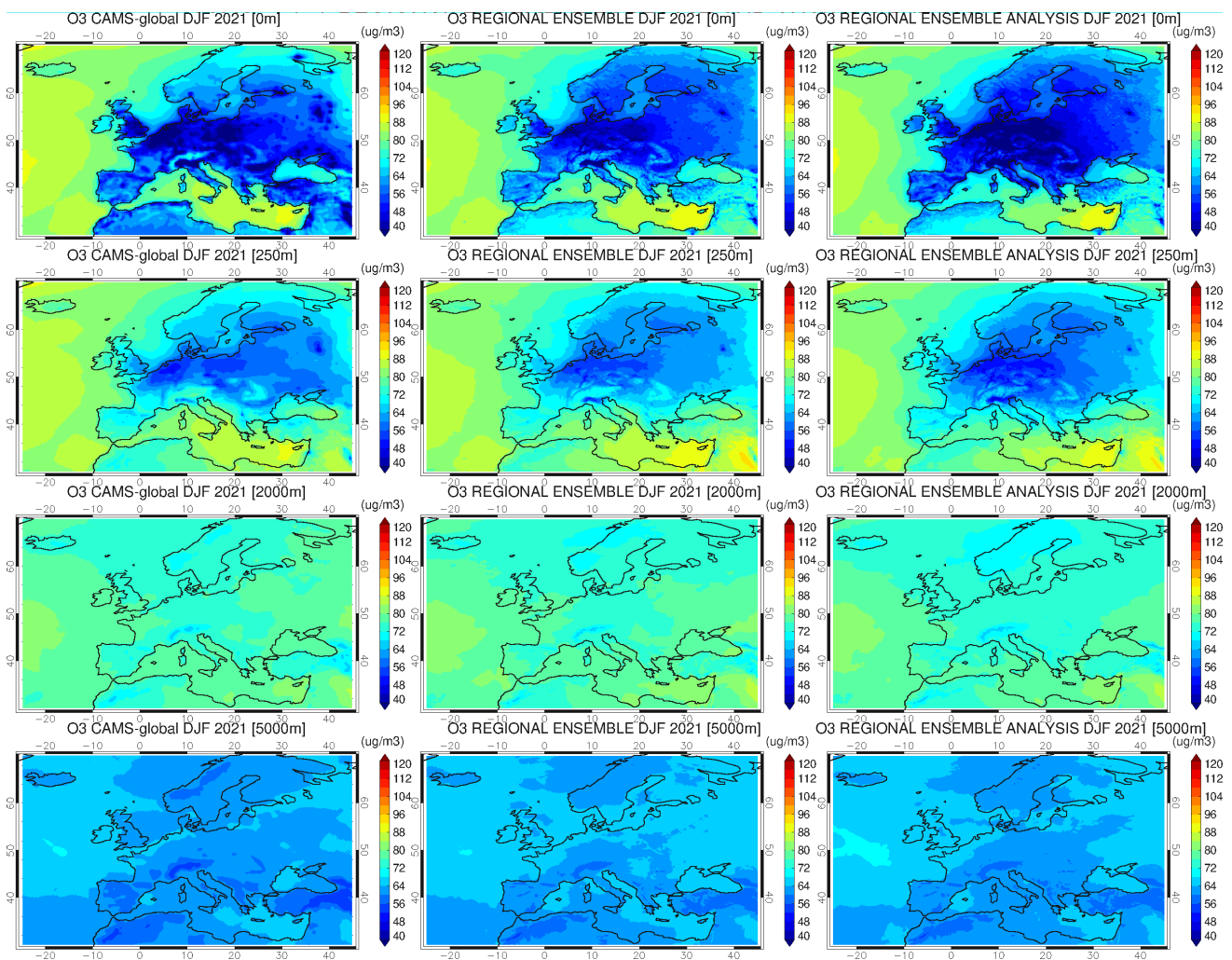


Figure S.1. CAMS global ozone forecast for day 1 (left), CAMS regional ENSEMBLE ozone forecast for day 1 (middle), CAMS regional ENSEMBLE ozone analysis (right). From top to bottom: 0, 250, 2000, 5000m altitude level. The results are averaged over the December 2020 to February 2021 period.

General conclusions for the regional ENSEMBLE

The comparison of the European regional CAMS ENSEMBLE air quality forecasts and analyses against above-surface observations of O_3 , NO_2 , CO for the period up to the 1st of March 2021 demonstrates that overall, the biases observed are small, often within the uncertainty of the validation approach while temporal correlations for ozone and CO are reasonable. Performance of the ENSEMBLE analysis product is found to be generally superior to that of the ENSEMBLE forecasts. The ENSEMBLE performs generally better than any of the individual models for ozone, NO_2 and CO , showing the strength of the ensemble approach adopted in CAMS.

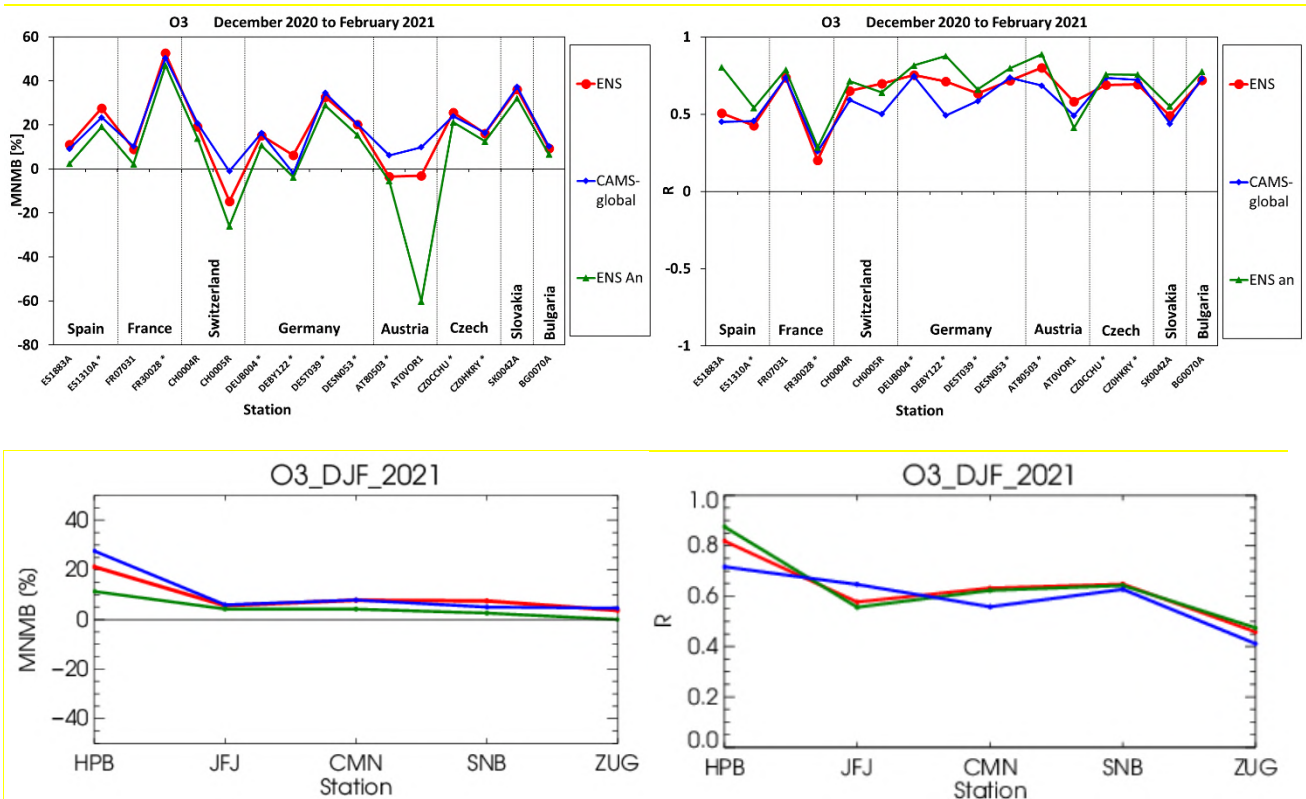


Figure S.2. Normalised bias (left) and correlation coefficient (right) for ozone for the high-altitude (above 1 km) EEA Air Quality e-reporting stations (top) and the 5 high-altitude European GAW stations (bottom). Lines represent ENSEMBLE forecast (solid red), ENSEMBLE analysis (solid green) and CAMS-global system (blue) for December 2020 - February 2021. The horizontal axis is the station identifier referring to Hohenpeissenberg (HPB), Jungfrauoch (JFJ), Monte Cimone (CMN), Sonnblick (SNB) and Zugspitze (ZUG).

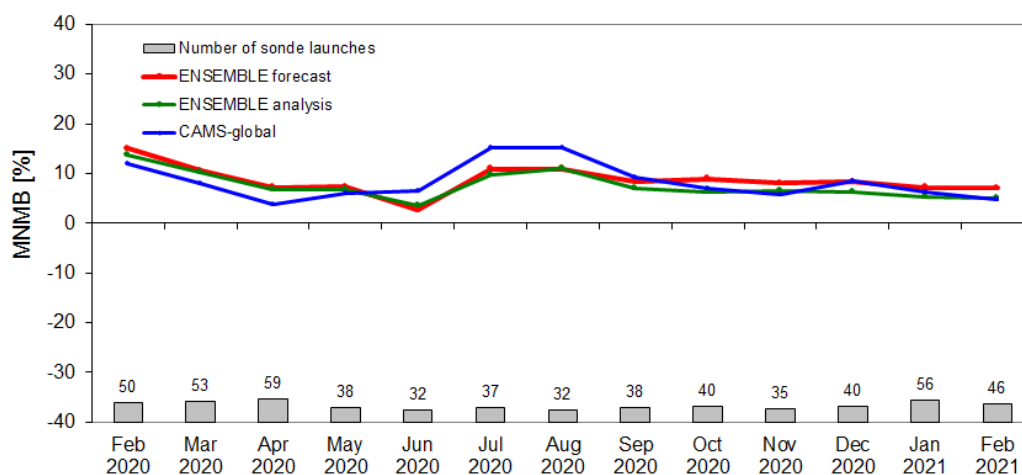


Figure S.3 Modified normalised mean bias (MNMB) against ozone sondes for the regional ENSEMBLE forecasts (red) and analyses (green) from February 2020 to February 2021 (horizontal axis). Ozone was averaged over the lower-middle free troposphere region, 500 hPa < p < 850 hPa.

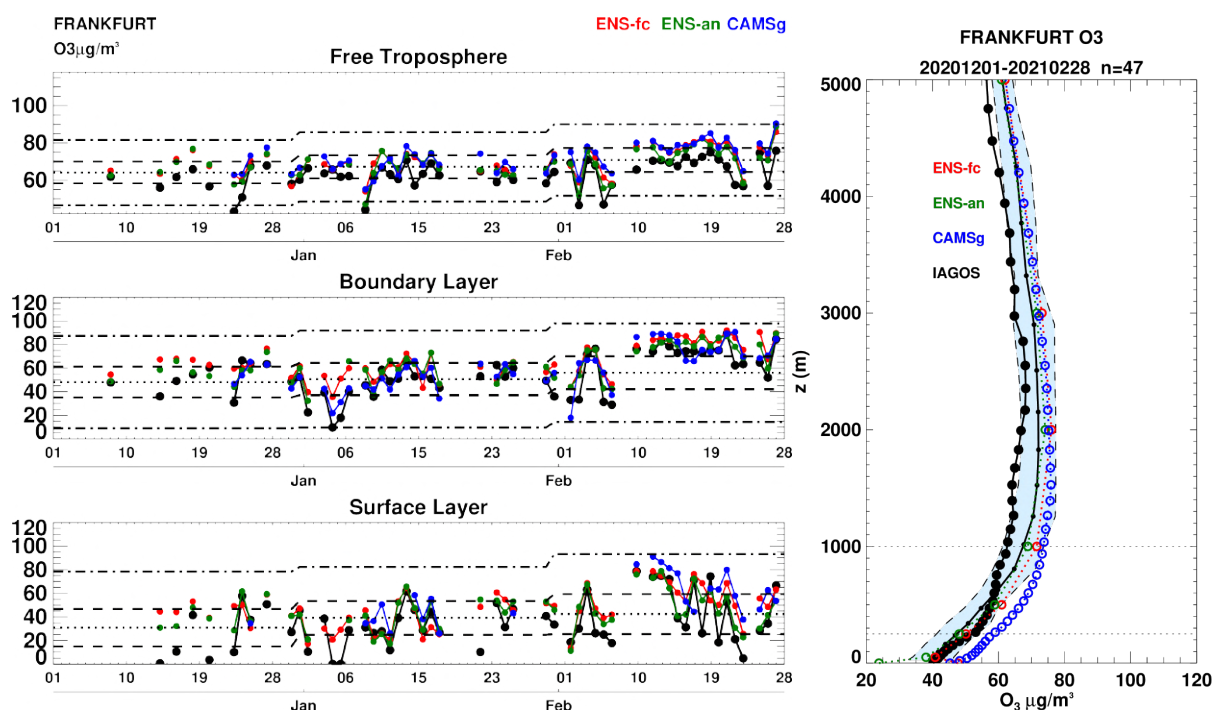


Figure S.4. Left: Daily time series of ozone at Frankfurt for December 2020 to February 2021. The black dotted line is the monthly mean of the observations over the period 2003-2016 (IAGOS/MOZAIC, Level 2 data), the black dashed line shows 1 standard deviation from the monthly mean and the black dotted-dashed line shows 3 standard deviations from the monthly mean. Right: Mean profile of ozone at Frankfurt for the period December 2020 - February 2021. The shaded area indicates the range of the mean climatology of the observations plus/minus one standard deviation during the same period for all years between 2003 and 2016 (IAGOS/MOZAIC, level 2 data). In both panels, IAGOS observations are shown in black, the regional ENSEMBLE and associated analysis are shown in red and green respectively, and the global o-suite is shown in blue.

Ozone

At the boundaries of the regional domain, the ENSEMBLE agrees well with CAMS-global, indicating that the implementation of the boundary conditions was done properly. Over the full domain there is a good match between the global and regional CAMS analyses and forecasts between 1 and 5 km altitude. These results are similar to previous quarters.

The differences between the global and regional systems reveal themselves in the boundary layer and at the surface over land, as expected. A comparison of the regional analysis product with the regional day 1 forecast (Fig. S.1) shows some differences between the regional ENSEMBLE forecast and analysis, with the analysis generally having lower concentrations near the surface.

For high altitude stations, Fig. S.2, mostly an overestimation between up to 50% is observed for the ENSEMBLE first day forecast, while comparison against ozone sondes shows a milder overestimation in the 5-20% range (also Fig. S.3). Time correlations at high-altitude and GAW stations range between 0.2 and 0.83 during this period with generally improved figures for the ENSEMBLE analyses as compared to the ENSEMBLE forecast, especially for the high-altitude stations.

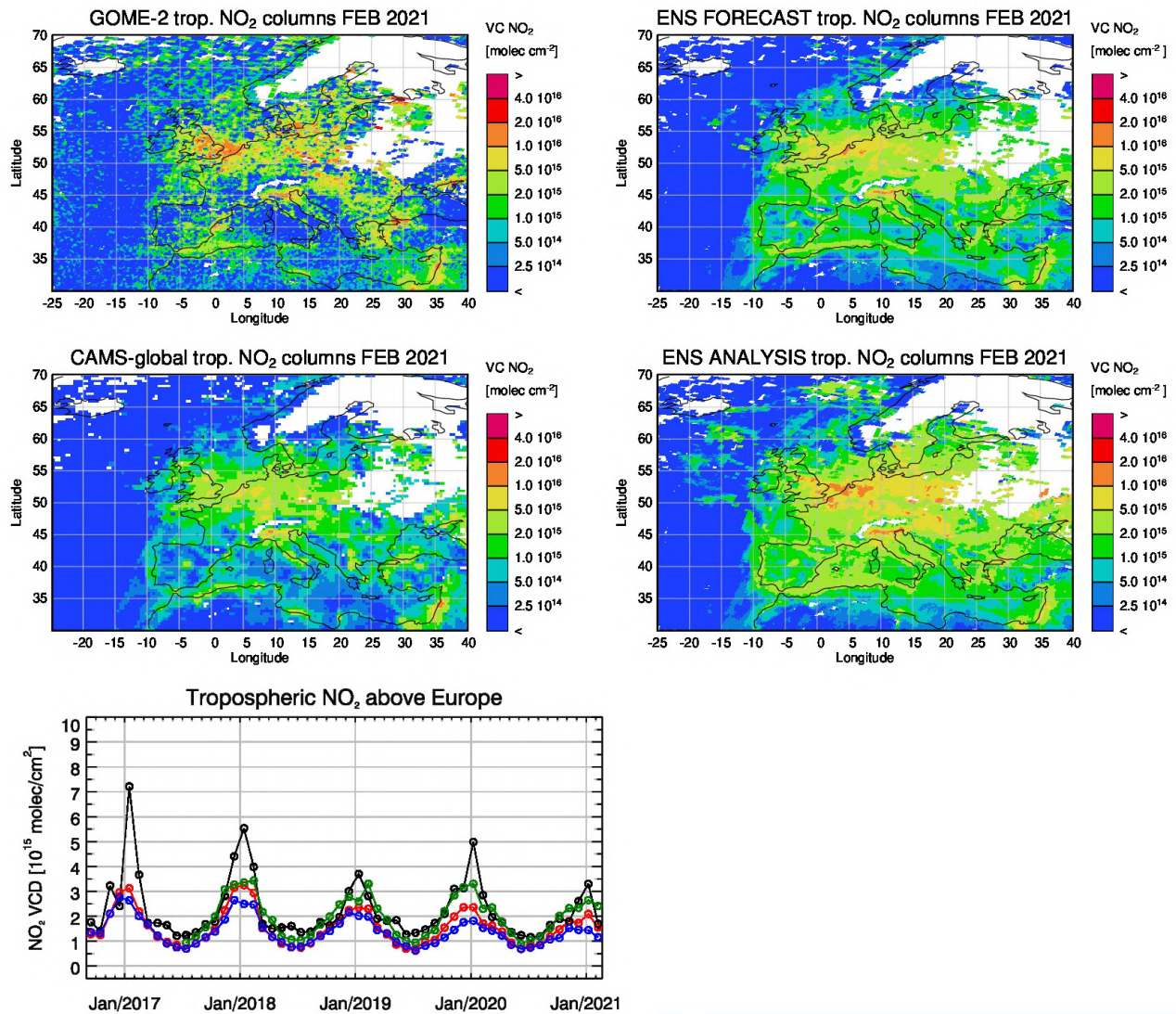


Figure S.5: Maps of satellite-retrieved and model-simulated tropospheric NO₂ columns [molecules cm⁻²] for February 2021 for GOME-2 (top left), regional ENSEMBLE forecasts (top right) CAMS-global forecasts (middle left) and regional ENSEMBLE analyses (middle right). The panel at the bottom shows corresponding time series of average tropospheric NO₂ columns [10¹⁵ molecules cm⁻²] from GOME-2 (black), regional ENSEMBLE forecasts (red), CAMS-global forecasts (blue) and regional ENSEMBLE analyses (green). GOME-2 data were gridded to regional model resolution (i.e., 0.1° x 0.1°). Model data were treated with the same reference sector (25°W - 20°E) subtraction approach as the satellite data and linearly interpolated to the satellite overpass time (9:30 LT).

Comparisons with IAGOS aircraft observations at Frankfurt, see Fig. S.4, reveals that ozone is mostly well reproduced by both the regional ensemble and CAMS-global in the low troposphere while larger overestimations are obtained in the free troposphere for all models.

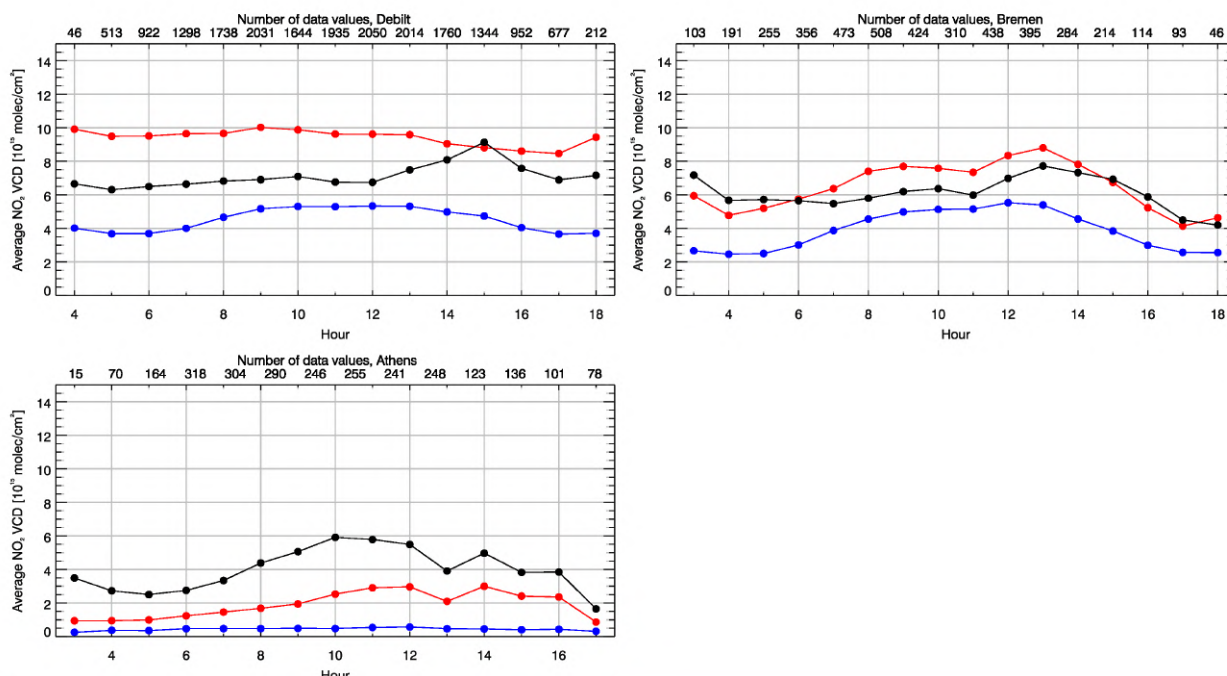


Figure S.6: Diurnal cycles (averages over hourly bins) of tropospheric NO₂ VCDs [10^{15} molec. cm⁻²] from MAX-DOAS and models for De Bilt (the Netherlands) (top left), Bremen (Germany) top right and Athens (Greece) (bottom). The coloured lines show (black) MAX-DOAS retrievals, (red) regional ENSEMBLE forecasts and (blue) CAMS-global. Period: June 2020 – February 2021.

Nitrogen dioxide (NO₂)

The overall spatial distribution of tropospheric NO₂ as observed from space by GOME-2A is reproduced by the ensemble during DJF2021 (see Fig. S.5) with better results by the analysis compared to the forecast. As described in previous reports, winter values over European emission hotspots simulated by the regional ensemble analysis, forecast and CAMS-global show smaller values than GOME-2A, this effect is however not so pronounced this year. Values over European emission hotspots are better represented by the regional ensemble analysis and forecasts than by CAMS-global.

Systematic uncertainties in the retrievals (on average on the order of 20% – 30% over polluted regions) depend on the season, with winter values and especially January in mid and high latitudes normally associated with larger error margins. Conclusions may differ for comparisons to other satellite NO₂ products (e.g. TEMIS GOME-2A, <http://www.temis.nl>). We note that GOME-2A observations were assimilated in the CAMS-global from 26 June 2018 until 5 May 2020. This was, however, a different retrieval product than the one used for validation reported here (University of Bremen retrieval).

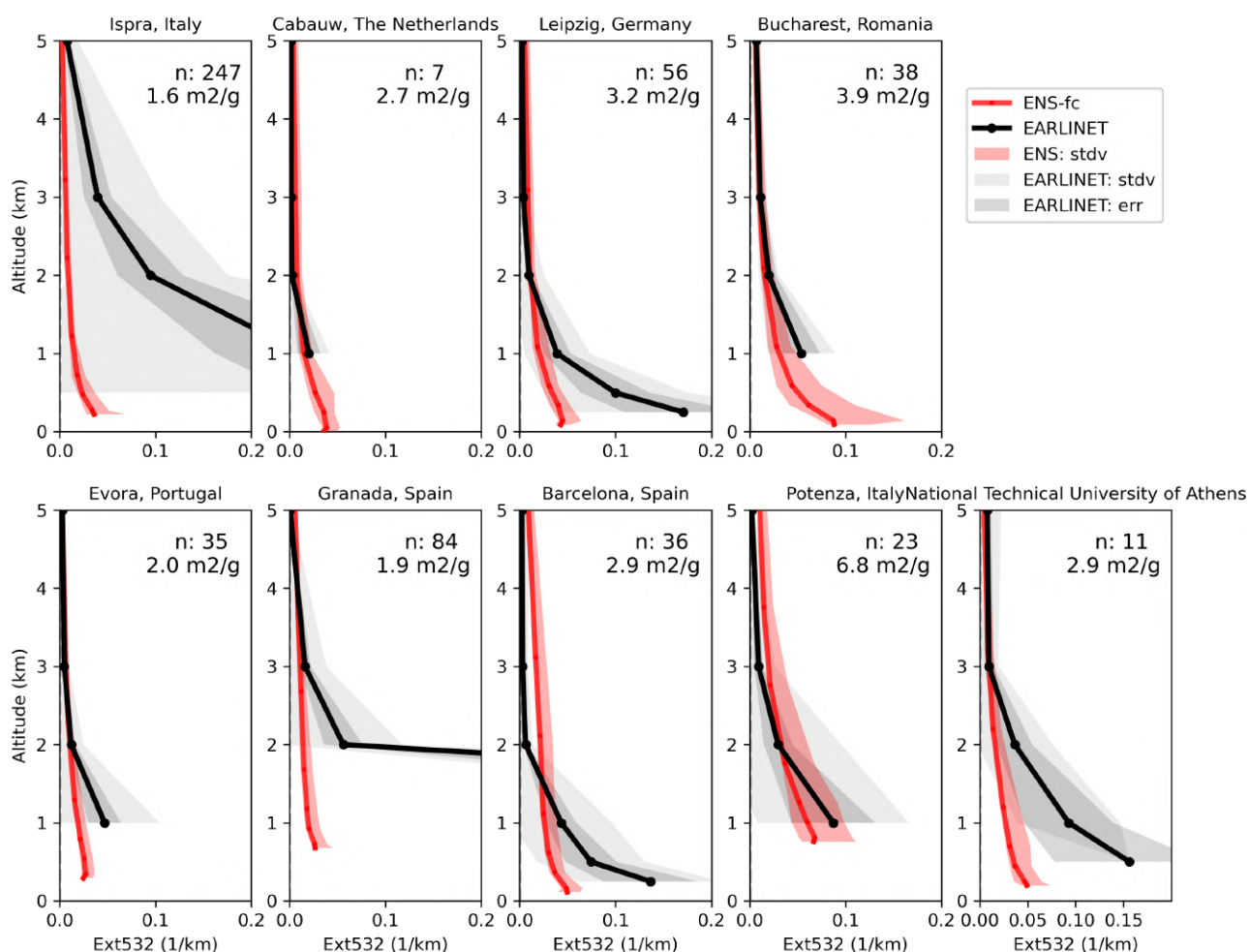


Figure S.7. Extinction profiles December 2020 - February 2021 derived from the ENSEMBLE forecast mass concentration profiles (red envelope) and from EARLINET (climatology) backscatter profiles (grey envelope: lidar ratio uncertainty, light grey: including sampling error). “n: XX means number of individual EARLINET profiles assembled (December - February 2006-2018). The EMC used for the calculation of the extinction from the concentration profiles is indicated for each station below the number of EARLINET profiles “n” used for the calculation of the climatology.

Comparisons to ground-based remote-sensing MAX-DOAS retrievals at three different European stations (see Figure S.6) for the time period of DJF2021 show that regional ENSEMBLE forecasts are closer to the urban station observations of Bremen and Athens than CAMS-global, mainly attributed to the difference in spatial resolution. For De Bilt, CAMS-global performs better than the regional ENSEMBLE. However, this may change with the next reports when more months of data will be included. The performance of simulations for diurnal cycles of tropospheric NO₂ columns depends on the location, but generally shows a moderate performance for the ENSEMBLE products for Bremen and Athens.

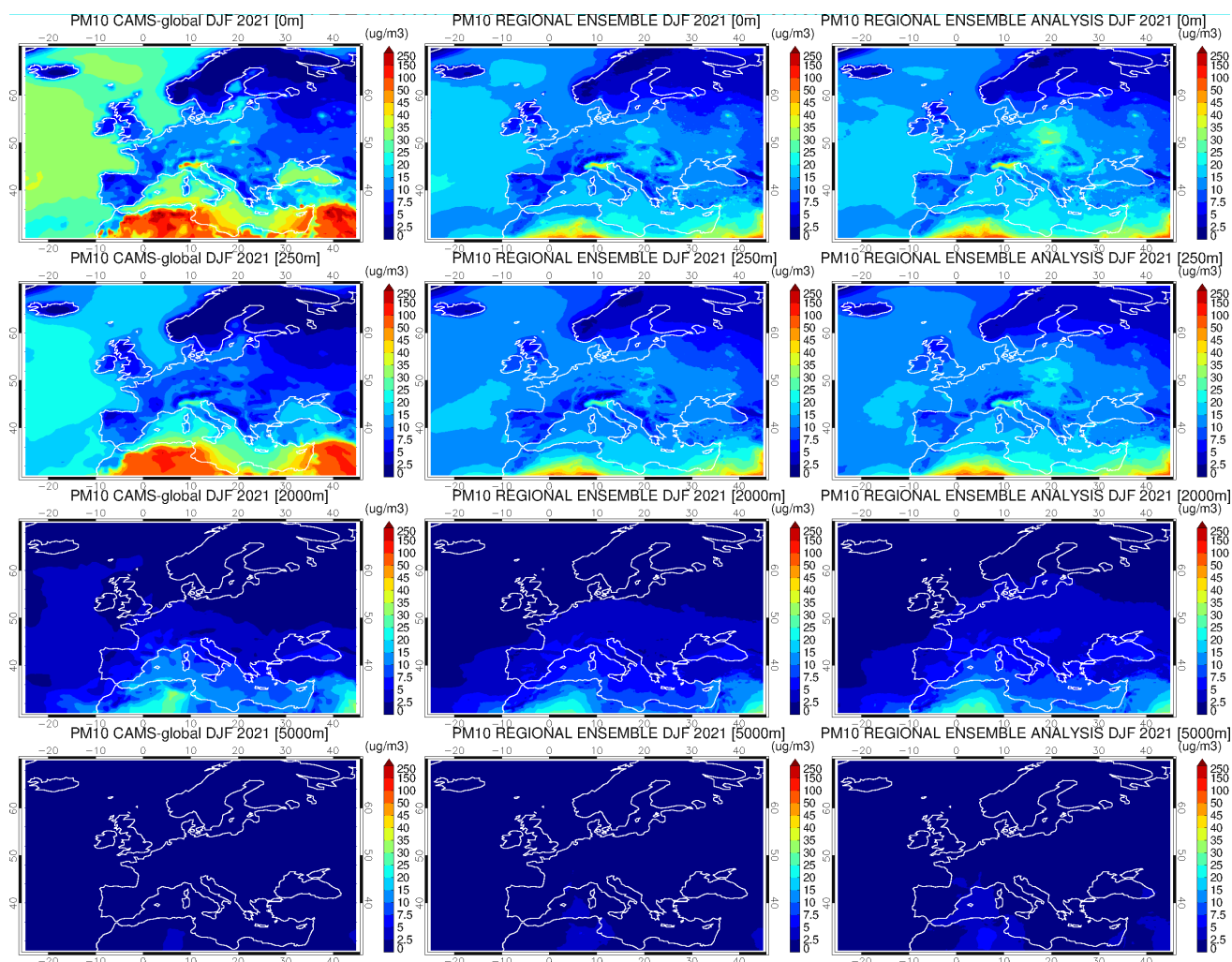


Figure S.8. CAMS global PM10 forecast for day 1 (left), CAMS regional ENSEMBLE PM10 forecast for day 1 (middle), CAMS regional PM10 analysis (right). From top to bottom: 0, 250, 2000, 5000m altitude level. Period: December 2020 to February 2021.

Aerosol / PM

The regional models are compared with EARLINET climatological lidar profiles for the same season (data from 2006-2020), Fig. S.7. The standard dissemination of CAMS-regional forecasts does not include information on composition, size, and humidity growth of the aerosol in the models. This introduces considerable uncertainty to the PM derived extinction, which conservatively spans a factor 10 for absolute extinction values. Relative differences among nearby stations and the form of extinction profiles are more certain. The order of magnitude in extinction is similar between the ENSEMBLE and the lidar profiles, with Ispra, Italy, close to the Alps and Granada, Spain being the most notable exceptions. The decrease in extinction with height seems to be generally steeper in the observations.

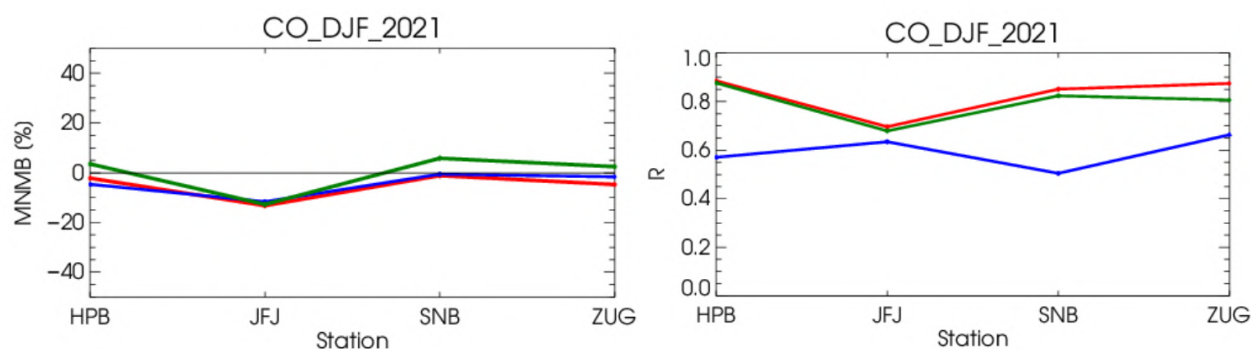


Figure S.9. MNMBs [%] (top) and correlation coefficients (bottom) for CO regional ENSEMBLE forecasts (red), analyses (green) and CAMS-global (blue) compared to observations at GAW stations. Period: December 2020 - February 2021.

The PM₁₀ concentrations in the regional ENSEMBLE forecasts and analyses are similar, but larger PM values are observed for the analysis over areas of Central and Western Europe at the surface level, see Figure S.8. CAMS-global shows higher dust loads over the Atlantic and the Mediterranean, especially at the lower levels. The agreement between CAMS-global and the ENSEMBLE products is better for PM₁₀ and PM_{2.5} in the upper layers compared to the lower layers, as expected.

Carbon monoxide (CO)

Comparison at the GAW stations reveal biases between -14% and 8% for the analysis and between -14% and ~0% for the forecast, while temporal correlations coefficients are between 0.68 and 0.88 for the analysis and between 0.69 and 0.89 for the forecast (Fig. S.9).

Comparisons with MOPITT CO satellite observations (version 8, Fig. S.10) data also reveal good agreement with the observations, exhibiting a slight negative bias within 10% (with some regional and temporal exceptions). The modelling data captured well both, relatively high CO values over the northern and middle parts of domain and relatively low values over the southern part. Analysis data are similar to the forecast data, showing slightly smaller negative bias.

General conclusions for the individual regional models

Regional models in general appear to have efficiently implemented the use of global CAMS boundary conditions and provide 3D fields that are comparable to those of the global model.

For ozone, bias and correlation at GAW and EEA e-reporting high altitude surface stations show a small spread among most of the models and an improved performance of the analysis compared to the forecast. The comparisons with ozone sondes in the lower free troposphere show similar results.

For NO₂, the validation with MAX-DOAS shows a sizeable spread between individual models, especially when considering the diurnal cycles. However, most of the simulated values probably fall within the uncertainty range of MAX-DOAS retrievals. The overall spatial distribution of tropospheric NO₂ as seen by GOME-2A/MetOp-A is reproduced by the regional models, but values over central European emission hotspots are underestimated by most models.

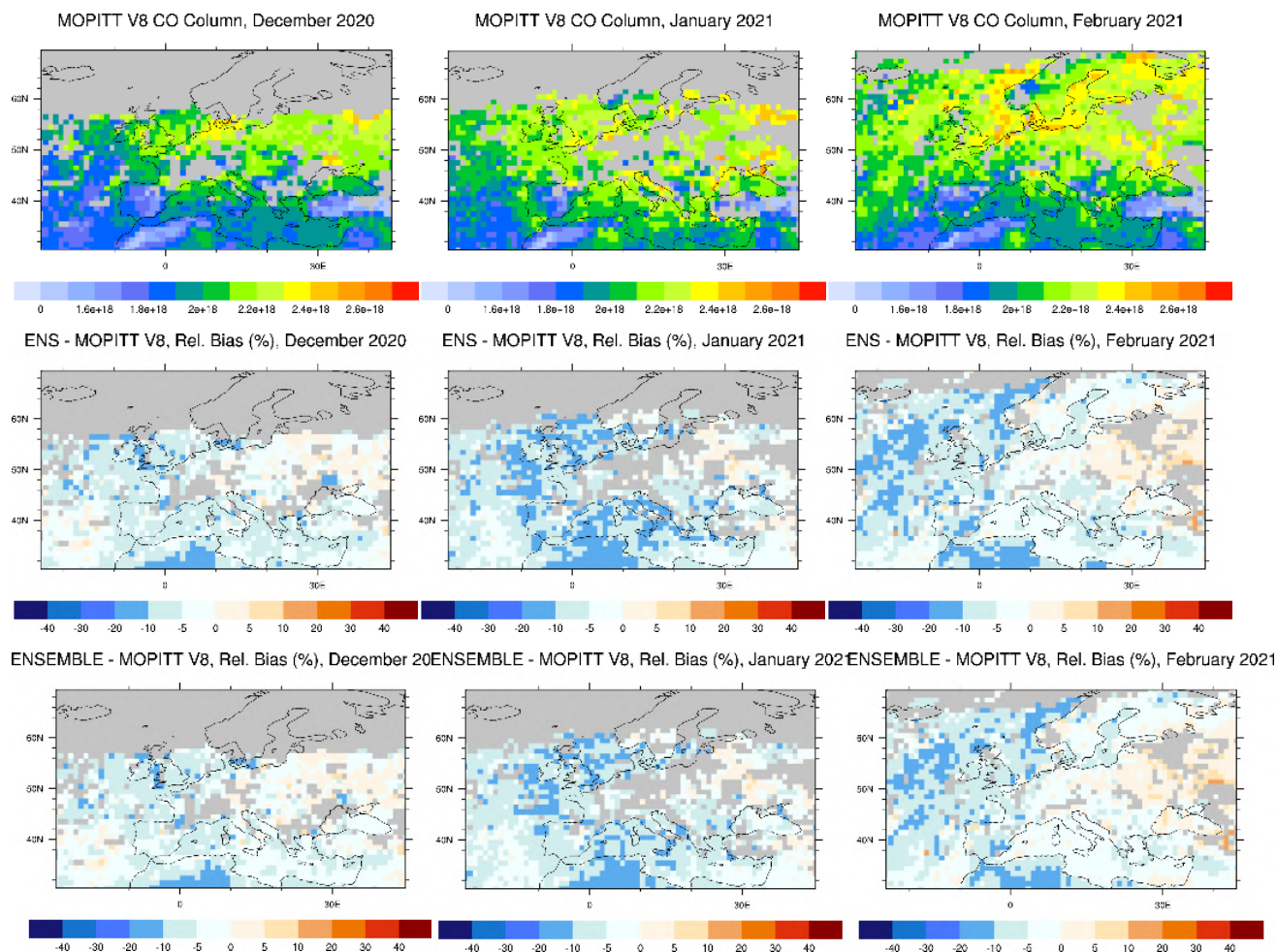


Figure S.10. CO total column for MOPITT v8 satellite retrievals (top row, in molecules/cm²), relative difference between the regional ENSEMBLE forecasts and MOPITT (middle row) and regional ENSEMBLE analyses and MOPITT (bottom row) for December 2020 (left column), January (middle column) and February 2021 (right column). Grey colour indicates missing values.

For CO, comparisons at GAW mountain stations show a slightly negative bias and small model spread for the forecasts. For the analyses, the bias is positive for some models and the spread is higher. The latter is especially true for the correlation coefficient. Comparisons with MOPITT retrievals reveal a small underestimation over the whole domain for most of the models.

For PM, observations from the EARLINET and AERONET networks reveal that the order of magnitude in extinction is similar between the models and the lidar profiles at most locations, while the decrease in extinction with height seems to be generally steeper in the observations.



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1 Introduction

The Copernicus Atmosphere Monitoring Service (CAMS, <https://atmosphere.copernicus.eu>) is a component of the European Earth Observation programme Copernicus. The CAMS near-real time services consist of daily analysis and forecasts with the IFS system with data assimilation of trace gas concentrations and aerosol properties. The global modelling system is also used to provide the boundary conditions for an ensemble of more detailed regional air quality models that are used to zoom in on the European domain and produce 4-day forecasts of air quality. The regional forecasting service provides daily 4-days forecasts of the main air quality species and analyses of the day before, based on the results from 9 state-of-the-art atmospheric chemistry models. The ensemble represents the median of these 9 model forecasts.

Routine validation of the regional models against surface observations from the European member states (EEA Air Quality e-reporting) is provided for each model individually as well as the ENSEMBLE in separate quarterly validation reports. Validation reports of the CAMS regional products are available in the following portal:

<https://atmosphere.copernicus.eu/regional-air-quality-production-systems>.

This web page provides access to the quarterly reports on the daily analyses and forecast activities and verification of the regional ENSEMBLE. An overview of the regional air quality forecasting system is provided by Marécal et al (2015).

Validation reports (e.g. Wagner et al., 2020) for the CAMS global products are available at <https://atmosphere.copernicus.eu/node/325>, including the evaluation on Earth's troposphere, stratosphere, aerosols and greenhouse gases, with state-of-the art observational datasets (GAW, IAGOS, MOPPIT, EMEP, GOME-2A, OMPS-LP, BASCOE, AERONET etc.). A published overview on the validation of reactive gases and aerosols in the global analysis and forecast system can be found in Eskes et al (2015). A validation study of the global surface ozone reanalysis for Europe is provided by Katragkou et al (2015).

Details of the various observational datasets can be found in Eskes et al. (2021), "Observations characterisation and validation methods document", also available at:

<https://atmosphere.copernicus.eu/eqa-reports-global-services>.

This document presents an evaluation of the concentrations above the surface as modelled by the set of the 9 individual regional models and the ensemble median derived from those 9 individual forecasts and analyses, as well as the consistency between the global and regional modelling systems of CAMS.



2 Regional and global CAMS forecasting systems

2.1 Regional models

The European Air Quality products are provided from the Copernicus Atmosphere Monitoring Service (<http://atmosphere.copernicus.eu/>). These data are available in NetCDF or Grib-Edition2 format. The files are available each day through ftp protocol from the Météo-France server (<ftp.cnrm-game-meteo.fr>). Since the beginning of February 2020, the data are also available through the Atmosphere Data store (<https://ads.atmosphere.copernicus.eu>), although the reliability of data retrievals from was relatively low during this quarter. The products are available in Near Real Time (NRT) for four forecast days, following the protocol below:

- Each day 96h model forecasts and 24h analyses for the previous day are provided with hourly resolution.
- Products are available at eight vertical height levels: surface, 50, 250, 500, 1000, 2000, 3000, 5000 meters.
- The pollutants are O₃, CO, NO₂, SO₂, PM_{2.5}, PM₁₀, NO, NH₃, NMVOC, PANs, dust aerosols (fraction below 10µm), secondary inorganic aerosols (fraction below 2.5µm), birch, grass, olive, ragweed and alder pollen. Since the upgrade of February 4th, 2020 three new aerosol species (PM from wildfires, EC from fossil fuels and EC from wood burning) are also available.
- The regional datasets cover the longitudes 335.05°E to 44.95°E every 0.1°, and latitudes 71.95°N – 30.05°N also at 0.1° resolution (~10km).
- The forecasts until the 48th hour are available before 7:30 UTC
- The forecasts for the 49-96th hour are available before 9:30 UTC
- The analyses are provided before 12:00 UTC
- Since the November 2020 upgrade, the regional models make use of the CAMS-REG-AP_v4_2 emissions dataset (reference year: 2017).

The NRT forecast and analysis regional air quality data are available for the nine air quality models and their ensemble median (CAMS-regional or ENSEMBLE):

- MOCAGE model (MFM)
- LOTOS-EUROS model (KNM)
- EMEP MSC-W model (EMP)
- MATCH model (SMH)
- EURAD-IM model (RIU)
- CHIMERE model (CHI)
- SILAM model (FMI)
- DEHM model (DEM)
- GEM-AQ model (GEM)

Before October 16th, 2019 the regional ensemble was calculated based on 7 regional models (except for DEHM and GEM-AQ). This is the third quarterly report which explicitly includes results and analysis from those two new models.



Every evening, a full download of the 96h forecasts and 24h analyses fields to KNMI at full resolution is performed. These fields are co-located to the set of surface stations used, and this largely reduced datasets is shared with all validation partners.

Documentation about the regional models may be found at the address <https://atmosphere.copernicus.eu/regional-air-quality-production-systems>. For the purposes of this report however, it's useful to indicate what kinds of observations are actively assimilated in each model (Table 2.1).

Table 2.1: Surface and other observations that are actively assimilated in regional models.













Model	Method	Surface	Other
CHIMERE	Kriging	O ₃ , NO ₂ , PM10, PM2.5	
EMEP	Intermittent 3D-var	O ₃ , SO ₂ , NO ₂ , CO, PM10, PM2.5	OMI NO ₂
EURAD	Intermittent 3D-var	O ₃ , SO ₂ , NO ₂ , PM10, PM2.5	OMI and MetOp/GOME-2 NO ₂ and SO ₂ , MOPITT and IASI CO
LOTOS-EUROS	Ensemble Kalman filter	O ₃ , NO ₂ , PM10, PM2.5	OMI NO ₂
MATCH	Intermittent 3D-var	O ₃ , NO ₂ , CO, PM10, PM2.5	
MOCAGE	3D-var	O ₃ , NO ₂ , PM10	
SILAM	Intermittent 3D-var	O ₃ , NO ₂ , CO, PM10, PM2.5	
DEHM	Optimal interpolation	O ₃ , NO ₂	
GEM-AQ	Optimal interpolation	O ₃ , NO ₂ , CO, SO ₂ , PM10, PM2.5	

Validation reports of the CAMS regional products are available in the following portal: <https://atmosphere.copernicus.eu/regional-services>.

Whenever possible, in this report, models follow the naming and colour scheme of Table 2.2.



Table 2.2: Naming and colour scheme followed throughout this report.

<i>Model</i>	<i>Short model name</i>	<i>Colour name</i>	<i>Colour</i>
CAMS-global	CAMSG	Blue	
ENSEMBLE forecast	ENS-fc	Red	
ENSEMBLE analysis	ENS-an	Green	
CHIMERE	CHIM	Yellow	
EMEP	EMEP	Brown	
EURAD	EURAD	Cyan	
LOTOS-EUROS	LOTOS	Purple	
MATCH	MATCH	Grey	
MOCAGE	MOCA	Pink	
SILAM	SILAM	Orange	
DEHM	DEHM	Fuchsia	
GEM-AQ	GEMAQ	Light Green	



2.2 Global CAMS system based on the ECMWF IFS model

The CAMS-global operational assimilation/forecast system consists of the IFS-CB05 chemistry combined with the MACC aerosol model. The chemistry is described in Flemming et al. (2015); aerosol is described by the bulk aerosol scheme (Morcrette et al., 2009). Dissemination of CAMS-global forecasts is twice a day, at about 10:00 and 22:00UTC. The forecast length is 120 h. Users can get access at <https://atmosphere.copernicus.eu/catalogue>. Table 2.3 provides information on the satellite data used in CAMS-global.

Table 2.3: Satellite retrievals of reactive gases and aerosol optical depth that are actively assimilated in CAMS-global.

Instrument	Satellite	Provider	Version	Type	Status
MLS	AURA	NASA	V4	O ₃ Profiles	20130107 -
OMI	AURA	NASA	V883	O ₃ Total column	20090901 -
GOME-2	Metop-A	Eumetsat	GDP 4.8	O ₃ Total column	20131007 - 20181231
GOME-2	Metop-B	Eumetsat	GDP 4.8	O ₃ Total column	20140512 -
GOME-2	Metop-C	Eumetsat	GDP 4.9	O ₃ Total column	20200505 -
SBUV-2	NOAA-19	NOAA	V8	O ₃ 21 layer profiles	20121007 - 20201005
OMPS	Suomi-NPP	NOAA / EUMETSAT		O ₃ Profiles	20170124 - 20190409, 20201006
OMPS	NOAA-20	NOAA / EUMETSAT		O ₃ 13-layer profile	20201006-20201215
TROPOMI	Sentinel-5P	ESA		O ₃ column	20181204-
IASI	MetOp-A	LATMOS/ULB EUMETSAT	-	CO Total column	20090901 - 20180621 20180622 - 20191118
IASI	MetOp-B	LATMOS/ULB EUMETSAT	-	CO Total column	20140918 - 20180621 20180622 -
IASI	MetOp-C	EUMETSAT		CO Total column	20191119-
MOPITT	TERRA	NCAR	V5-TIR V7-TIR V7-TIR Lance V8-TIR	CO Total column	20130129 - 20160124 - 20180626 20180626 20190702
OMI	AURA	KNMI	DOMINO V2.0	NO ₂ Tropospheric column	20120705 -
GOME-2	METOP A	Eumetsat	GDP 4.8	SO ₂ Total column	20150902-20200504
GOME-2	METOP B	Eumetsat	GDP 4.8	SO ₂ Total column	20150902-
GOME-2	METOP C	Eumetsat	GDP 4.9	SO ₂ Total column	20200505-
TROPOMI	Sentinel-5P	ESA		SO ₂ Total column	20201005-
MODIS	AQUA / TERRA	NASA	Col. 6 Deep Blue	Aerosol total optical depth, fire radiative power	20090901 - 20150902 -



PMAp	METOP-A METOP-B	EUMETSAT		AOD	20170124 - 20170926 -
GOME-2	METOP A	Eumetsat	GDP 4.8	NO ₂ Tropospheric column	20180624-20200504
GOME-2	METOP B	Eumetsat	GDP 4.8	NO ₂ Tropospheric column	20180626-
GOME-2	METOP C	Eumetsat	GDP 4.9	NO ₂ Tropospheric column	20200505-

The most recent upgrade relevant to this report, was the upgrade to cycle 47R1 which took place on 6 October 2020 and involved a number of changes, including an update in anthropogenic emissions, improved parametrisations sea salt and desert emissions, an improved stratospheric ozone scheme and the assimilation of volcanic SO₂ from the Sentinel-5p/TROPOMI.

A detailed changelog and the corresponding validation reports for this last upgrade can be found on the following page:

<https://atmosphere.copernicus.eu/cycle-47r1>.

Upgrade and version information is available here:

<https://atmosphere.copernicus.eu/changes-cams-global-production-system>.

Documentation on the global system can be found here:

<https://atmosphere.copernicus.eu/global-production-system>.



3 Consistency between the global and regional modelling components of CAMS

3.1 Summary

This chapter reports on the consistency between the global and regional modelling components of CAMS, and the impact of global CAMS boundary conditions on regional forecasts. The current evaluation includes ozone (O₃) Carbon Monoxide (CO), Nitrogen Dioxide (NO₂) and aerosol (PM₁₀/PM_{2.5}) forecasts covering the period from December 2020 to February 2021.

Global and regional ensemble forecasts: The two forecast products compare well, some differences are seen in aerosol (PM₁₀), which is higher in CAMS-global over the southern boundary and over the Atlantic in altitudes < 2Km.

Regional forecast variability: The regional ensemble members exhibit the expected regional variability for O₃, CO, NO₂ and particulate matter. Minor deviations (compared to CAMS-global and the majority of the regional models) are generally noted, with MOCAGE having the most distinct PM₁₀ behavior over western (sea salt) and southern (dust) Europe. CHIMERE has a problematic O₃/CO boundary condition implementation.

Daily time series. There is a good temporal agreement for O₃, CO, NO₂ and PM₁₀ between the regional ensemble members and CAMS-global, SILAM and GEMAQ only exhibit occasionally higher PM peaks than CAMS-global.

Regional forecast and analysis: The comparison of regional analyses and forecasts shows strong model and species dependence.

3.2 Methodology for the comparisons of CAMS-global and CAMS-regional

Operational download

The daily regional CAMS forecasts are retrieved on a daily basis by the AUTH-server. This includes the 3-hourly (0,3,6,9 etc.) regional forecast data (ensemble members and regional ENSEMBLE) for all provided species at all vertical layers for the 5 forecast days extracted from the Météo-France ftp server, and the 3-hourly (0, 3, 6, 9 etc.) global forecast data for 5 forecast days extracted from the ECMWF CAMS ftp server

Methodology of global-regional comparison

The following methodology is used to a) convert CAMS-global species from mass mixing ratio (kg/kg) to concentration ($\mu\text{g}/\text{m}^3$) and, b) extract CAMS-global species concentrations from the vertical levels that lie closest to the regional height levels.

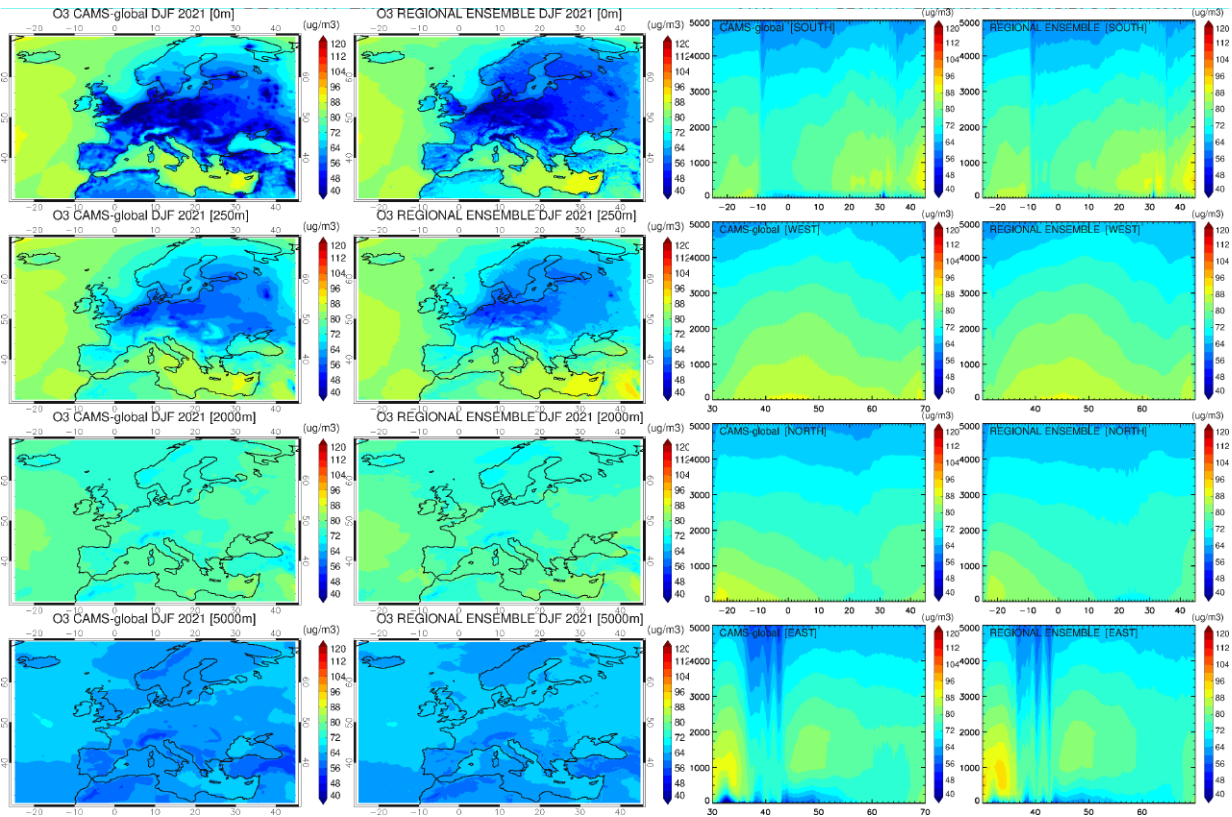


Figure 3.1. Left: Mean global and regional ENSEMBLE forecast ozone fields for four different vertical layers (0, 250, 2000, 5000 m) for DJF2021. Right: Cross sections for the same period of the global and regional ENSEMBLE ozone boundaries (south, west, north, east).

The following parameters are used from the CAMS-global model: hybrid layer coefficients; temperature, surface pressure; "GEMS" ozone; carbon monoxide; "aermr01-11" and "aermr16-18" (aerosol species, kg/kg). Data from the first 78 vertical layers (from the surface) are used.

The thickness of each vertical layer Δz (m) is calculated:

$$\Delta z_k = \frac{R * T_k}{M_{air} * g} * \ln \left(\frac{p_{i_{k+1}}}{p_{i_k}} \right) \quad (E. 1)$$

where $R=8.314 \text{ J/mol}\cdot\text{K}$ the gas constant, T the temperature at vertical layer midpoint, $M_{air}=28.97\cdot 10^{-3} \text{ kg/mol}$ the molecular weight of air and $g=9.8 \text{ m/s}^2$ the gravity acceleration.

The mass-mixing ratio (kg/kg) for ozone (go_3) and carbon monoxide (CO) is initially provided. Conversion from mass mixing ratio (kg/kg) to concentration ($\mu\text{g/m}^3$) is performed using the following approach:

$$\rho_{O_3} = m_{mr_{O_3}} * \left(\frac{p_m * M_{air}}{R * T} \right) \quad (E. 2)$$

where ρ_{O_3} is the ozone concentration (kg/m^3) and $m_{mr_{O_3}}$ the ozone mass mixing ratio (kg/kg). The expression inside the parentheses in E.2 corresponds to the air concentration (kg/m^3). The same approach is also used for the CO unit conversion from kg/kg to $\mu\text{g/m}^3$.

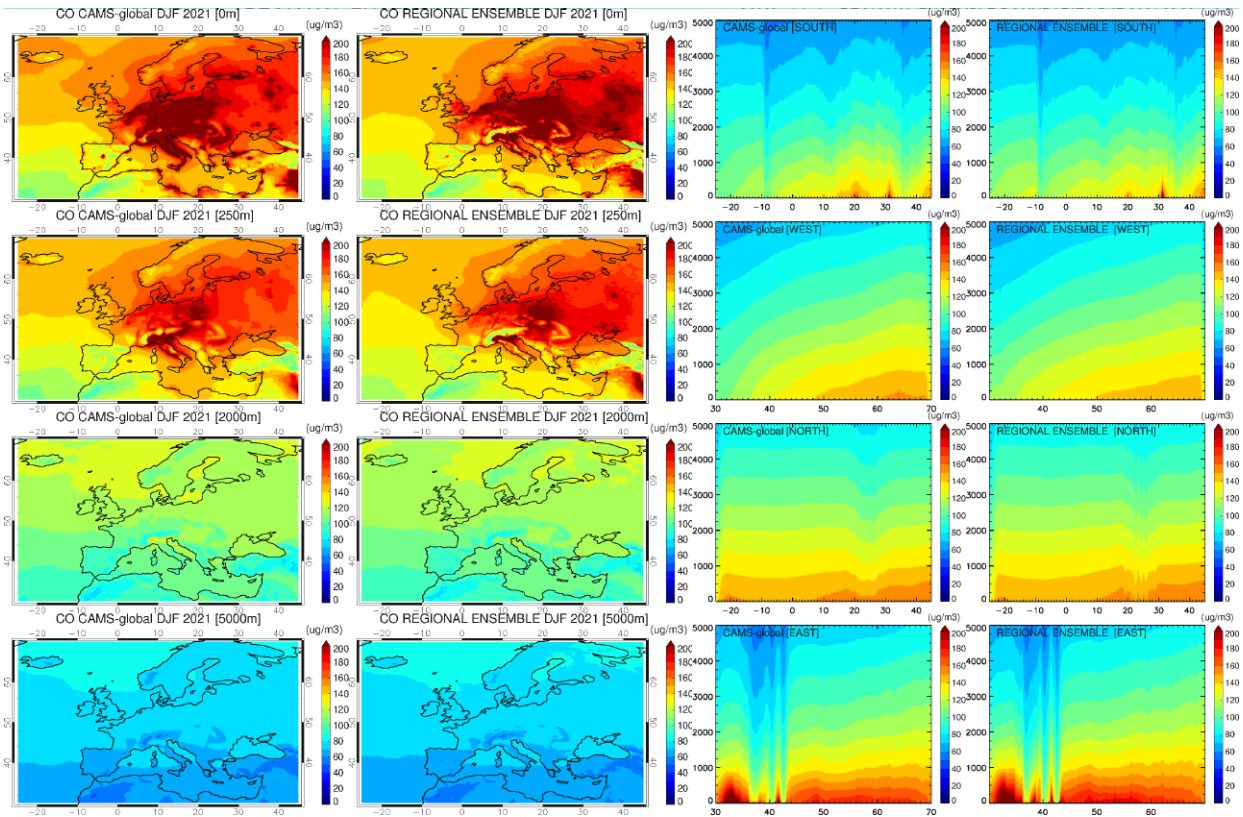


Figure 3.2. Left: Mean global and regional ENSEMBLE forecast CO fields for four different vertical layers (0, 250, 2000, 5000 m) for DJF2021. Right: Cross sections for the same period of the global and regional ENSEMBLE CO boundaries (south, west, north, east).

The mass mixing ratio for all aerosol species (see Table 3.1) is initially provided. The PM10 and PM2P5 species are converted to $\mu\text{g}/\text{m}^3$ as follows:

$$\rho_{\text{PM10}} = \left[\frac{\text{aermr01}}{4.3} + \frac{\text{aermr02}}{4.3} + \text{aermr04} + \text{aermr05} + 0.4 * \text{aermr06} + \text{aermr07} + \text{aermr08} + \text{aermr11} + \text{aermr09} + \text{aermr10} + \text{aermr16} + \text{aermr17} + \text{aermr18} \right] * \left(\frac{p_m}{R_{\text{spec}} * T} \right) \quad (\text{E. 3})$$

$$\rho_{\text{PM2P5}} = \left[\frac{\text{aermr01}}{4.3} + \frac{\text{aermr02} * 0.5}{4.3} + \text{aermr04} + \text{aermr05} + 0.7 * \text{aermr07} + 0.7 * \text{aermr08} + 0.7 * \text{aermr11} + \text{aermr09} + \text{aermr10} + 0.7 * \text{aermr16} + 0.25 * \text{aermr17} + 0.7 * \text{aermr18} \right] * \left(\frac{p_m}{R_{\text{spec}} * T} \right) \quad (\text{E. 4})$$

where $R_{\text{spec}}=287.058 \text{ J}/(\text{kg}\cdot\text{K})$ is the specific gas constant for dry air. The expression inside the parentheses in E.3 and E.4 corresponds to the dry air concentration (kg/m^3).

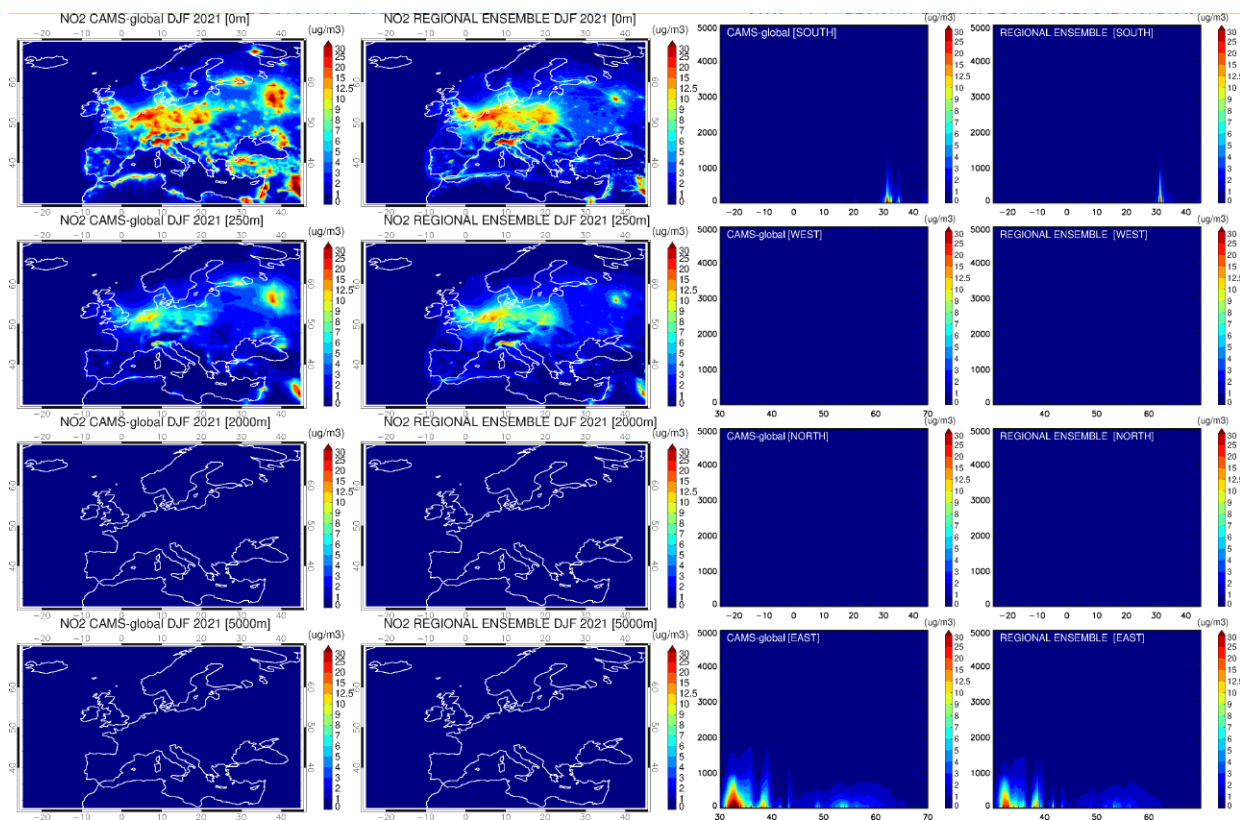


Figure 3.3. Left: Mean global and regional ENSEMBLE forecast NO₂ fields for four different vertical layers (0, 250, 2000, 5000 m) for DJF2021. Right: Cross sections for the same period of the global and regional ENSEMBLE NO₂ boundaries (south, west, north, east).

Regional model products are provided at the height levels of 0, 50, 250, 500, 1000, 2000, 3000 and 5000m. For every grid point and time step of the CAMS-global model, the differences between the height of each vertical layer midpoint z_m and the regional model height (e.g., 5000m) is calculated. The layer midpoint that exhibits the minimum height difference is the one that lies closest to the regional height level and is therefore selected for extraction of both chemical and aerosol species concentrations. The above procedure is performed for every regional height level. The final global product contains the O₃, CO, PM₁₀ and PM_{2.5} concentrations in eight height levels that correspond to the CAMS-global vertical levels that lie closest to the regional height levels.

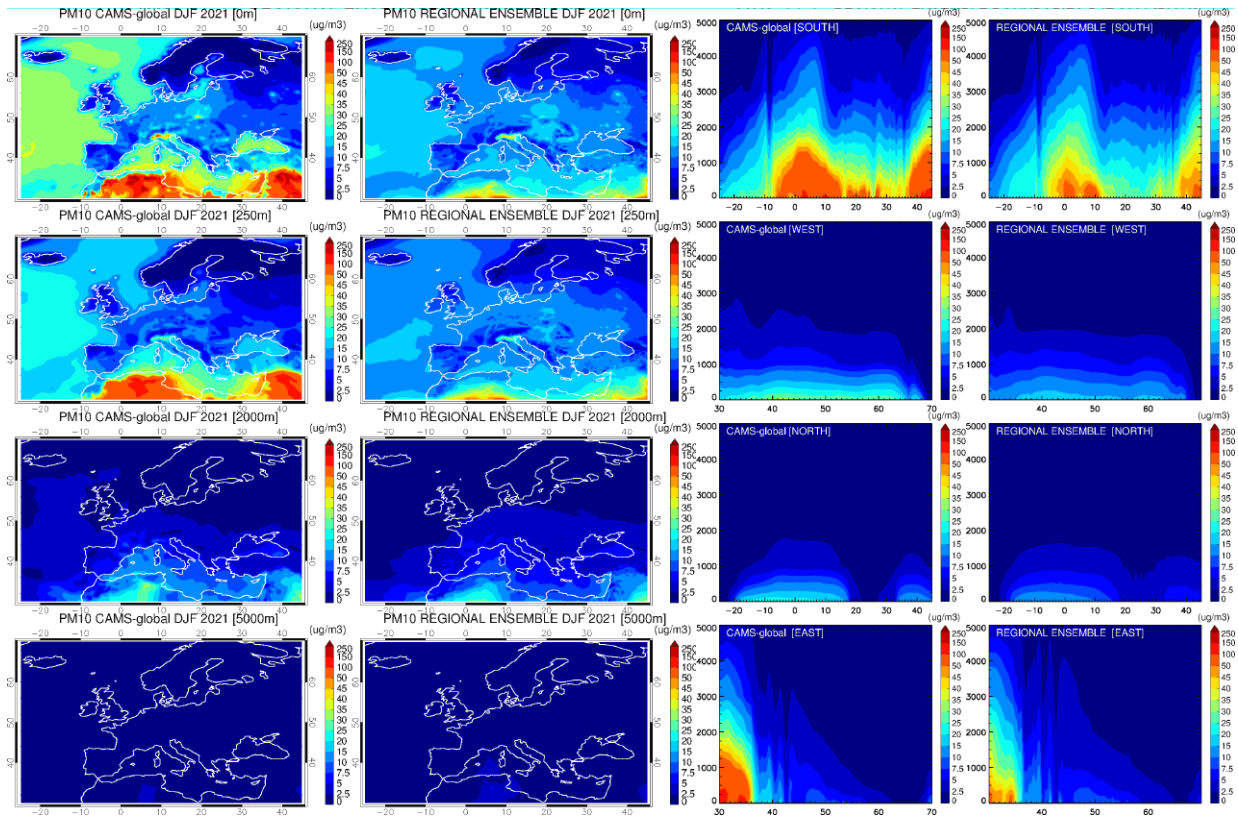


Figure 3.4 Left: Mean global and regional ensemble forecast PM10 fields for four different vertical layers (0, 250, 2000, 5000 m) for DJF2021. Right: Cross sections for the same period of the global and regional ensemble PM10 boundaries (south, west, north, east).

Table 3.1: Aerosol species description.

Label		Name	Size (μm)
aermr01	SS1	Sea Salt Aerosol	0.03-0.5
aermr02	SS2	Sea Salt Aerosol	0.5-5
aermr03	SS3	Sea Salt Aerosol	5-20
aermr04	DD1	Dust Aerosol	0.03-0.55
aermr05	DD2	Dust Aerosol	0.55-0.9
aermr06	DD3	Dust Aerosol	0.9-20
aermr07	OM1	Hydrophobic Organic Matter Aerosol	
aermr08	OM2	Hydrophilic Organic Matter Aerosol	
aermr09	BC1	Hydrophobic Black Carbon Aerosol	
aermr10	BC2	Hydrophilic Black Carbon Aerosol	
aermr11	SU1	Sulphate Aerosol	
aermr16	NI1	Nitrate fine mode aerosol	
aermr17	NI2	Nitrate coarse mode aerosol	
aermr18	NH3	Ammonium aerosol	

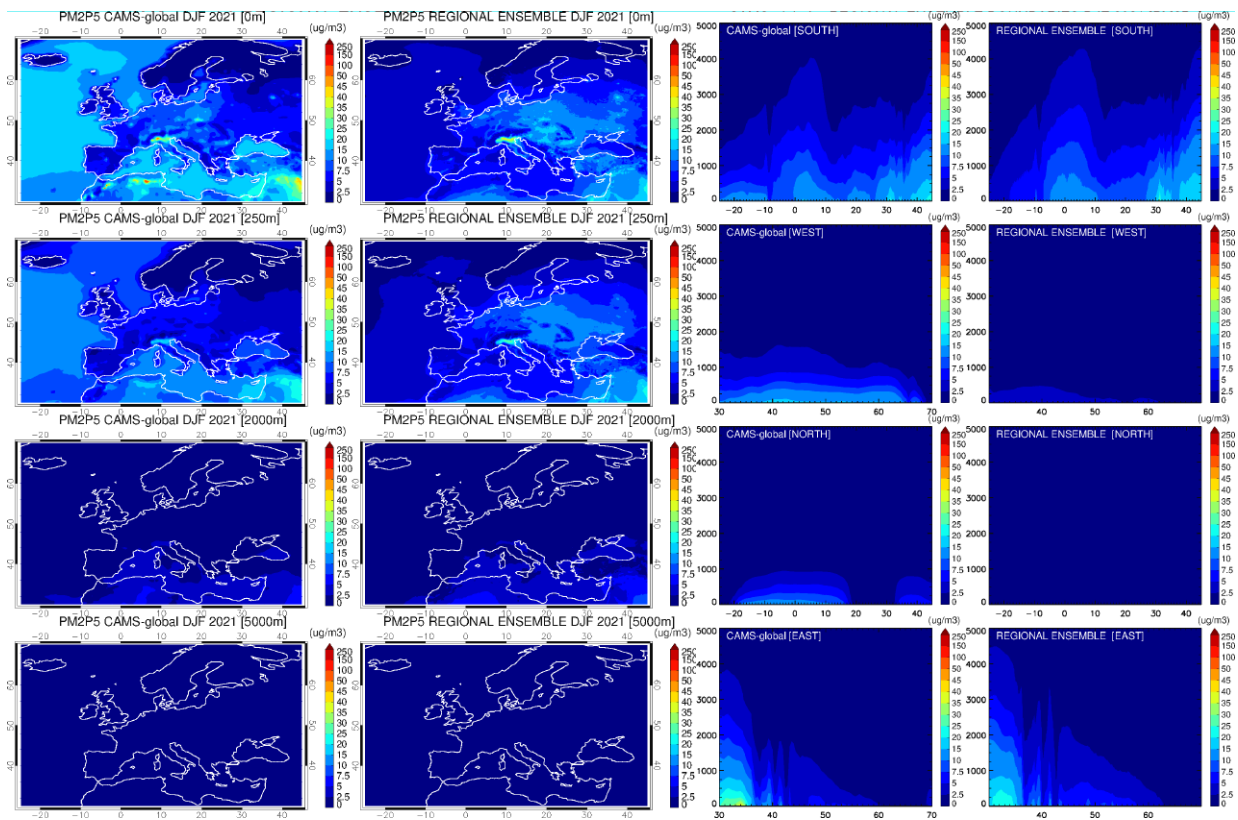


Figure 3.5 Left: Mean global and regional ENSEMBLE forecast $PM_{2.5}$ fields for four different vertical layers (0, 250, 2000, 5000 m) for DJF2021. Right: Cross sections for the same period of the global and regional ENSEMBLE ozone boundaries (south, west, north, east).

3.3 Consistency between the global and regional forecasts

This section reports on the consistency of the global and regional ENSEMBLE forecast. The analysis is performed for O_3 , CO, NO_2 , PM_{10} and $PM_{2.5}$ at four levels (0, 250, 2000 and 5000 m) for the time period from December 2020 to February 2021 (DJF2021).

Ozone (O_3)

Figure 3.1 presents the average DJF2021 spatial distribution of O_3 for different vertical layers (left) and the cross sections of the lateral boundaries (right) for the CAMS-global forecast and the regional ENSEMBLE. Overall, CAMS-global and ENSEMBLE agree well for O_3 , with CAMS-global exhibiting somehow lower ozone near the surface over continental Europe and North Africa, while the opposite stands for Scandinavia. The agreement in the four boundaries is very good, with minor inconsistencies at the eastern boundaries at $\sim 40^\circ$ N above 3000 m.

Carbon monoxide (CO)

Figure 3.2 shows the seasonal mean fields of CO and the cross sections of lateral boundaries (right) for ENSEMBLE and CAMS-global. There are more local surface CO maxima in the global forecast



product, over eastern Europe, Italy, and the coast of North Africa. Probably, this is due to the different emissions in the two products. The comparison in the boundaries is quite good.

Nitrogen dioxide (NO₂)

Figure 3.3 illustrates the seasonal mean fields of NO₂ and the cross sections of lateral boundaries (right) for ENSEMBLE and CAMS-global. Overall, there are higher surface NO₂ concentrations in CAMS-global compared to ENSEMBLE over Europe, Middle-East and North Africa. The ship emissions over the Mediterranean are mostly seen in the ENSEMBLE. From 2000m and above, both CAMS-global and ENSEMBLE exhibit very low concentrations (<1µg/m³). The comparison in the boundaries is good.

Aerosols (PM₁₀ and PM_{2.5})

Figures 3.4 and 3.5 illustrate the CAMS-global and ENSEMBLE spatial distributions (left) and lateral boundary cross sections (right) of PM₁₀ and PM_{2.5} mean fields, respectively. The main inconsistencies for particulate matter are summarized below:

- Higher PM₁₀ and PM_{2.5} in CAMS-global compared to ENSEMBLE in the boundary layer (mostly seen at the surface) over the Atlantic. This presumably results from the latest CAMS-global upgrade (CY47R1) and the new sea-salt emissions scheme implementation.
- Higher PM₁₀ in CAMS-global compared to ENSEMBLE over the southern and south-eastern boundaries of the domain up to 2000 m altitude.
- High PM₁₀/PM_{2.5} surface concentrations over Middle East seen in CAMS-global are not found in ENSEMBLE.
- Higher PM₁₀/PM_{2.5} at the west and north boundaries probably related to the abovementioned CAMS-global upgrade.

3.4 Regional variability

Ozone (O₃)

Figure 3.6 shows ozone mean fields for DJF2021 of the individual regional ensemble members and CAMS-global (bottom panel) for selected altitudes (0, 250, 2000, 5000 m). SILAM, LOTOS-EUROS, and EURAD-IM exhibit lower surface ozone concentrations over central Europe compared to CAMS-global. GEMAQ, LOTOS-EUROS, and MOCAGE models have lower O₃ values over the Atlantic. MOCAGE exhibits higher concentrations over the Mediterranean near the surface and over central Europe at 250m compared to CAMS-global. CHIMERE exhibits higher O₃ concentrations up to 2000m compared to CAMS-global.

Carbon monoxide (CO)

The DJF2021 mean fields of carbon monoxide for the regional ensemble members as well as for CAMS-global are presented in Figure 3.7 for different vertical layers. No particular outliers are found, as CO in the regional models is within the expected variability. GEMAQ, EMEP, SILAM, and MOCAGE have spatially more extended CO maxima than all other regional models and CAMS-global near the surface. DEHM exhibits lower CO values near the surface over central Europe than all other regional models and CAMS-global. LOTOS-EUROS has lower CO values at the 20°-40° E, 50°-60° N region near the surface, which may be due to this being the first full quarter where the vertical resolution of the



model for the forecasts has been increased from 5 to 12 vertical levels. CHIMERE has higher CO values at 5000m, unlike other regional models and the CAMS-global.

Nitrogen Dioxide (NO₂)

The DJF2021 mean fields of nitrogen dioxide for the regional ensemble members as well as for CAMS-global are illustrated in Figure 3.8 for several vertical layers. The comparison is generally good, with no obvious outliers. EMEP exhibits the lowest NO₂ levels near the surface, CHIMERE and MATCH at 250m altitude, among the regional models. Low NO₂ levels (< 1µg/m³) above 2000m are seen in all regional models and the CAMS-global.

Aerosols (PM₁₀ and PM_{2.5})

The mean PM₁₀ and PM_{2.5} fields for DJF2021 are presented in Figures 3.9 and 3.10, respectively. Major features are noted below:

- MOCAGE and DEHM have higher surface PM₁₀ over the Atlantic in comparison to all other regional models. In particular the MOCAGE model has higher PM₁₀ even at 250 m altitude, which is in disagreement with CAMS-global.
- MOCAGE, EURAD, MATCH and DEHM exhibit relatively lower PM₁₀ levels at the south boundary.
- EMEP, SILAM and GEMAQ have higher PM_{2.5} levels in the PBL near the south boundaries compared to CAMS-global, while LOTOS-EUROS, MOCAGE, MATCH, and DEHM have lower.

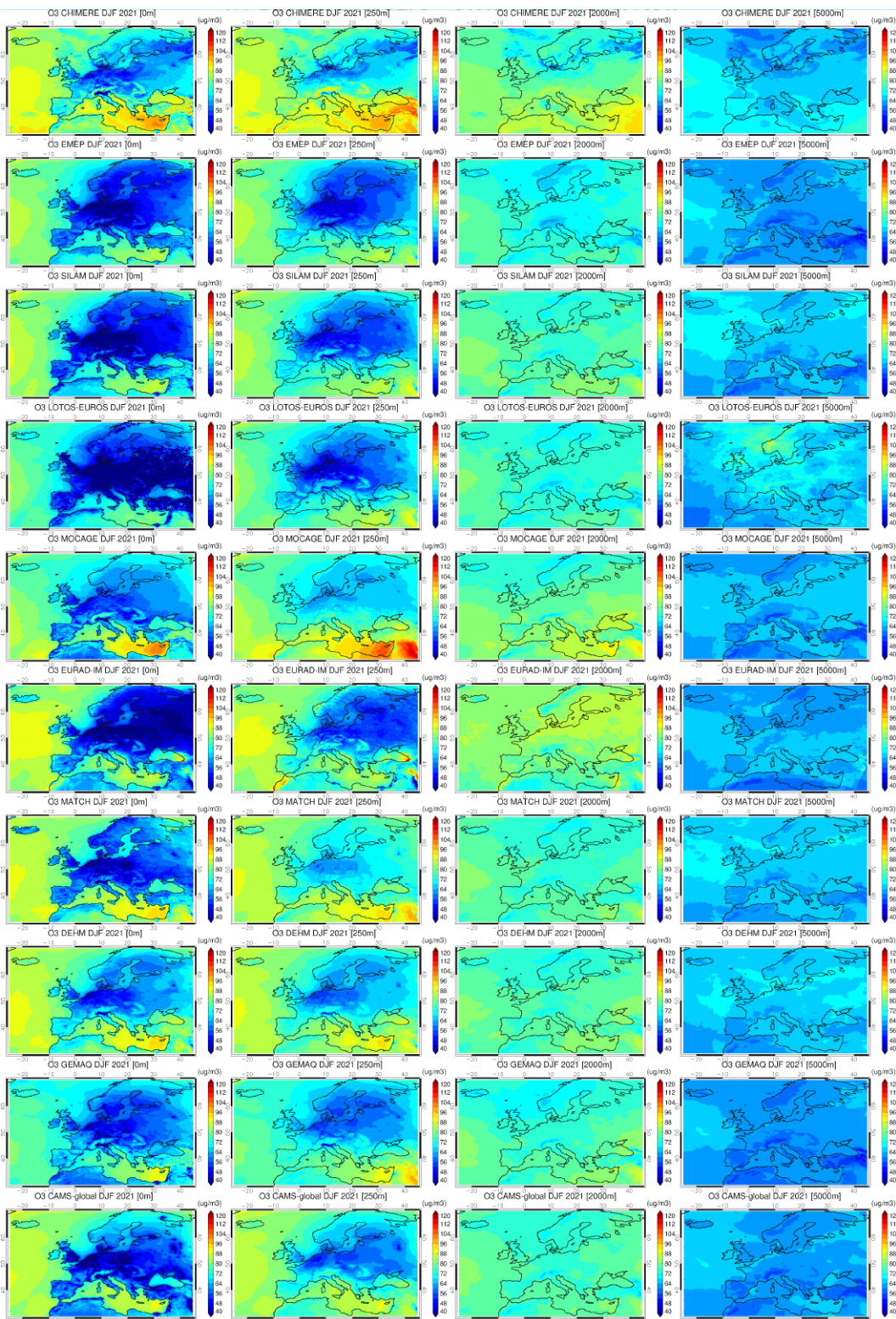


Figure 3.6 Mean regional ozone forecasts for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global).

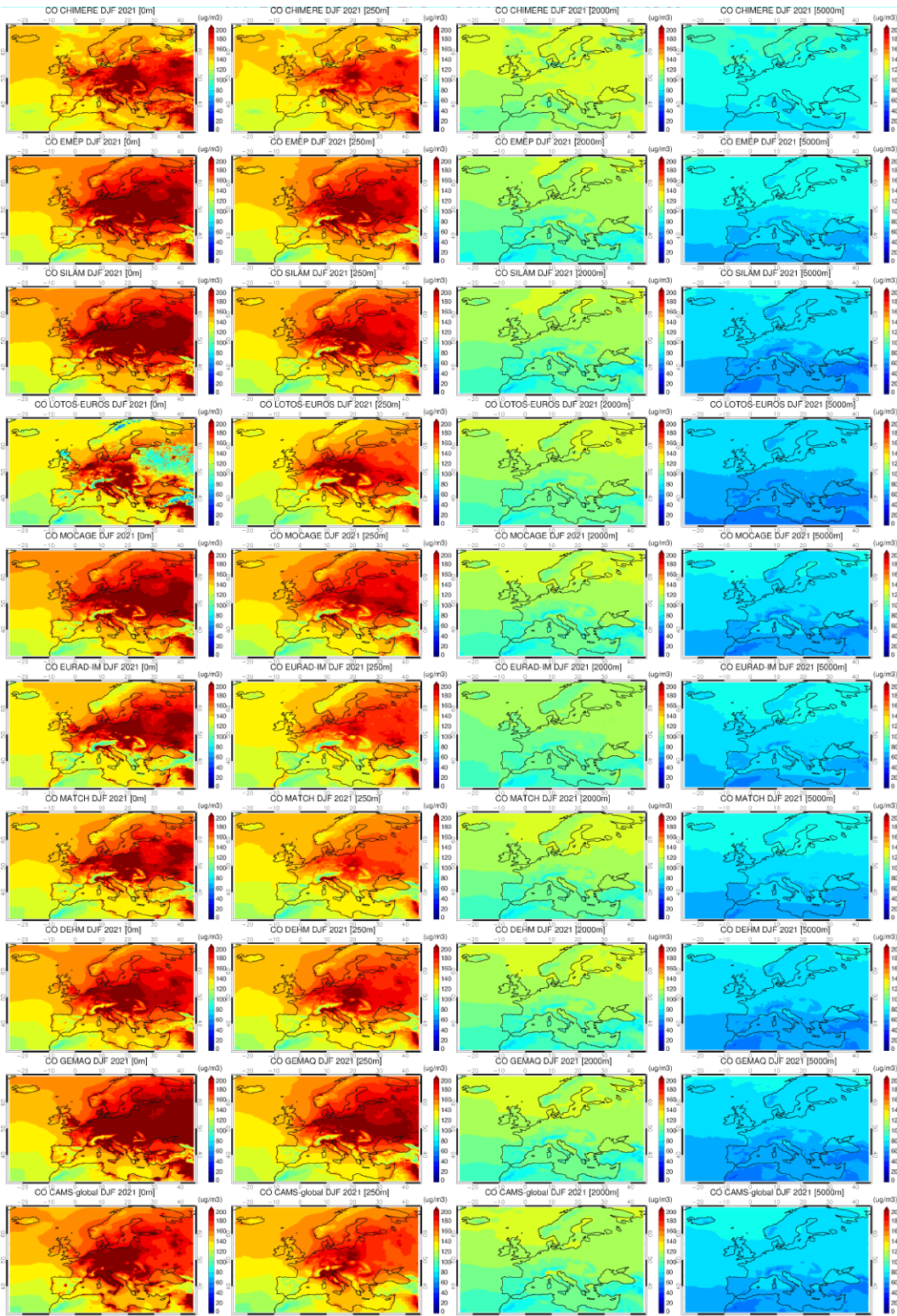


Figure 3.7. Mean regional CO forecasts for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAQE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global).

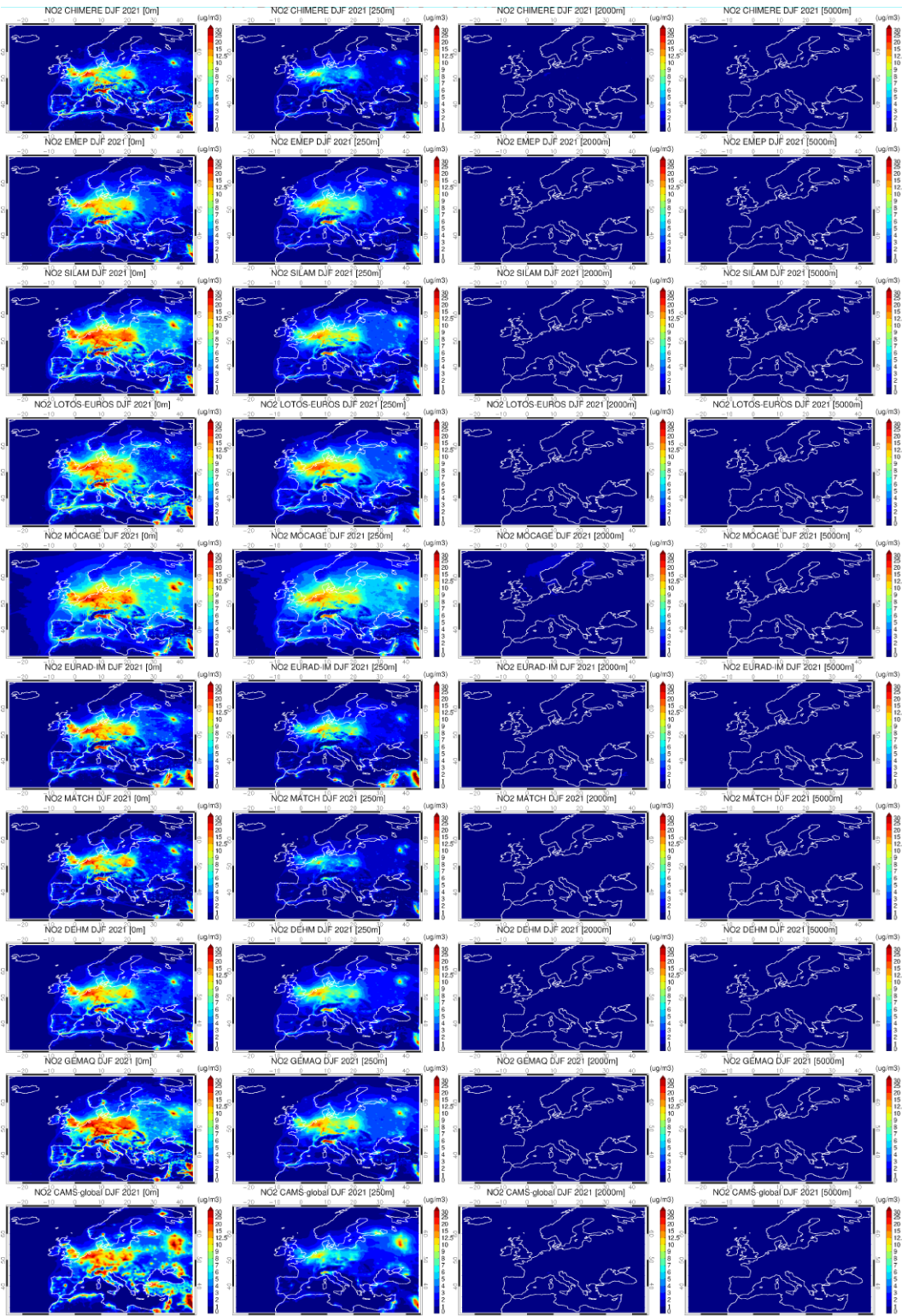


Figure 3.8. Mean regional NO₂ forecasts for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global).

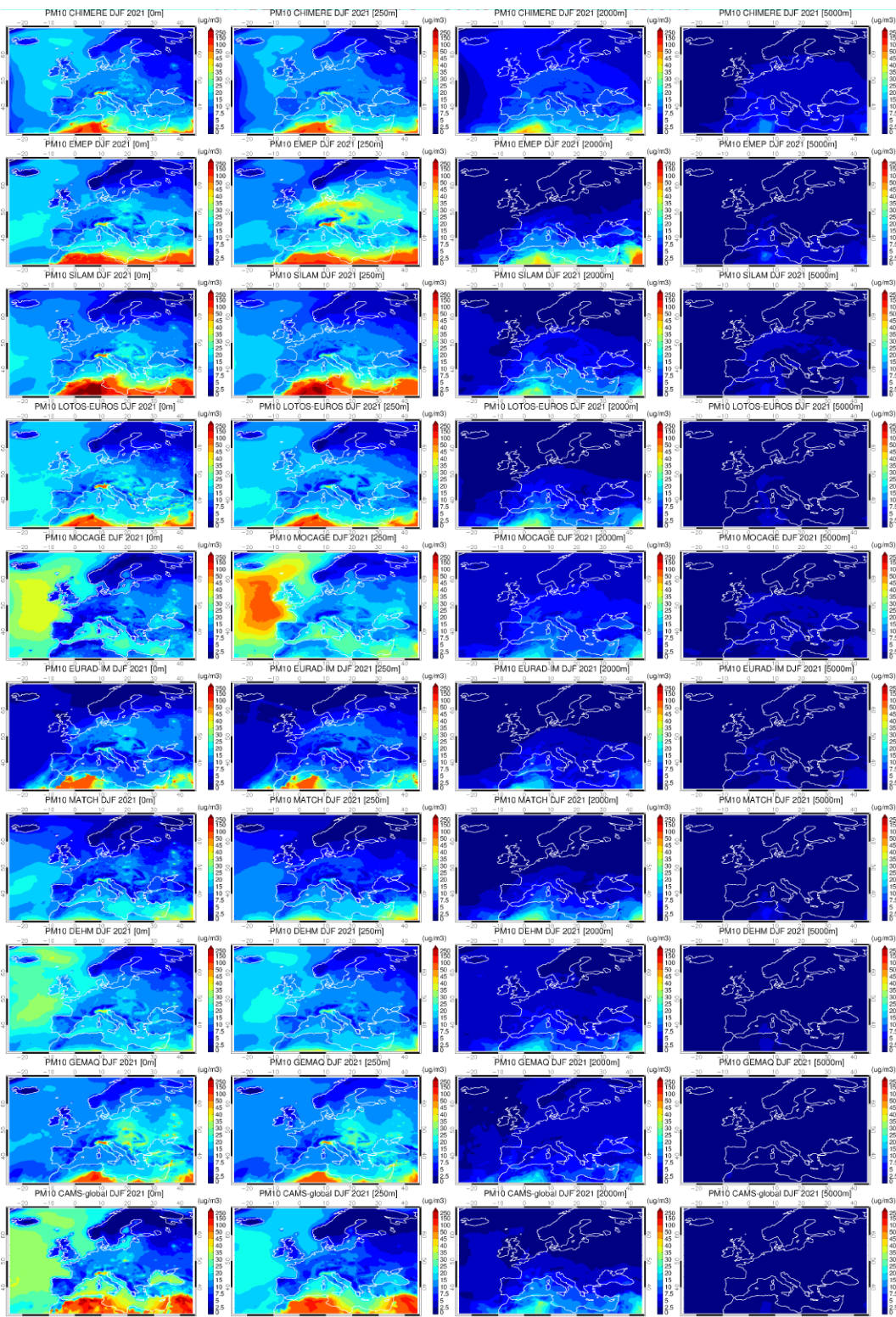


Figure 3.9. Mean regional PM₁₀ forecasts for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global).

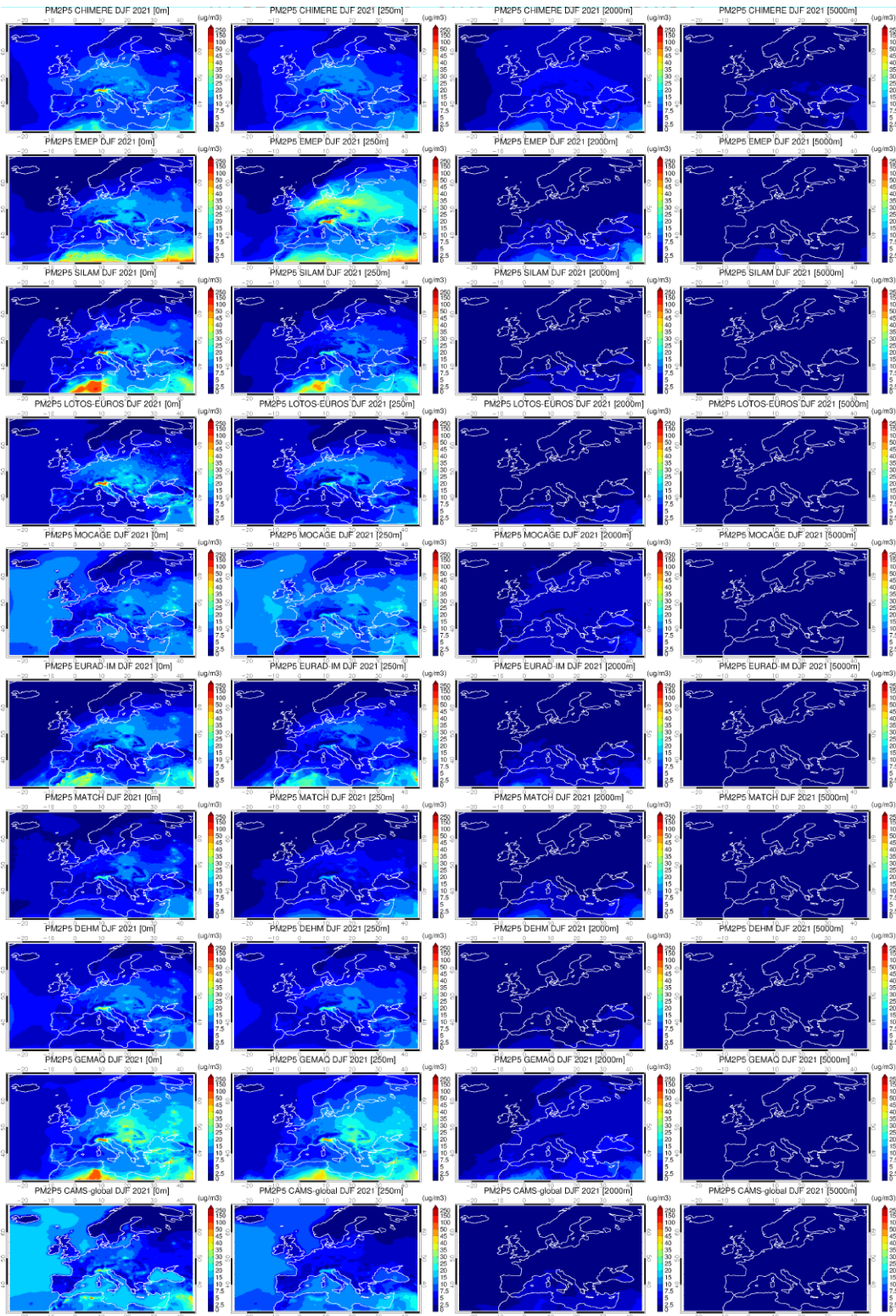


Figure 3.10. Mean regional PM_{2.5} forecasts for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global).



3.5 Time series

Figure 3.11 shows the mean daily time series for DJF2021 for the five species, namely O₃, CO, NO₂, PM₁₀ and PM_{2.5} (from left to right) for different European sub-regions (from top to bottom): Alps (AL), British Isles (BI), East Europe (EA), France (FR), Iberian Peninsula (IP), Mediterranean (MD), Mid-Europe (ME), Scandinavia (SC).

Each subregion is defined with the following latitude/longitude boundaries:

Name = (BI, IP, FR, ME, SC, AL, MD, EA)

West = (-10, -10, -5, 2, 5, 5, 3, 16)

East = (2, 3, 5, 16, 30, 15, 25, 30)

South = (50, 36, 44, 48, 55, 44, 36, 44)

North = (59, 44, 50, 55, 70, 48, 44, 55)

The letter R denotes the temporal correlation between the two products. Only concentrations over land are used.

For O₃ the temporal variability between CAMS-global and ENSEMBLE is in very good agreement (R values of 0.88 to 0.96).

The temporal correlation for CO is also very good (0.6 to 0.95), with CAMS-global exhibiting higher levels mostly for the Alps.

For NO₂ the temporal variability between CAMS-global and ENSEMBLE is very good (R values of 0.75 to 0.94).

The agreement in the PM₁₀ daily timeseries between the global and regional forecasts ranges is very good for the majority of the regions, with the exception of BI (0.36).

The magnitude of PM₁₀ in CAMS-global is close to the regional ensemble, however CAMS-global PM₁₀ is higher over AL.

GEMAQ (green) and SILAM (orange) exhibit higher PM₁₀ compared to CAMS-global during CAMS-global peaks

The temporal agreement in surface PM_{2.5} is quite good (0.6 to 0.92).

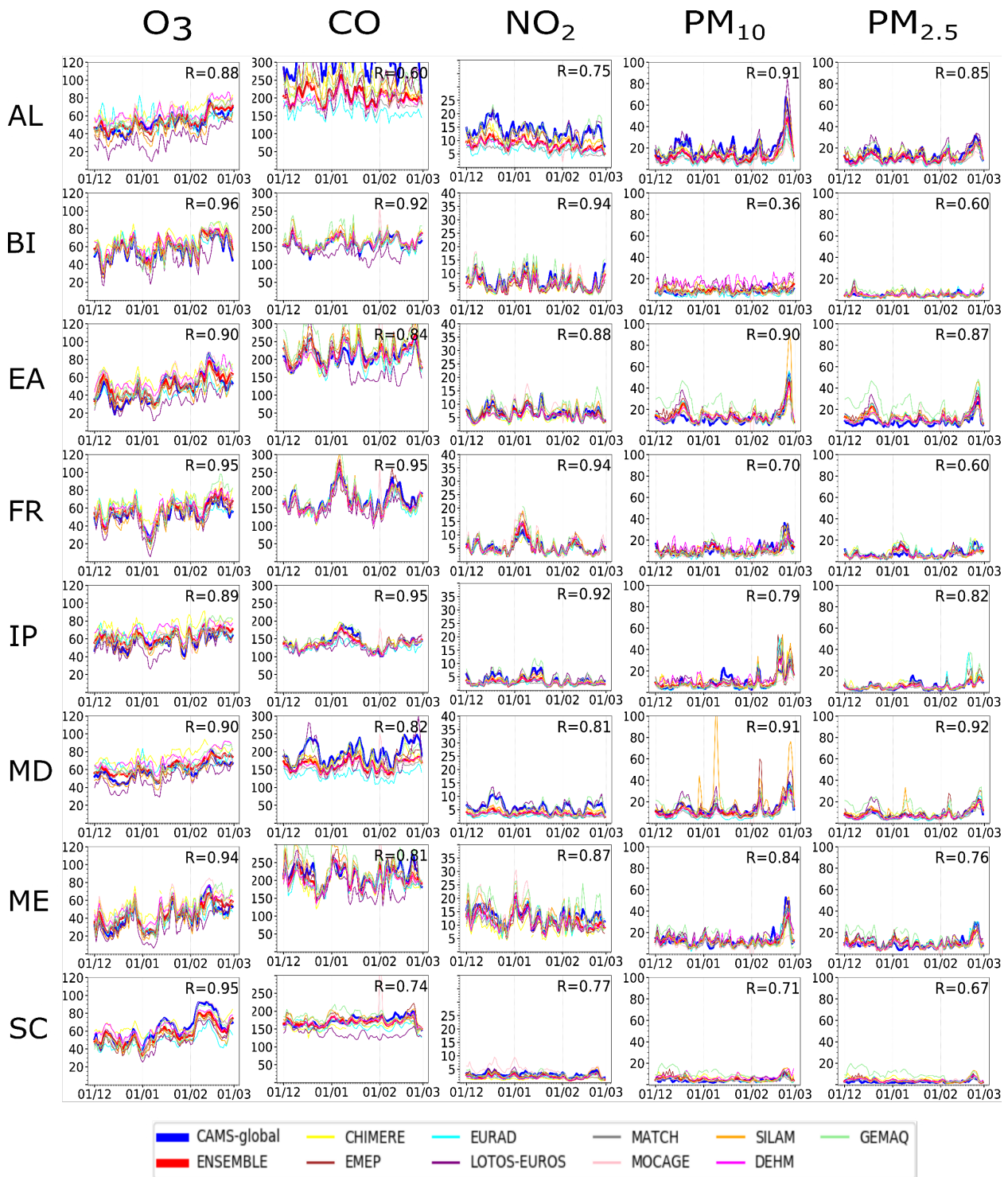


Figure 3.11. Mean daily time series for surface O₃, CO, NO₂, PM₁₀ and PM_{2.5} for DJF2021 (unit $\mu\text{g}/\text{m}^3$). The blue line is CAMS-global and the red line the ENSEMBLE forecasts. Each line in the composite plot denotes a different European subregion.



3.6 Diurnal cycles

Figure 3.12 presents the diurnal cycles for surface O_3 , CO, NO_2 , PM_{10} and $PM_{2.5}$ averaged over the period DJF2021 for different European sub-regions (from top to bottom): Alps (AL), British Isles (BI), East Europe (EA), France (FR), Iberian Peninsula (IP), Mediterranean (MD), Mid-Europe (ME), Scandinavia (SC). The red colour is used for the regional ensemble and the blue for CAMS-global.

There is a good agreement between the O_3 CAMS-global and the regional diurnal cycles, both in timing and amplitude of the diurnal cycle.

The diurnal range for surface CO is less pronounced for the regional products over some regions, mostly over the Alps and to a lesser extent over the Mediterranean.

The diurnal cycle of NO_2 at the surface reveals that regional product and the CAMS-global forecasts agree on the shape, yet the diurnal range is higher in CAMS-global mostly over AL, MD, and ME.

There is a mediocre agreement of the PM_{10} and $PM_{2.5}$ diurnal cycles between the global and the regional product.

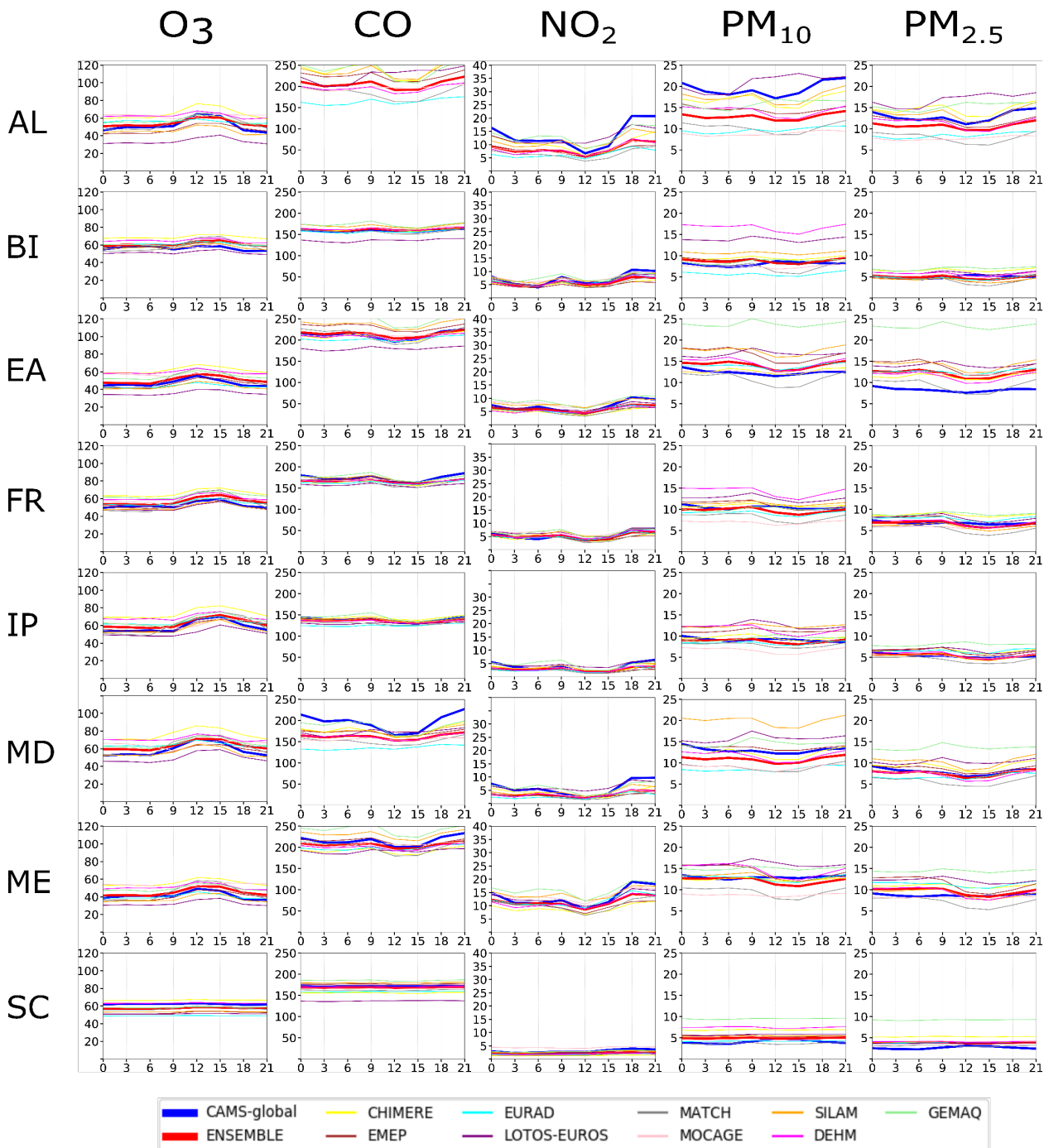


Figure 3.12. Diurnal cycles for surface O₃, CO, NO₂, PM₁₀ and PM_{2.5} for the period DJF2021 (unit: $\mu\text{g}/\text{m}^3$). The blue line is CAMS-global and the red line the ENSEMBLE forecast. Each line in the composite plot denotes a different European sub-region.



3.7 Regional domain boundary cross sections

Ozone (O₃)

Figure 3.13 shows the regional variability in the lateral domain boundary cross sections of O₃ (from left to right): south, west, north, east and the different ensemble members and CAMS-global (from top to bottom) averaged over the period DJF2021. CHIMERE in all four boundaries has spurious implementation of O₃ boundaries, while EURAD and MOCAGE at the south boundaries differ slightly from the rest of the regional models.

Carbon monoxide (CO)

Figure 3.14 presents the regional variability in the lateral cross sections of CO (from left to right): south, west, north, east and the different ensemble members and CAMS-global (from top to bottom) averaged over the period DJF2021. CHIMERE appears to have the same issue with the boundaries implementation as with ozone.

Nitrogen dioxide (NO₂)

Figure 3.15 shows the regional variability in the lateral cross sections of NO₂ (from left to right): south, west, north, east and the different ensemble members and CAMS-global (from top to bottom) averaged over the period DJF2021. Overall, no particular outlier for NO₂ boundaries. MOCAGE shows the most vertically extended NO₂ concentrations at the east boundaries of the domain.

Aerosols (PM₁₀ and PM_{2.5})

Figure 3.16/3.17 shows the regional variability in the lateral cross sections of PM₁₀/PM_{2.5} respectively (left to right): south, west, north, east and the different ensemble members and CAMS-global (top to bottom). We note the following inconsistencies:

- MOCAGE, EURAD, and DEHM exhibit lower PM₁₀ concentrations over the south boundaries than CAMS-global.
- EMEP exhibits somehow higher PM₁₀ than CAMS-global at the south boundaries.
- MOCAGE has higher PM₁₀ than CAMS-global at the west and north boundaries, probably due to sea salt emissions.
- MOCAGE, EURAD, MATCH, DEHM, and GEMAQ have lower PM₁₀ at the east boundaries.
- All regional models (except MOCAGE) exhibit lower PM₁₀ at the west boundaries, possibly related with the latest CAMS-global upgrade (sea salt emissions scheme).
- All models except LOTOS-EUROS, MOCAGE, MATCH and DEHM have higher southern PM_{2.5} boundaries than CAMS-global.
- EMEP, and to a lesser extent EURAD and GEMAQ, have additionally higher eastern PM_{2.5} boundaries than CAMS-global.

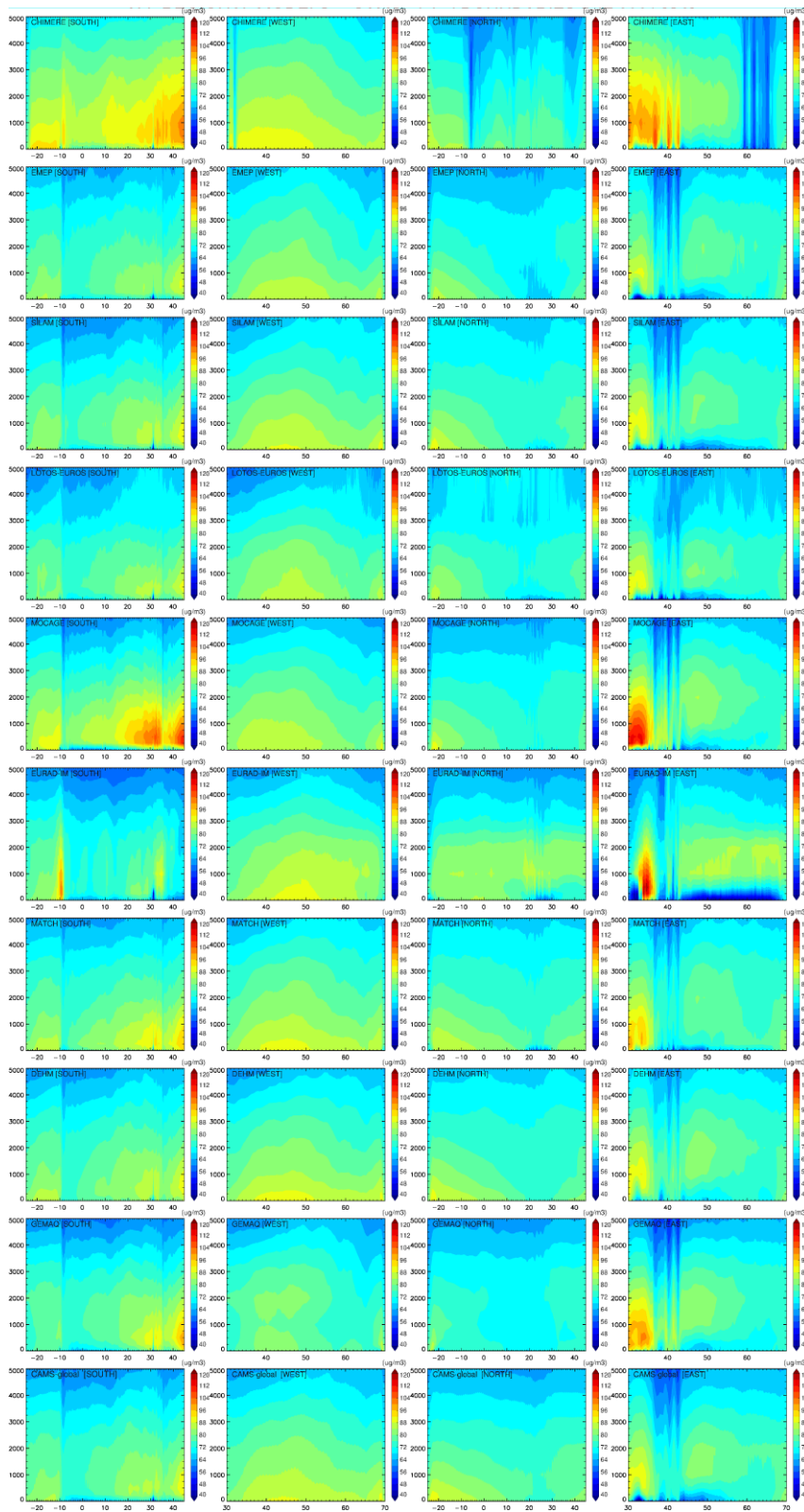


Figure 3.13. Ozone cross sections for DJF2021 for the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global) and the lateral boundaries (left to right: south, west, north, east).

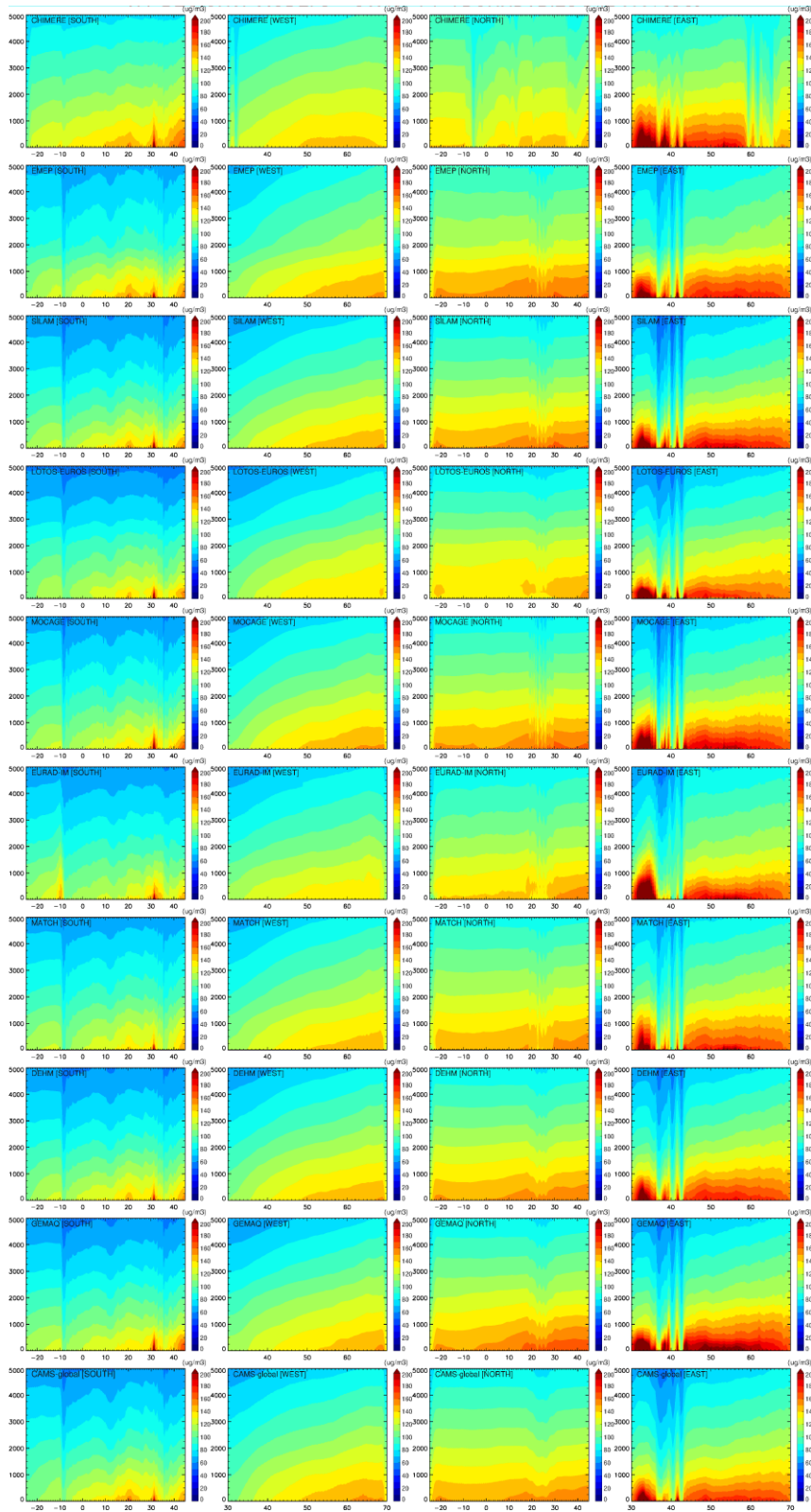


Figure 3.14. Carbon Monoxide cross sections for DJF2021 for the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global) and the lateral boundaries (left to right: south, west, north, east).

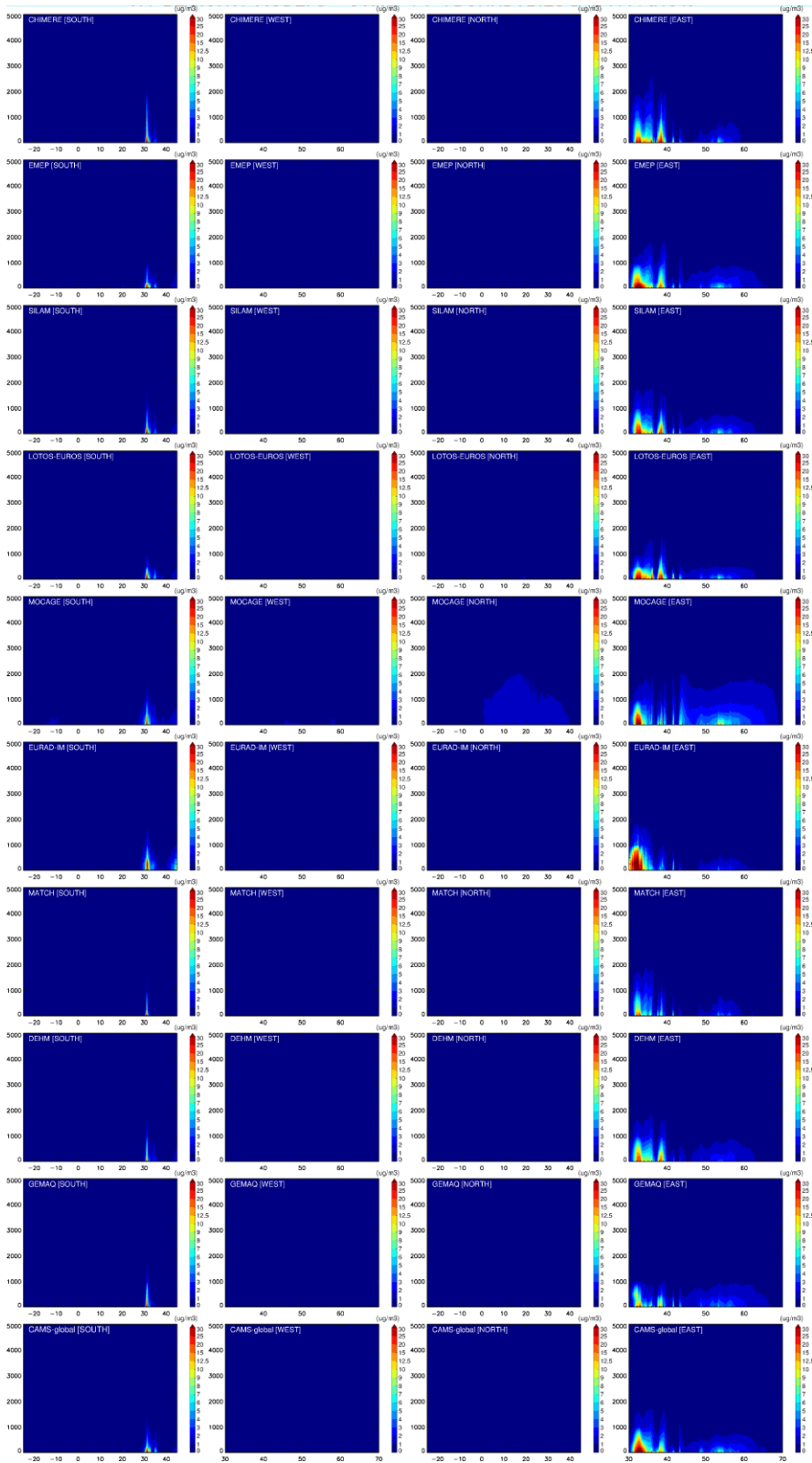


Figure 3.15. Nitrogen dioxide cross sections for DJF2021 for the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global) and the lateral boundaries (left to right: south, west, north, east).

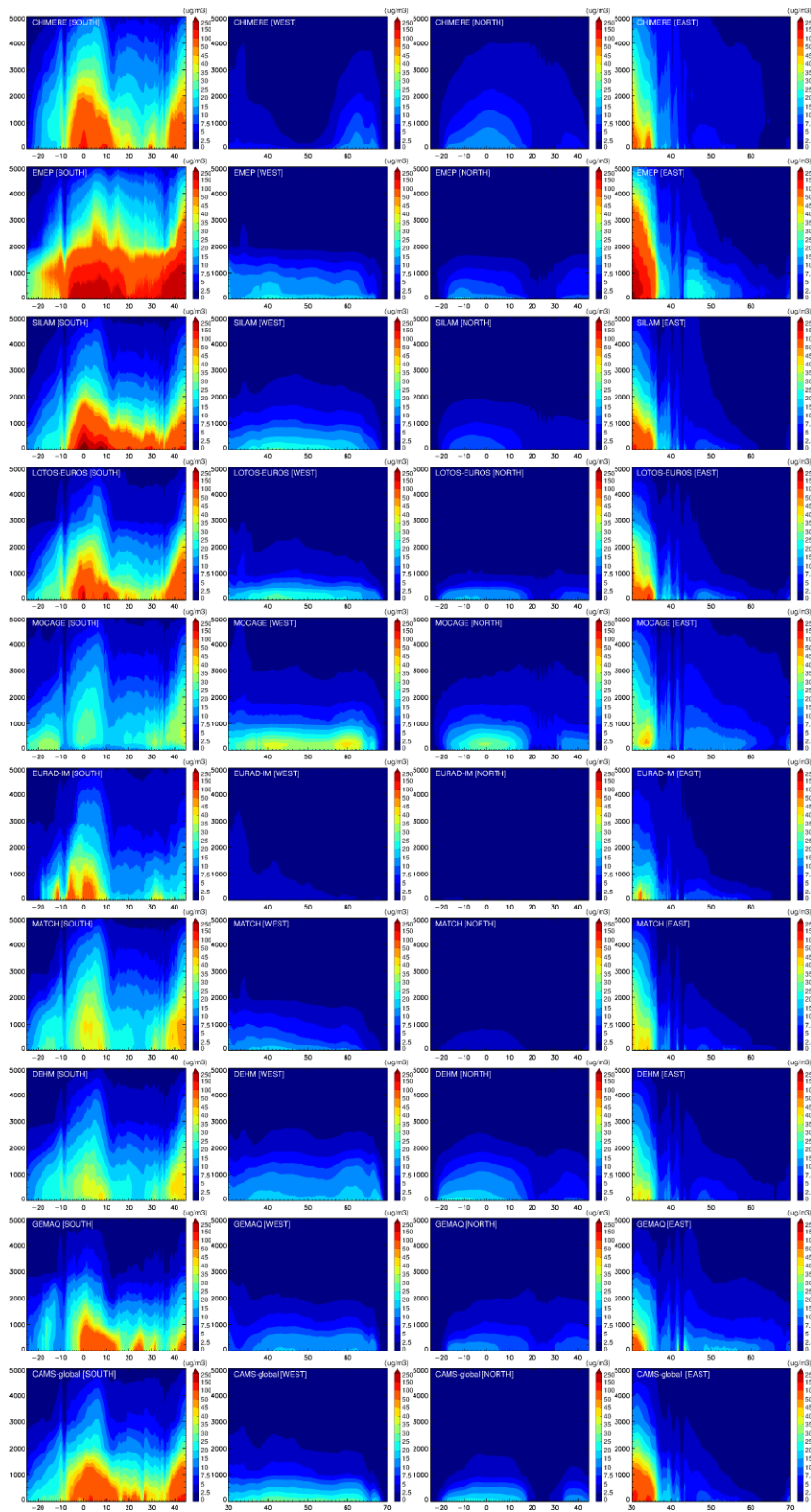


Figure 3.16. Aerosol PM₁₀ cross sections for DJF2021 for the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global) and the lateral boundaries (left to right: south, west, north, east).

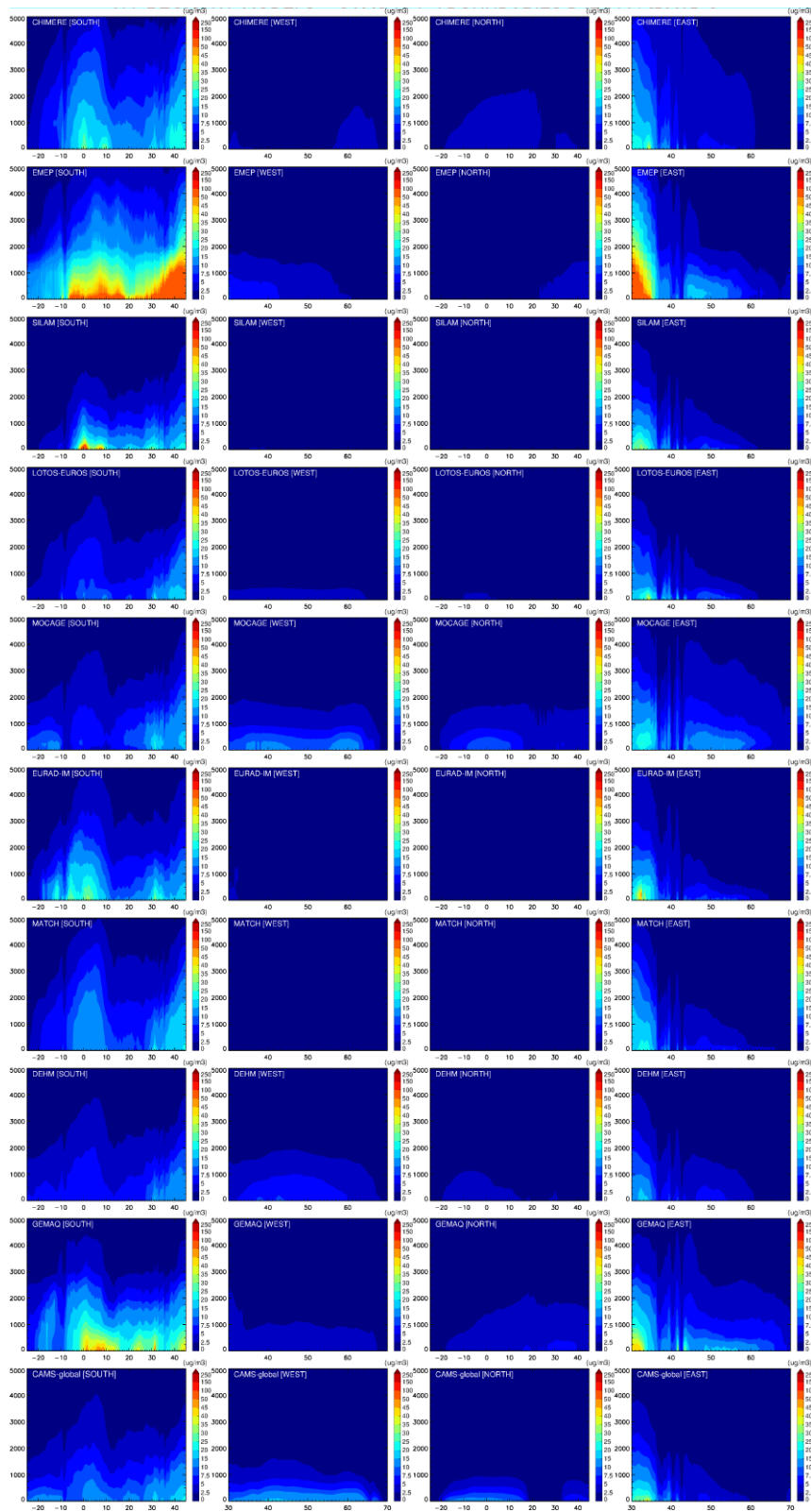


Figure 3.17 Aerosol PM_{2.5} cross sections for DJF2021 for the nine ensemble members and CAMS-global (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and CAMS-global) and the lateral boundaries (left to right: south, west, north, east).



3.8 Regional analysis vs. regional forecasts

In the following sections we compare the regional analysis products with the regional forecasts (Day1). The four following figures (3.18-3.22) show the mean regional differences between analysis and forecasts for the time period DJF2021 at four different vertical layers (0, 250, 2000, 5000 m, left to right) including O₃, CO, NO₂, PM₁₀ and PM_{2.5}, respectively.

Regional models with the largest differences between the analyses and the 1st day forecasts are for:

Ozone

- CHIMERE and SILAM in the surface layer.
- EMEP, MOCAGE, and DEHM mostly within the PBL (patchy patterns)
- MATCH and LOTOS-EUROS in all levels. For LOTOS-EUROS, this difference can be attributed to the difference in vertical resolution between the forecast and analysis suites.

Carbon monoxide

Differences are seen in:

- MATCH, DEHM and SILAM (up to 2000m) exhibit much lower CO levels compared to analysis.
- CHIMERE and GEMAQ at the surface.

Nitrogen dioxide

- Lower NO₂ forecast compared to analysis at the surface for CHIMERE and SILAM, and in the boundary layer for EMEP.

PM₁₀ and PM_{2.5}

- Minor differences in some models, most pronounced in MATCH for PM₁₀ and PM_{2.5} with lower forecast compared to analysis.

It is important to note that the differences observed between the forecast and the analysis do not only reflect the impact of the assimilation but may also result from differences in the model setup between the analysis systems and the forecast systems.

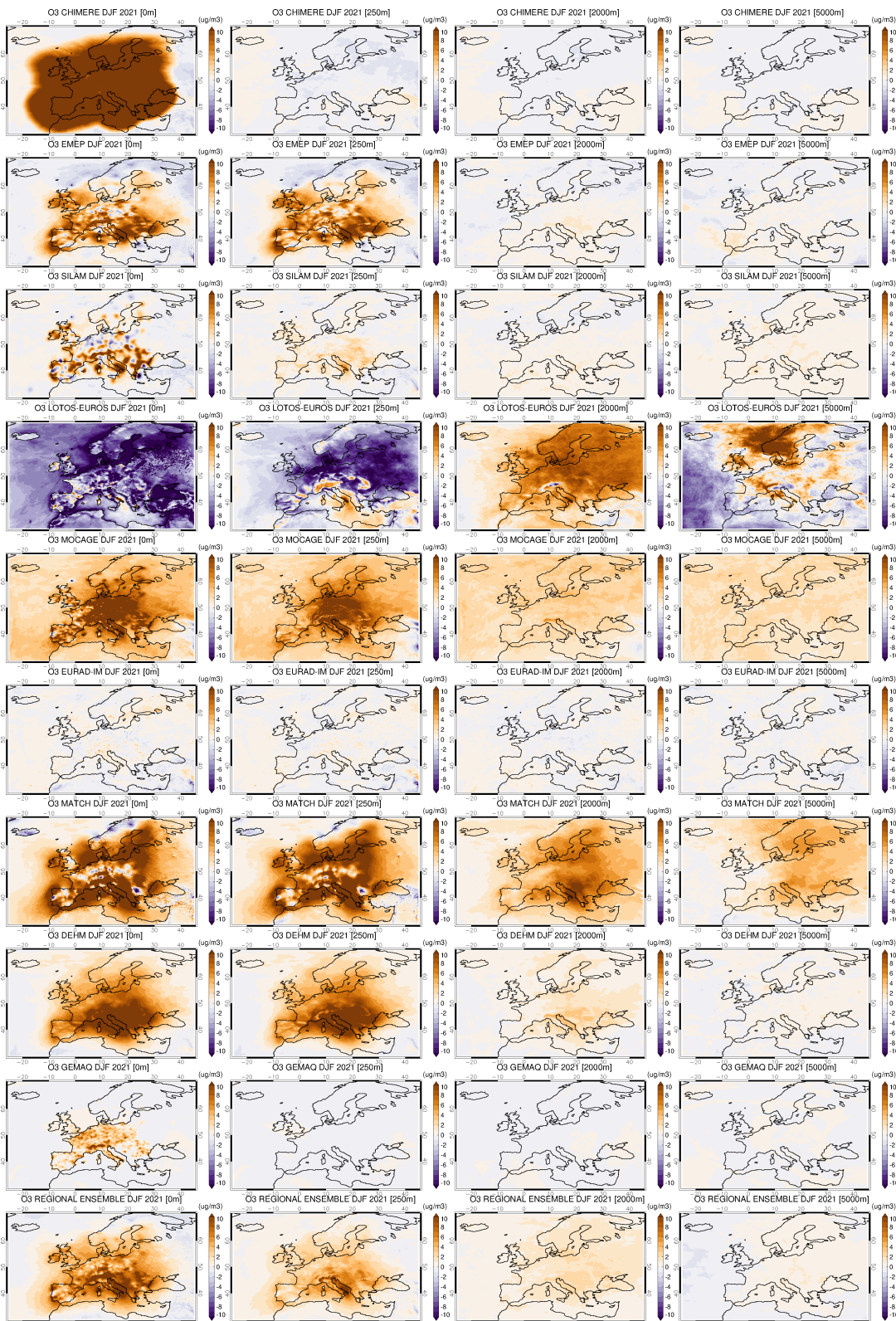


Figure 3.18. Mean regional O₃ differences between analysis and forecast for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from regional ENSEMBLE and individual ensemble members (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOÇAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and ENSEMBLE).

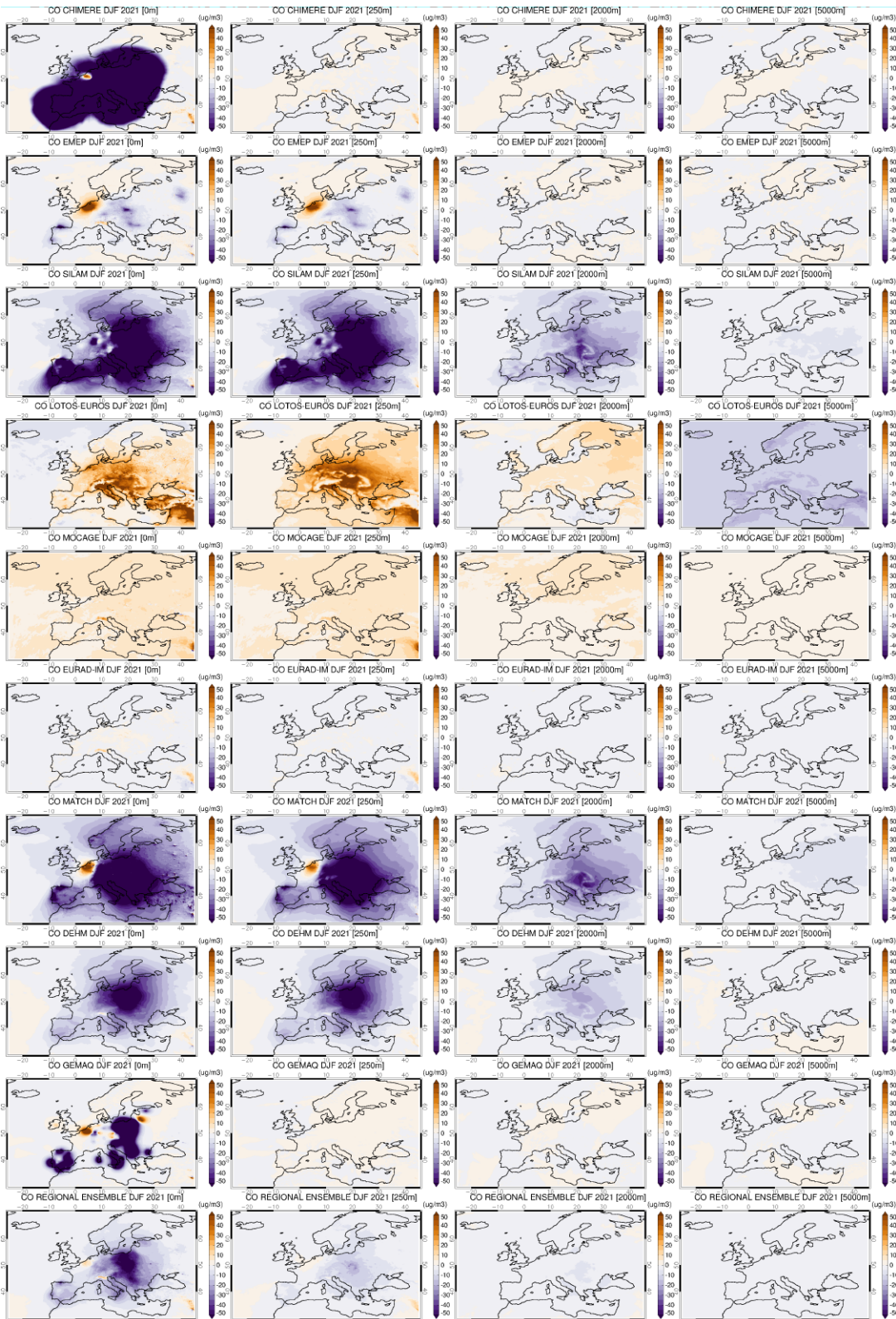


Figure 3.19. Mean regional CO differences between analysis and forecast for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from regional ENSEMBLE and individual ensemble members (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOcAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and ENSEMBLE).

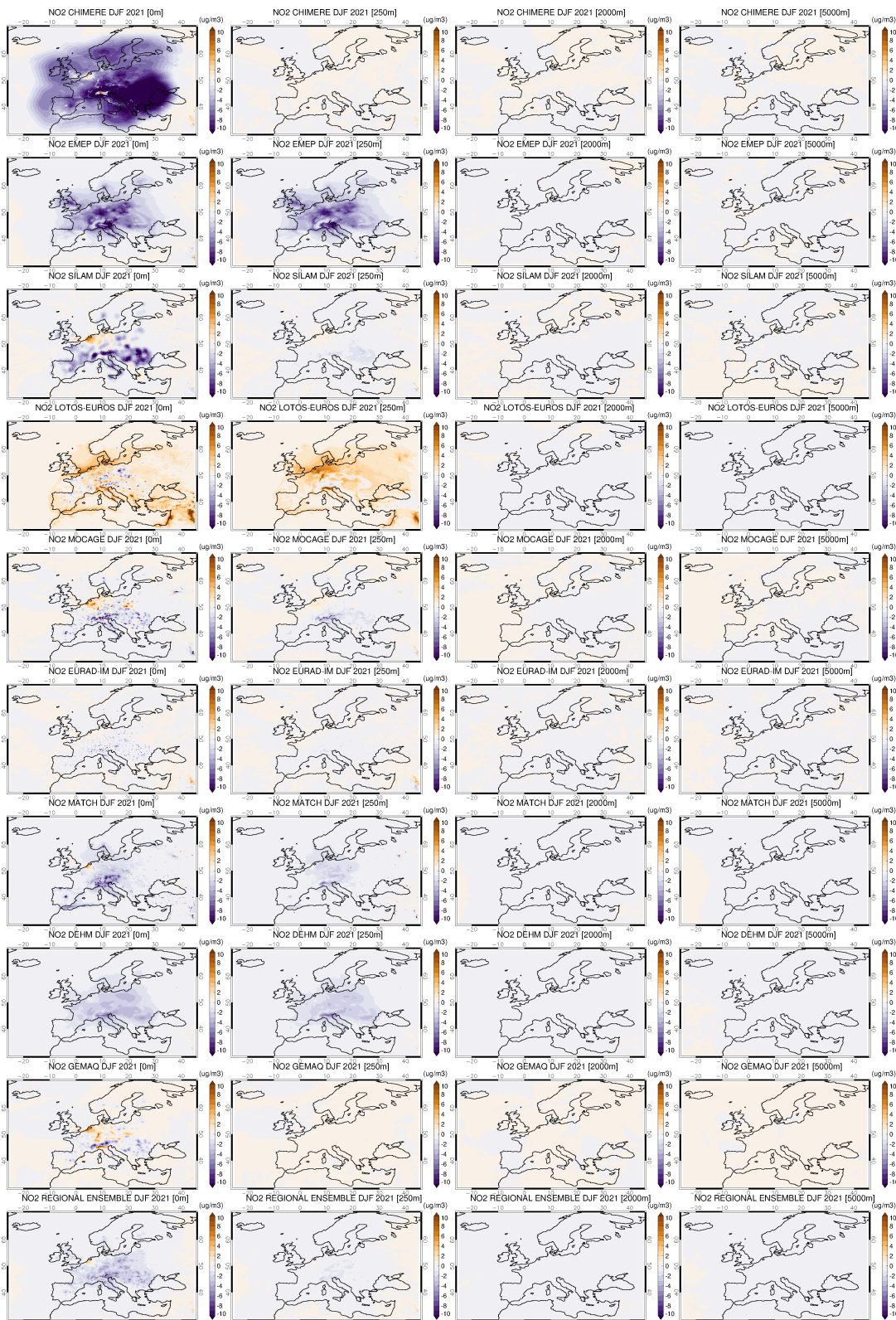


Figure 3.20. Mean regional NO₂ differences between analysis and forecast for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from regional ENSEMBLE and individual ensemble members (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and ENSEMBLE).

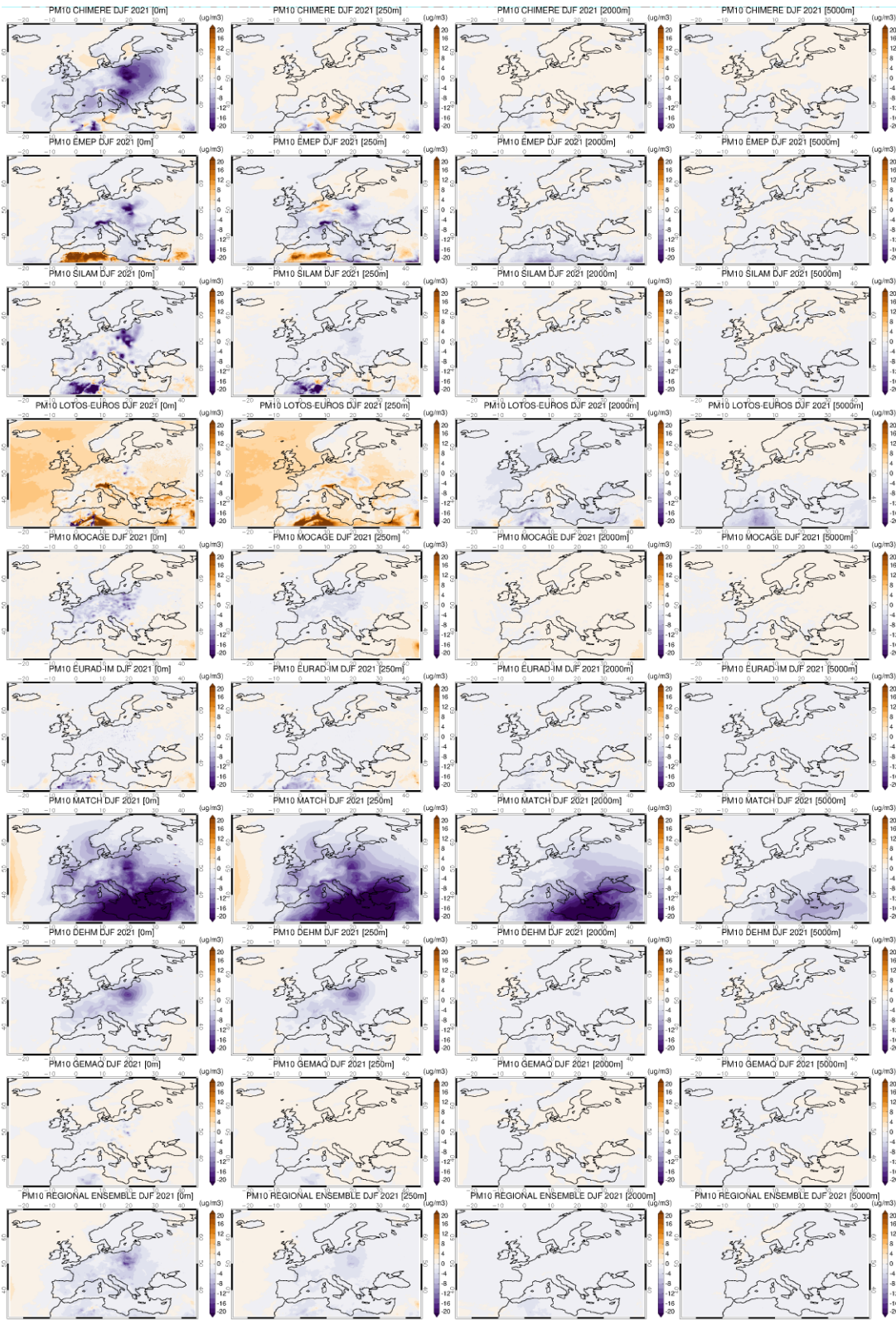


Figure 3.21. Mean regional PM₁₀ differences between analysis and forecast for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from regional ENSEMBLE and individual ensemble members (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEM-AQ and ENSEMBLE).

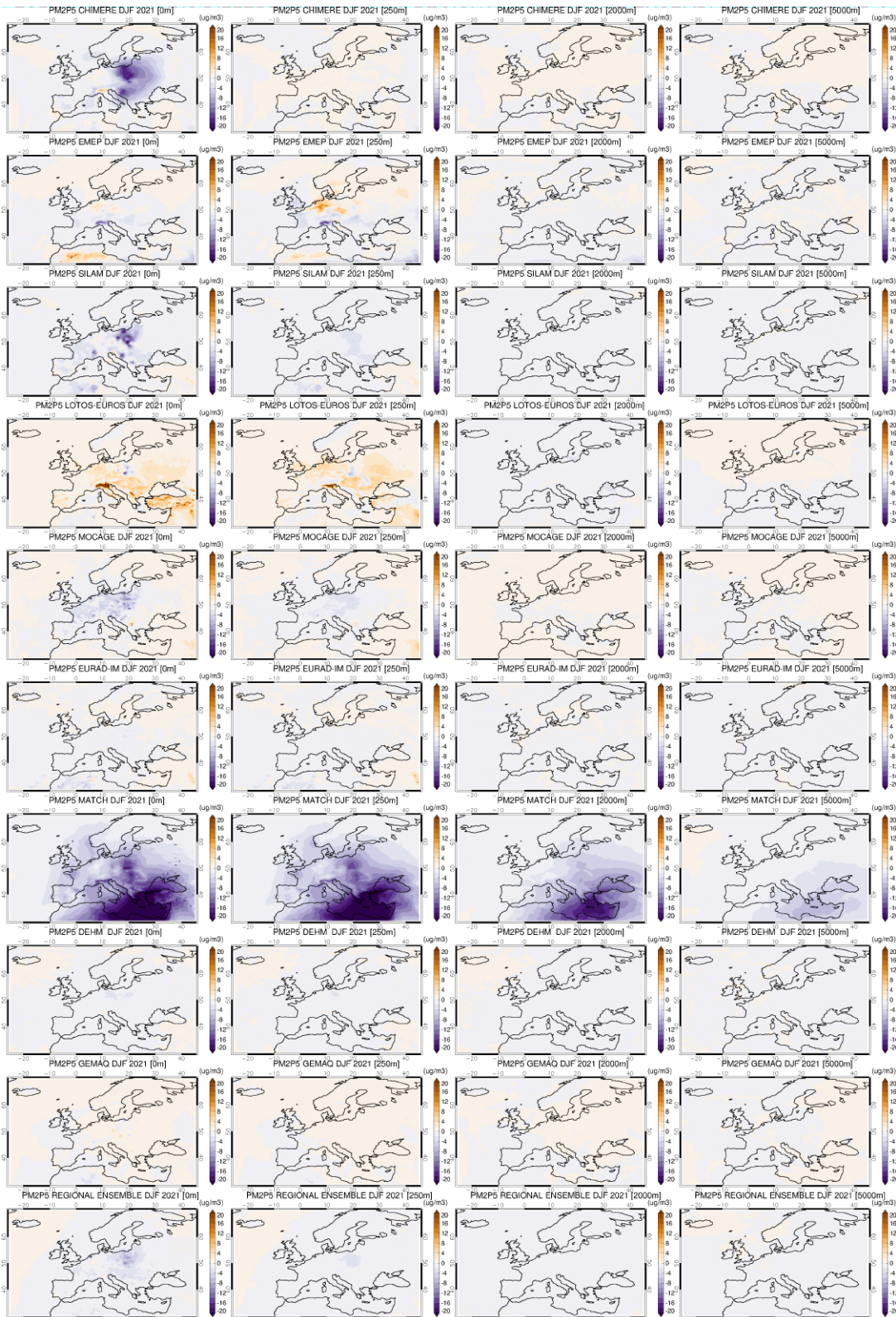


Figure 3.22. Mean regional PM_{2.5} differences of forecast from analysis for DJF2021 for four different vertical layers (0, 250, 2000, 5000 m) from regional ensemble and individual ensemble members (top to bottom: CHIMERE, EMEP, SILAM, LOTOS-EUROS, MOCAGE, EURAD-IM, MATCH, DEHM, GEMAO and ENSEMBLE).



4 Vertical profile and column aerosol comparisons

4.1 Summary for the EARLINET lidar and Aeronet comparisons

The regional models are compared with climatological lidar profiles for each season (EARLINET/ACTRIS data from 2006-2018). Missing information on composition, size and humidity growth of the aerosol in the models introduces considerable uncertainty to the PM derived extinction, which conservatively spans up to a factor 10 for absolute extinction values. Aeronet data are used to calibrate the conversion from modelled mass to optical property aerosol extinction. This way the order of magnitude in extinction is similar between the models and the lidar profiles, but also significant differences appear at some stations in the lowest layers (Granada, Athens). Relative differences in the form of extinction profiles are more certain. We choose the most representative five stations to compare in retrospective the seasonal average aerosol profiles since 2016. The retrospective of the seasonal comparisons since 2016 shows very similar profiles during this season. The respective overestimation or underestimation of the extinction found in 2016 are usually also found in following years with the ENSEMBLE.

4.2 Introduction

The vertical distribution of aerosol reflects processes like atmospheric mixing, removal, and aerosol transport from outside of the domain or formation of secondary aerosol. The vertical mixing processes determine ground concentrations in polluted areas. Long-range transported aerosol, often carried aloft, may contribute to pollution in clean regions. Evaluation of the simulated aerosol column and vertical profiles are thus valuable for the performance characterisation of air quality models.

The 9 regional models provide mass concentration vertical profiles (PM_{2.5} and PM₁₀) over Europe and may thus be evaluated for their aerosol vertical distribution. However, only very few aircraft campaigns and mountain sites are available to validate aerosol mass at altitude. In contrast frequent measurements of vertical profiles of aerosol backscatter, extinction or its integral, aerosol optical depth, exist. Deriving aerosol optical properties from the mass concentrations is thus needed, assuming lidar ratios and mass extinction coefficients, at least until the models provide more specific output on aerosol composition and optical properties.

In order to assess the mass extinction coefficients chosen, the aerosol optical depth derived from the model mass profiles is first compared to AERONET AOD measurements. Secondly, we document a comparison of the extinction profiles derived from modelled mass concentration with climatological extinction profiles derived from European EARLINET/ACTRIS lidars.

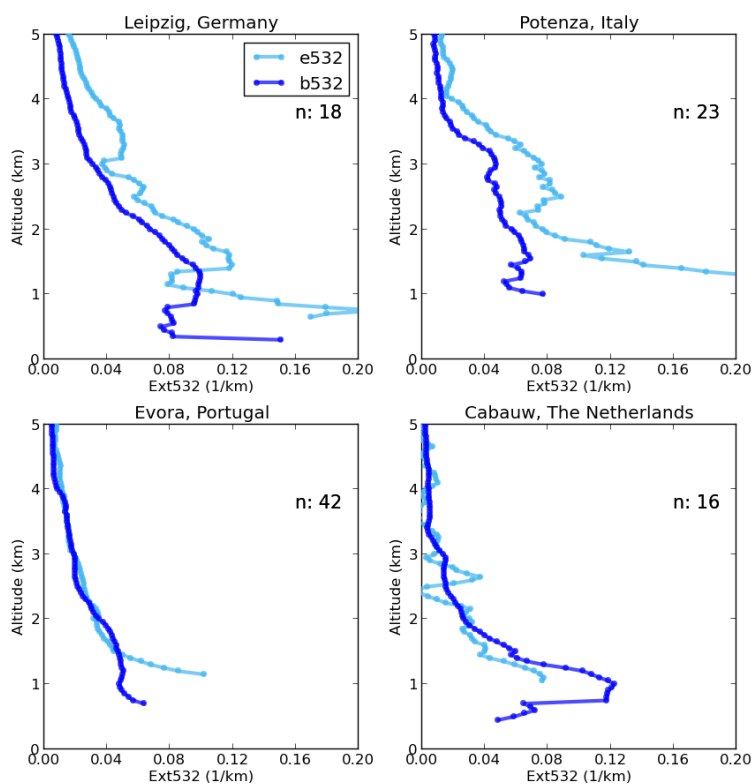


Figure 4.1. Comparison of mean profiles of two aerosol optical properties derived from the same Earlinet Raman lidar at the same time: a) aerosol extinction @532nm (light blue line) and b) extinction @532nm derived from the aerosol backscatter coefficient (dark blue) using a lidar ratio of 50 sr. The profiles use data taken at simultaneous times at each station; number of profiles given as n: x.

4.3 Methodology

AERONET data

The AERONET sun-photometers measure in non-cloudy conditions the aerosol optical depth at several wavelength and in near real time. The spatial distribution of the instruments allows a good coverage of aerosol observation over Europe. The version 3 level 1.5 data has been used in the reporting period presented here. This version and quality level ensures an efficient filtering of the residual clouds (mainly cirrus) for data in near real time. Daily AERONET aerosol optical depth, measured at 550 nm, has been averaged over summer for the European sites available in CAMS model output.

Lidar data

The EARLINET/ACTRIS Lidars are distributed over several locations in Europe and allow comparison across different climates in Europe (Pappalardo et al., 2014). Yet, up to now, there are no near-real-time data available. Regular measurements in EARLINET are sparse and often acquired once per week, with gaps due to maintenance or funding restrictions. A climatology has been computed per station and per season with all measurements available between 2006 and 2018.

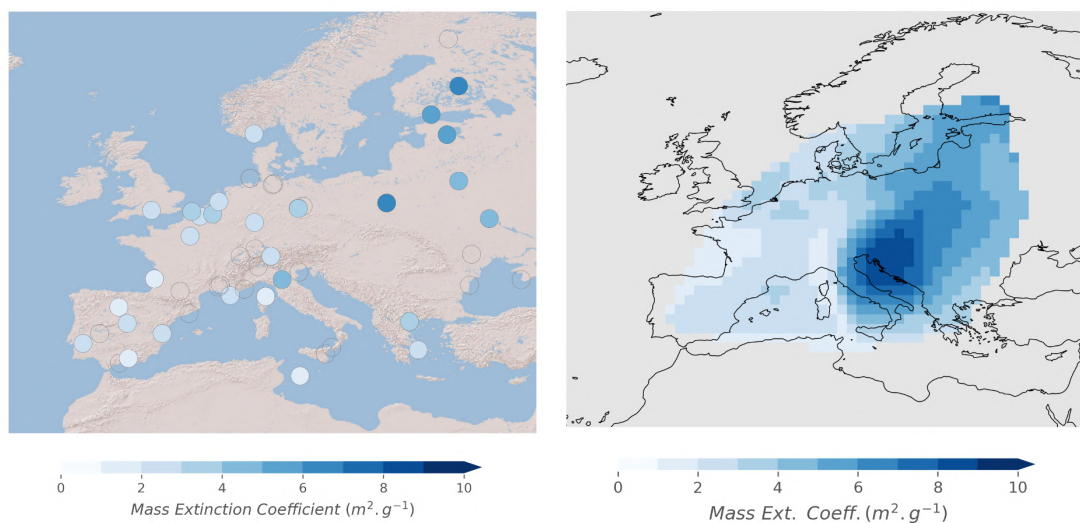


Figure 4.2. Mass extinction coefficient estimated at AERONET location sites (blue dots) and EARLINET stations (triangles) for DJF2021 (left). A European map of the Mass Extinction Coefficient has been constructed with cubic interpolation in the inner part of the region covered by AERONET on the model grid (right), and with the nearest neighbours in the outer part of this region (not shown).

The backscatter coefficient and extinction profiles at 532 nm have been extracted from the EARLINET database. The more frequently measured backscatter profiles are considered here with priority. An aerosol extinction coefficient profile is computed from the backscatter coefficient using a range of plausible lidar ratios. This latter parameter depends on the aerosol type and is more likely decreasing with the size of the aerosol. Minimum values are observed for sea salt aerosol (below 30 sr at 550 nm, [Ackermann et al., 1998, Omar et al., 2009]), while larger values are related to urban particles (55 sr in [Muller et al., 2007], 70 sr in [Cattrell et al., 2005]). Desert dust is associated with intermediate lidar ratios, ranging from 30 sr to 60 sr depending on the sources and the transport regime. A climatology of aerosols in West Africa published in Mortier et al. [2016] revealed an average lidar ratio (over 9 years) of about 30 ± 15 sr. Due to the location of the stations involved in this study, both dust and urban aerosols and any a mix of them might occur. In order to represent the uncertainty on the nature of aerosols, we show the range in likely mean extinction using a lidar ratio extending from 30 to 70 sr.

As a test, the conversion of lidar backscatter to extinction coefficient is performed assuming a constant average lidar ratio of 50 sr at locations where both backscatter and extinction coefficients are measured in EARLINET. This allows consistent comparison and visualization of some of the error associated to our simplified constant lidar ratio assumption. We have excluded from this comparison cases where local extinction coefficient was above 0.5 km^{-1} to avoid outliers. The profiles shown in figure 4.1 from 4 stations reveal an error in the mean profile of 0-30% in extinction, which is small compared to the model spread documented below. The extinction profiles derived from the backscatter coefficients look vertically smoother. We have excluded from this comparison cases where local extinction coefficient was above 0.5 km^{-1} to avoid outliers.

In addition to this aerosol typing uncertainty, a sampling error should be accounted for. The observations are sporadic, while the models predict the aerosol concentration continuously. Therefore, seasonal averages are not computed with the same coverage in model and observation.



Our earlier model-based bootstrap studies revealed, that, depending on the station, a set of ca. 30 daily observations allows reproducing the seasonal average with an error of about 10% [ACTRIS Deliverable WP6/D6.21]. In our case, this error might be larger since the synoptic situation is very different between our EARLINET climatological dataset, covering 2006-2018, and the season covered in this report. An overall uncertainty of about 20% has been chosen to represent the sampling error.

Model data

The ensemble and the underlying 9 regional CAMS models are investigated. For each of these models, the hourly PM₁₀ and PM_{2.5} vertical profiles are extracted at the EARLINET station locations from the first day of each daily forecast at levels 0, 50, 250, 500, 1000, 2000, 3000, 5000 m.

The conversion of PM₁₀ mass concentration to extinction requires a mass extinction coefficient (MEC). MEC depends on the size distribution, refractive index and density of the particles. This information is not yet available from the models. For different kind of aerosols, MEC values can vary from about 0.5 m²g⁻¹ in the case of desert dust aerosols up to 8 m²g⁻¹ for urban particles [Chin et al., 2002]. No variation with height or aerosol type is taken into account, mainly because the models provide no further info on aerosol speciation. We derive the MEC value to convert the model profile data to extinction profiles from a combination of the modelled mass column load and consistent Aeronet AOD data.

For the Aeronet based computation of the MEC, the model data are picked at the location and on the day when sun photometer observations were available. The CAMS-regional mass concentrations have been averaged for coincident days (with the measurements) and averages are converted, with a seasonal and site dependent mass extinction coefficient estimated with AERONET retrievals, into extinction profiles. A seasonal and site dependent mass extinction coefficient is obtained when combining it with AERONET AOD retrievals. Values of MEC are ranging from 1 m²g⁻¹ in South-West of Europe to more than 10 m²g⁻¹ in the North-East. Since some of the EARLINET stations are not co-located with Sun photometers, a European map of MEC has been constructed, for each season, by interpolating (cubic interpolation) and extrapolating (nearest neighbour) the available AERONET based MEC calculations on the grid of the model (figure 4.2). One can notice a gradient with the longitude with lower values found in the Western part while the highest values are observed in the Eastern part of Europe. Also, the values are generally lower as compared to last year, which might reveal a higher concentration in coarse particles (dust). The seasonal AERONET-based MEC is then used at each EARLINET station to calculate the model extinction from concentration profiles. The uncertainty on the MEC being removed allows more accurate comparisons with the observed vertical profiles than using an average MEC over whole Europe.

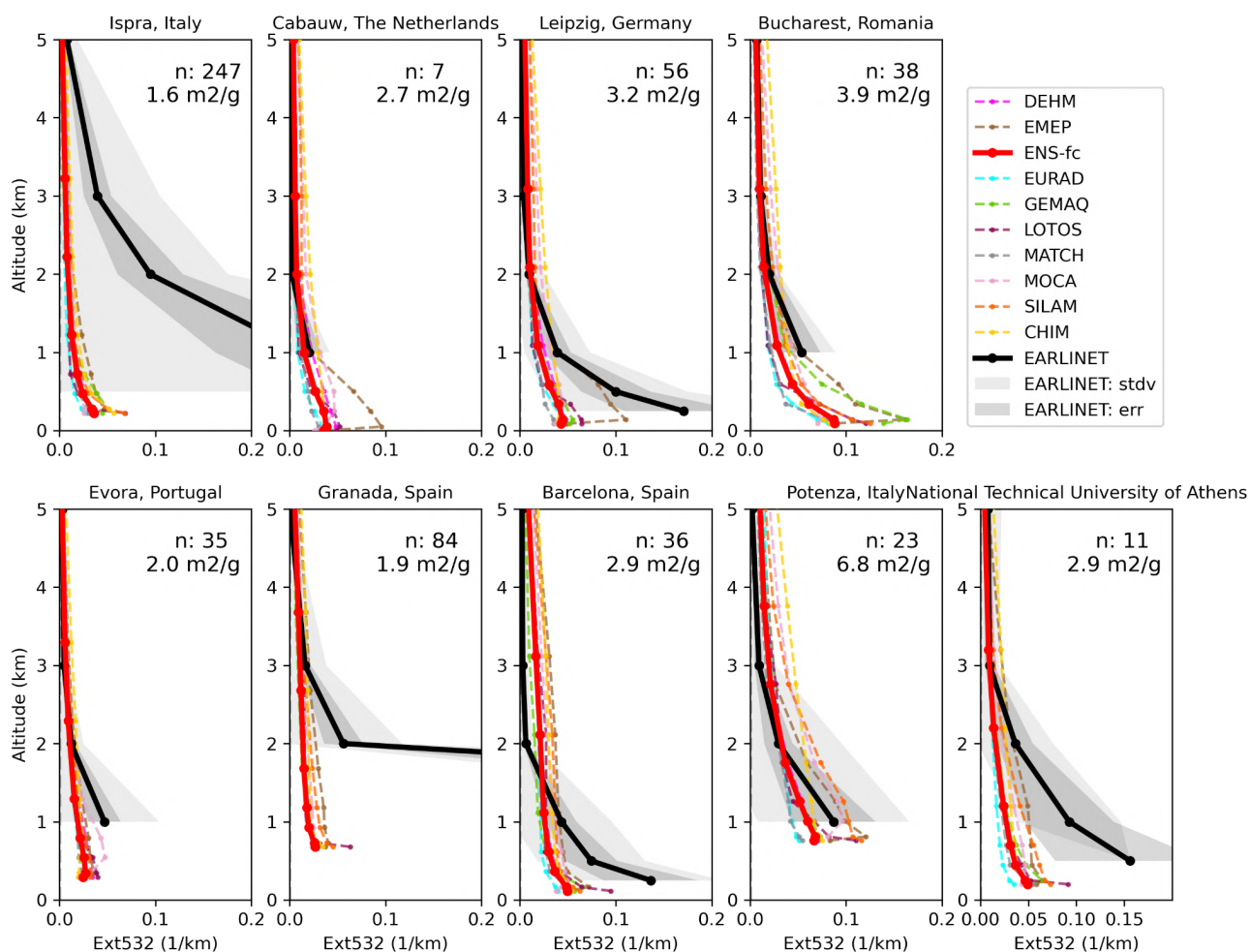


Fig 4.3. Extinction profiles December 2020 - February 2021 derived from the ENSEMBLE forecast mass concentration profiles (red envelope) and from EARLINET (climatology) backscatter profiles (grey envelope: lidar ratio uncertainty, light grey: including sampling error). “n: XX means number of individual EARLINET profiles assembled (December - February 2006-2018). The EMC used for the calculation of the extinction from the concentration profiles is indicated for each station below the number of EARLINET profiles “n” used for the calculation of the climatology.

4.4 Results

4.4.1 Comparison of extinction profiles

The extinction profiles estimated from the 9 CAMS models and the ENSEMBLE and EARLINET measurements are compared for DJF2021 (figure 4.3).

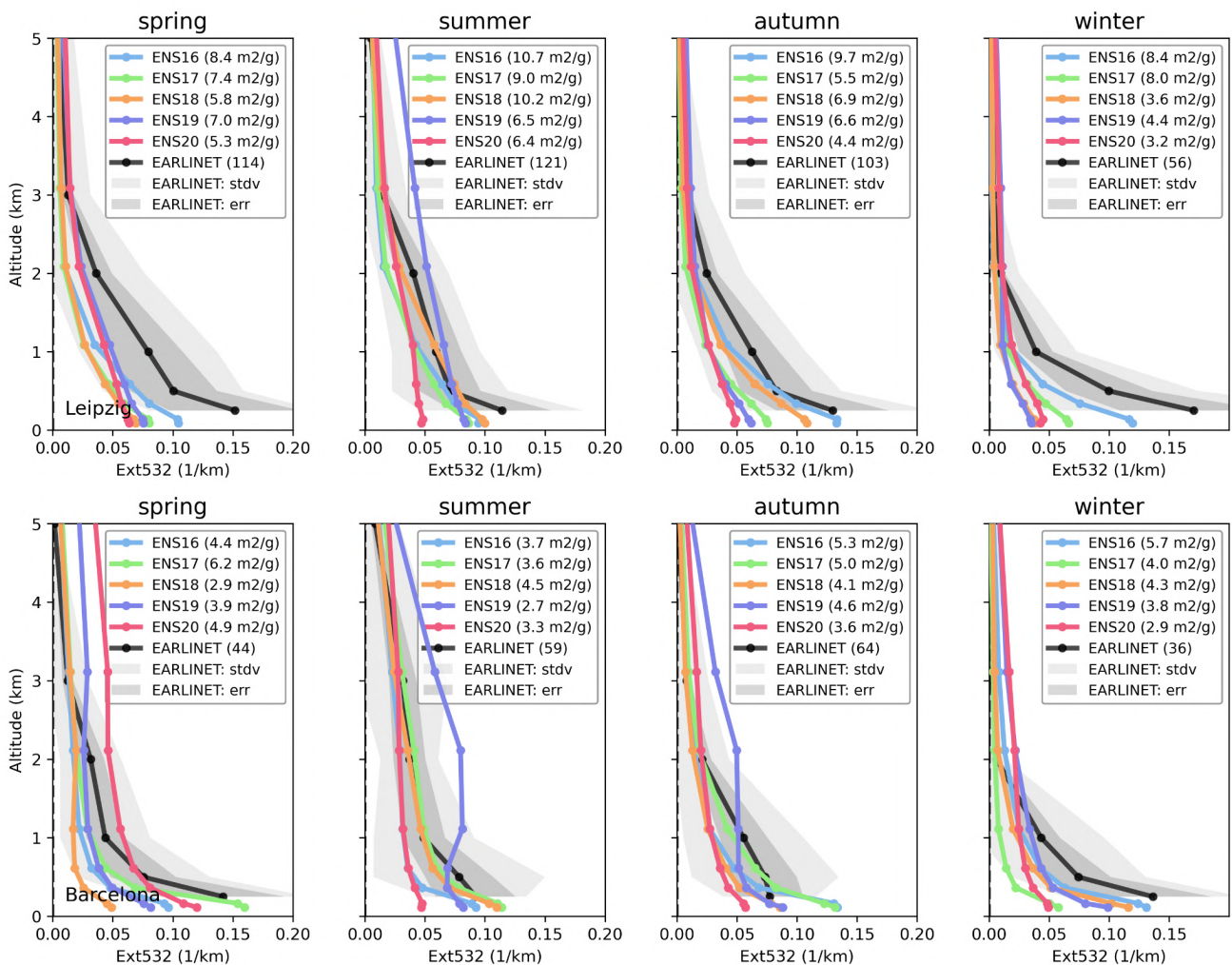
One observes generally a good agreement between the two datasets at the exception of Ispra and Granada where the observation datasets exhibit large extinction values. In Cabauw, a good agreement is found above 1 km of altitude, where the first observation point is available. In the other locations, all of the models show lower as compared to EARLINET climatology values this winter. The



reason is probably due to the relatively low EMC coefficients obtained this season due to the several Saharan dust intrusions observed in February.

4.4.2 Seasonal variability

In order to investigate the performance of the model in reproducing the vertical profiles, it is interesting to observe the inter-annual variability for the different seasons. This will be of use for the development of a score providing an assessment of the models skills, and is also useful to investigate the models synoptic variability. The seasonal profiles have been reported since 2016 at 5 stations (Leipzig, Barcelona, Potenza, Evora and Bucharest) in Figure 4.4, using the new EARLINET climatology. Performances of the models in 2021 look similar to the performances observed in winter 2020. In Potenza, the models are in better agreement with the observations due to the higher values, which can be explained by the higher EMC values obtained this year. At other locations, due to lower EMC, the negative bias is usually slightly increased as compared to last year.



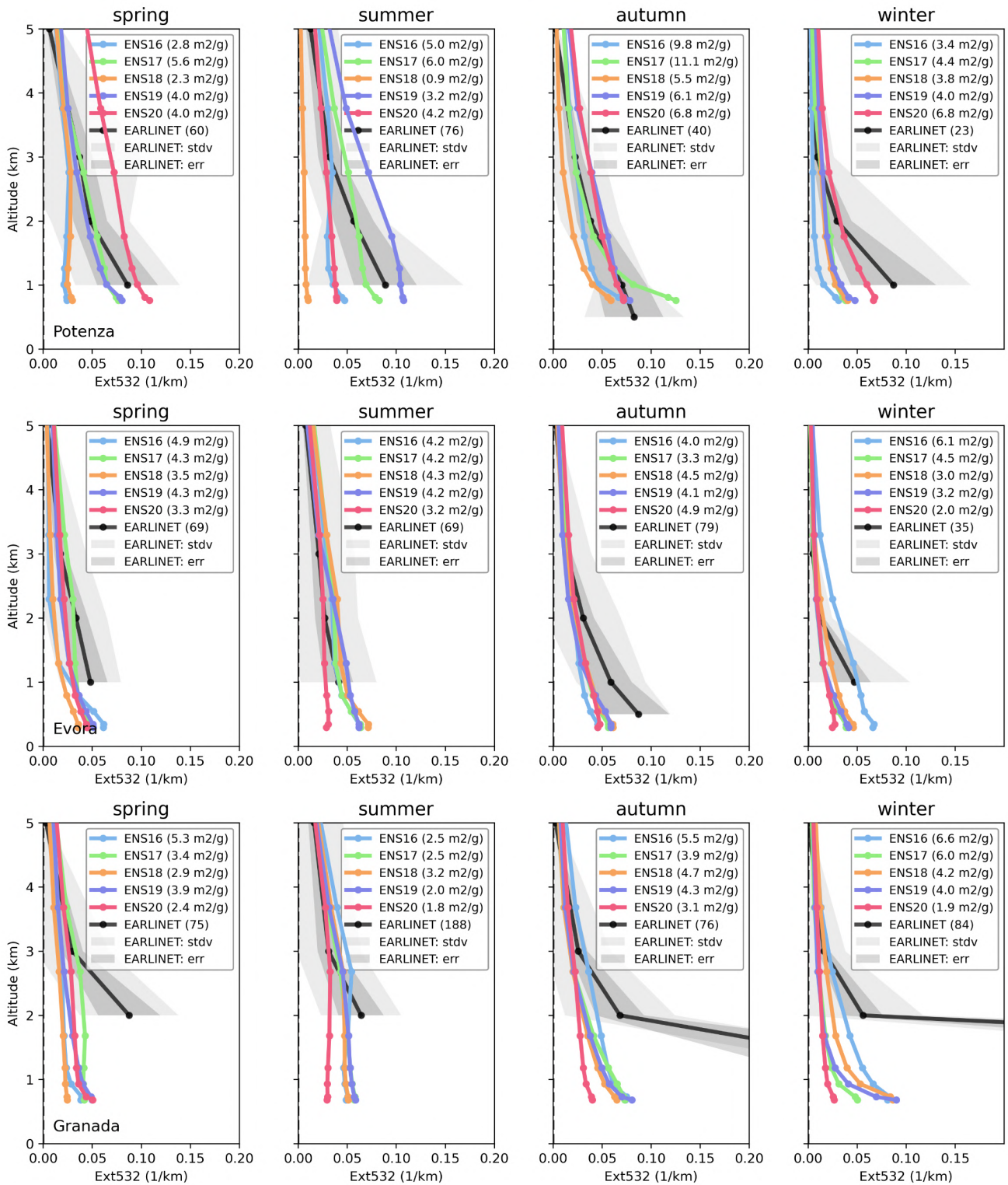


Fig 4.4: Seasonal extinction profiles derived from ENSEMBLE forecast mass concentration profiles for 2016 (ENS16), 2017 (ENS17), 2018 (ENS18), 2019 (ENS19), 2020 (ENS20) and EARLINET climatology. The parenthesis indicates for the CAMS profiles, the MEC used for the extinction estimation, and for the EARLINET, the number of profiles used for constructing the climatology.



5 IAGOS aircraft CO and O₃ profile comparisons

5.1 Summary

Routine observations of ozone and CO over European airports are available from the IAGOS fleet. Take-off and landing profiles were sampled from the hourly model 3D forecasts along the flight tracks.

Ozone in-situ: In the lowest layers, ozone is well represented by the models with small overestimations and sometimes a better performance from the regional ensemble especially when ozone values are high with respect to climatology. The bias in the free troposphere is larger with overestimations from both models.

Carbon monoxide: CO is mostly underestimated in all layers by the regional ENSEMBLE and CAMS global which present similar behaviour in the low troposphere. In the free troposphere the agreement is better for both models and CAMS-global performs better than the regional ENSEMBLE.

IAGOS Validation Method

Validation is possible at the European airports visited by the IAGOS fleet. For the European-based carriers, there are regular profiles at the home airports. There are two aircraft operated by Lufthansa, one operated by Air France. Thus, when the fleet is fully operational, there are daily profiles Frankfurt and Paris (CDG). IAGOS is also installed on two aircraft operated by the Asian-based carrier China Airlines. Aircraft fly regularly from Taipei to Amsterdam or Vienna and sometimes to Rome. Other airports may be visited depending on the operational schedules of the airlines. Due to persisting restrictions associated to the COVID-19 crisis, IAGOS operations still remain impacted in this period although more observations are available compared to other quarters of 2020. Over Europe only the airport of Frankfurt has been visited as shown in Figure 5.1.

We download the daily latitude-longitude datasets for the 7 regional models and the ENSEMBLE for two species (carbon monoxide and ozone) on 8 vertical levels (surface, 50m, 250m, 500m, 1000m, 2000m, 3000m, 5000m). The aircraft takes about 10 minutes to climb or descend the 5000m vertical extent covered by the regional models. During this time and travelling at up to 166 m s⁻¹, it covers about 120km and therefore traverses many grid-boxes of resolution 10km. We perform a spatial interpolation from the grid of the regional models to the aircraft's trajectory. The IAGOS measurements in ppbv are converted to µg m⁻³ using the temperatures measured by IAGOS. The data are validated by the PI but are not yet calibrated. Calibration takes place after an operational period of about 6 months.

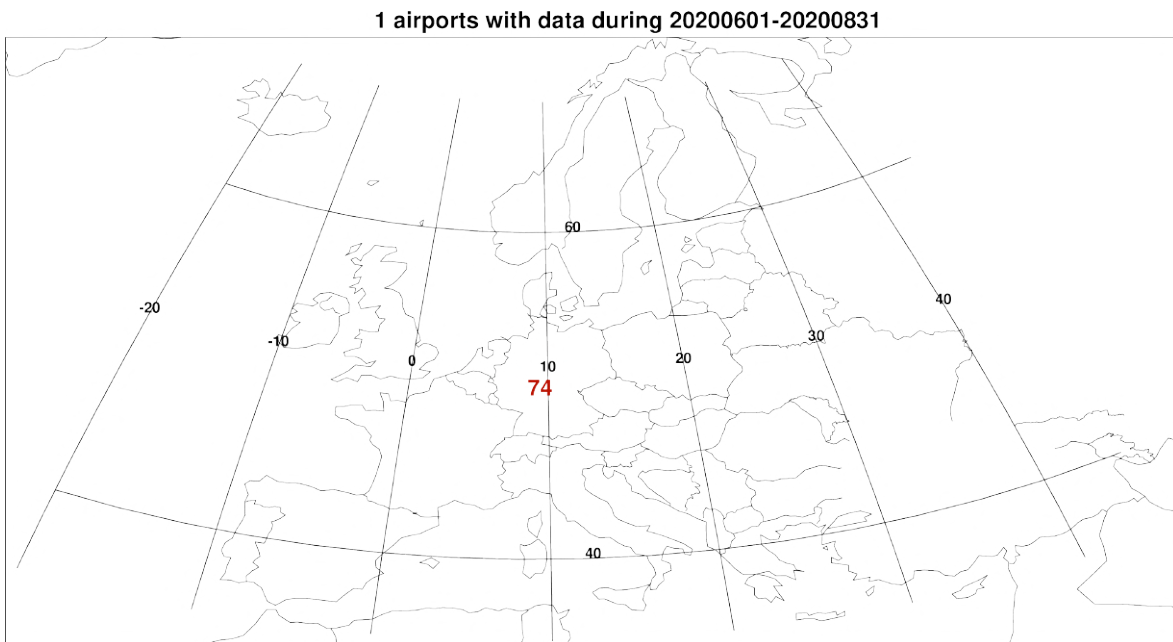


Figure 5.1. Map showing the number of profiles available in the period December 2020 - February 2021.

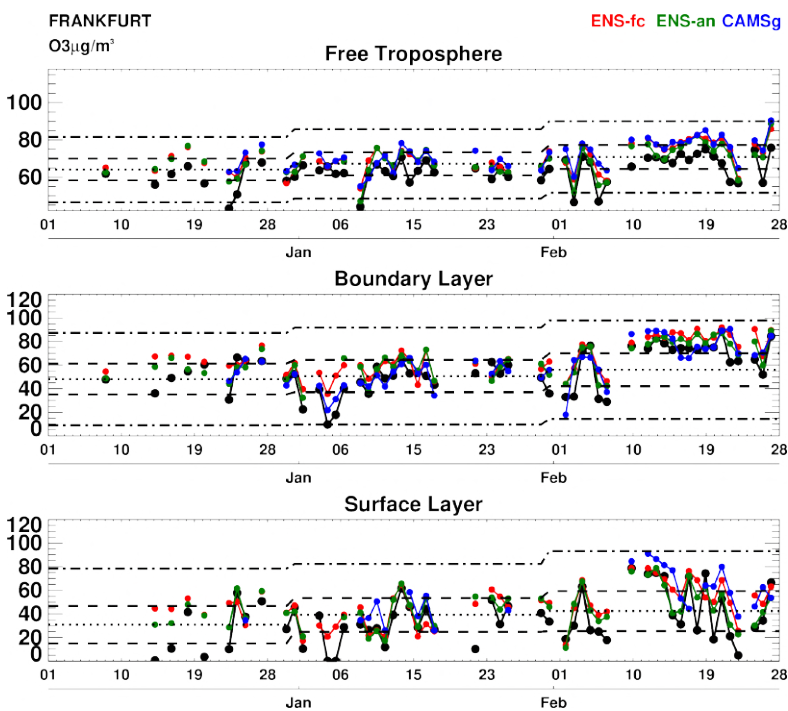


Figure 5.2: Daily time series of ozone at Frankfurt for the period DJF 2021. IAGOS observations are shown in black, the regional ENSEMBLE and associated analysis are shown in red and green respectively, and CAMS-global (o-suite) is shown in blue. The black dotted line is the monthly mean of the observations over the period 2003-2016 (IAGOS/MOZAIC, Level 2 data), the black dashed line shows 1 standard deviation from the monthly mean and the black dotted-dashed line shows 3 standard deviations from the monthly mean. (Units: $\mu\text{g}\cdot\text{m}^{-3}$).

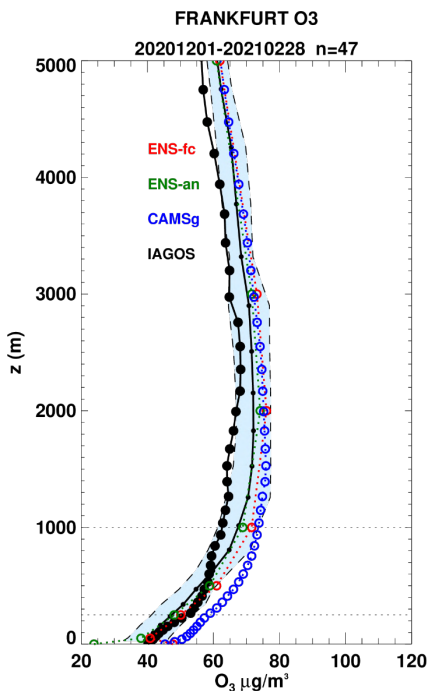


Figure 5.3: Mean profile of ozone at Frankfurt for the period DJF2021. IAGOS observations are shown in black, the regional ENSEMBLE and associated analysis are shown in red and green respectively, and the global o-suite is shown in blue. The shaded area indicates the range of the mean climatology of the observations plus/minus one standard deviation during the same period for all years between 2003 and 2016 (IAGOS/MOZAIC, level 2 data). (Units: $\mu\text{g}\cdot\text{m}^{-3}$).

5.2 IAGOS Ozone

For the period December 2020 - February 2021, observations are available only at Frankfurt with discontinuities: poor sampling in the second half of December and gaps at the end of January and second week of February. The daily time series are presented in Figure 5.2 and associated averaged profiles over the whole period are presented in Fig. 5.3. In all plots, the forecast and analysis of the regional ENSEMBLE is shown in red and green respectively, and CAMS-global analysis is shown in blue. Fig. 5.3 shows that the regional and global models differ mostly from the surface up to 1 km. On average, in the lowest layers the ENSEMBLE performs better than CAMS global presenting small overestimations (Fig. 5.3). In the upper layers, the regional and global models behave more similarly and mostly overestimate (Fig. 5.3).

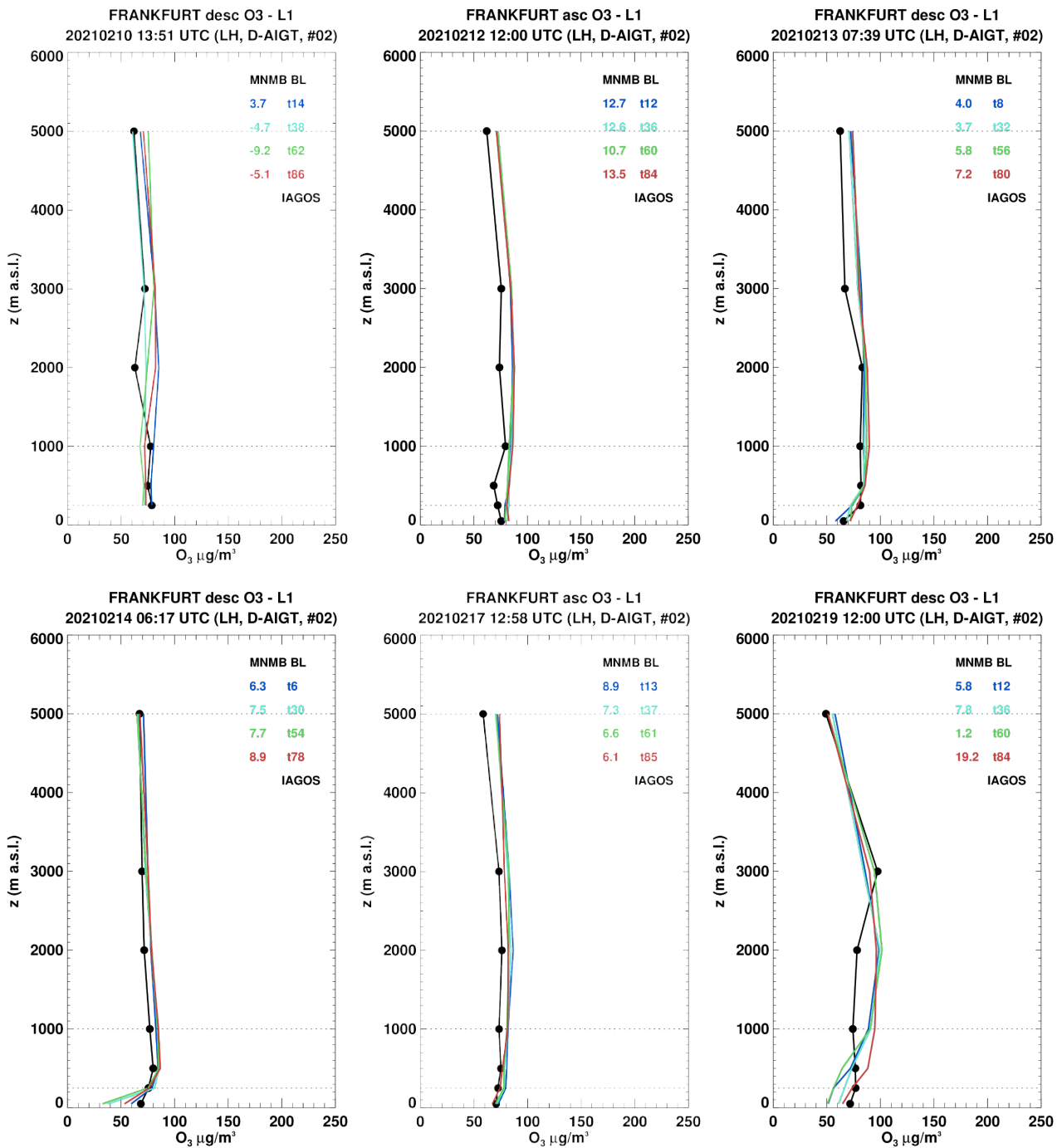


Figure 5.4.a. Selection of ozone profiles at Frankfurt during the period DJF 2021. IAGOS is shown in black and the ENSEMBLE is shown at 4 forecast times (blue: 1-day; cyan: 2-day; green: 3-day; red: 4-day). (Units: $\mu\text{g m}^{-3}$).

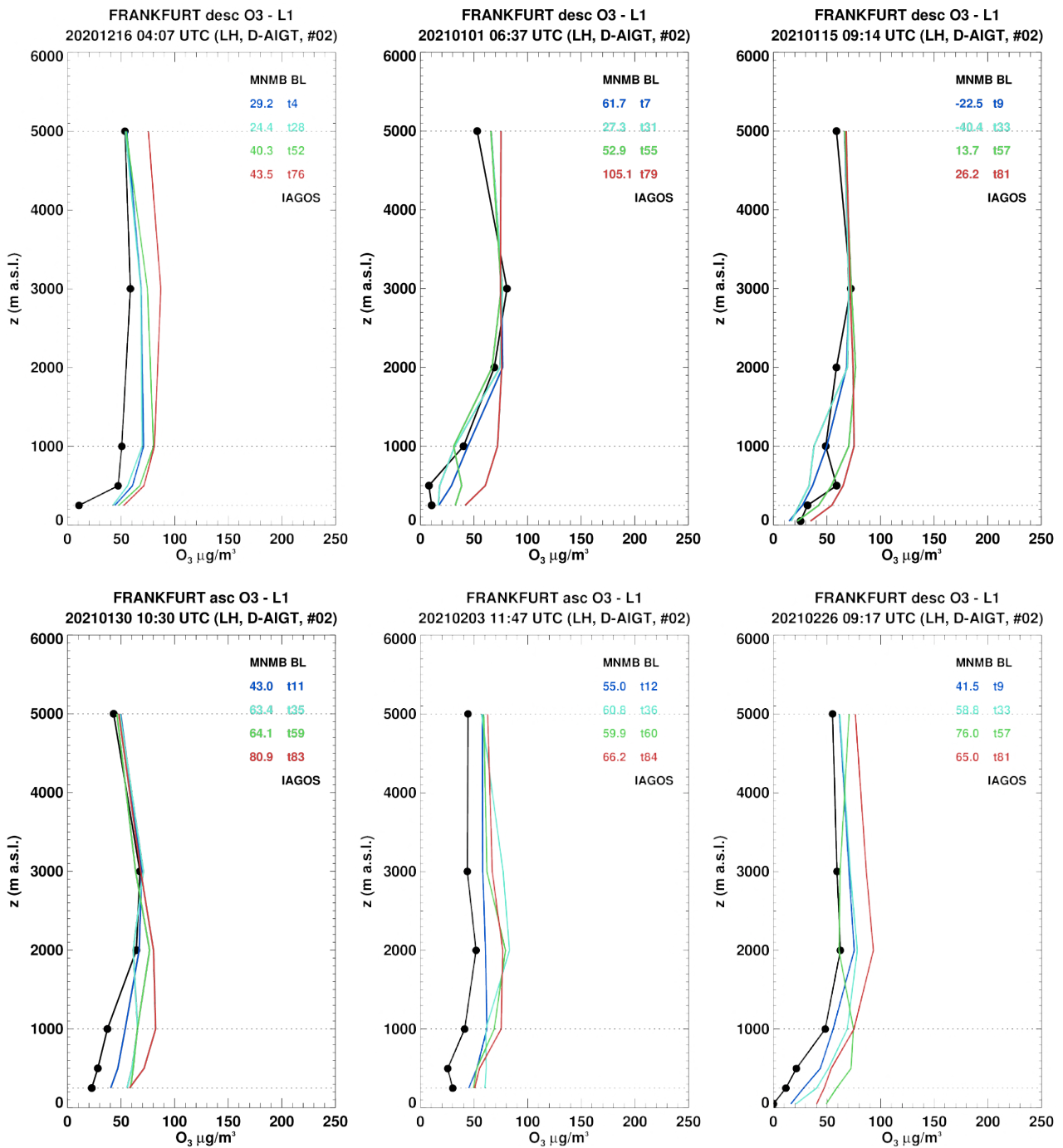


Figure 5.4.b. Selection of ozone profiles at Frankfurt during the period DJF2021. IAGOS is shown in black and the ENSEMBLE is shown at 4 forecast times (blue: 1-day; cyan: 2-day; green: 3-day; red: 4-day). (Units: $\mu\text{g m}^{-3}$).

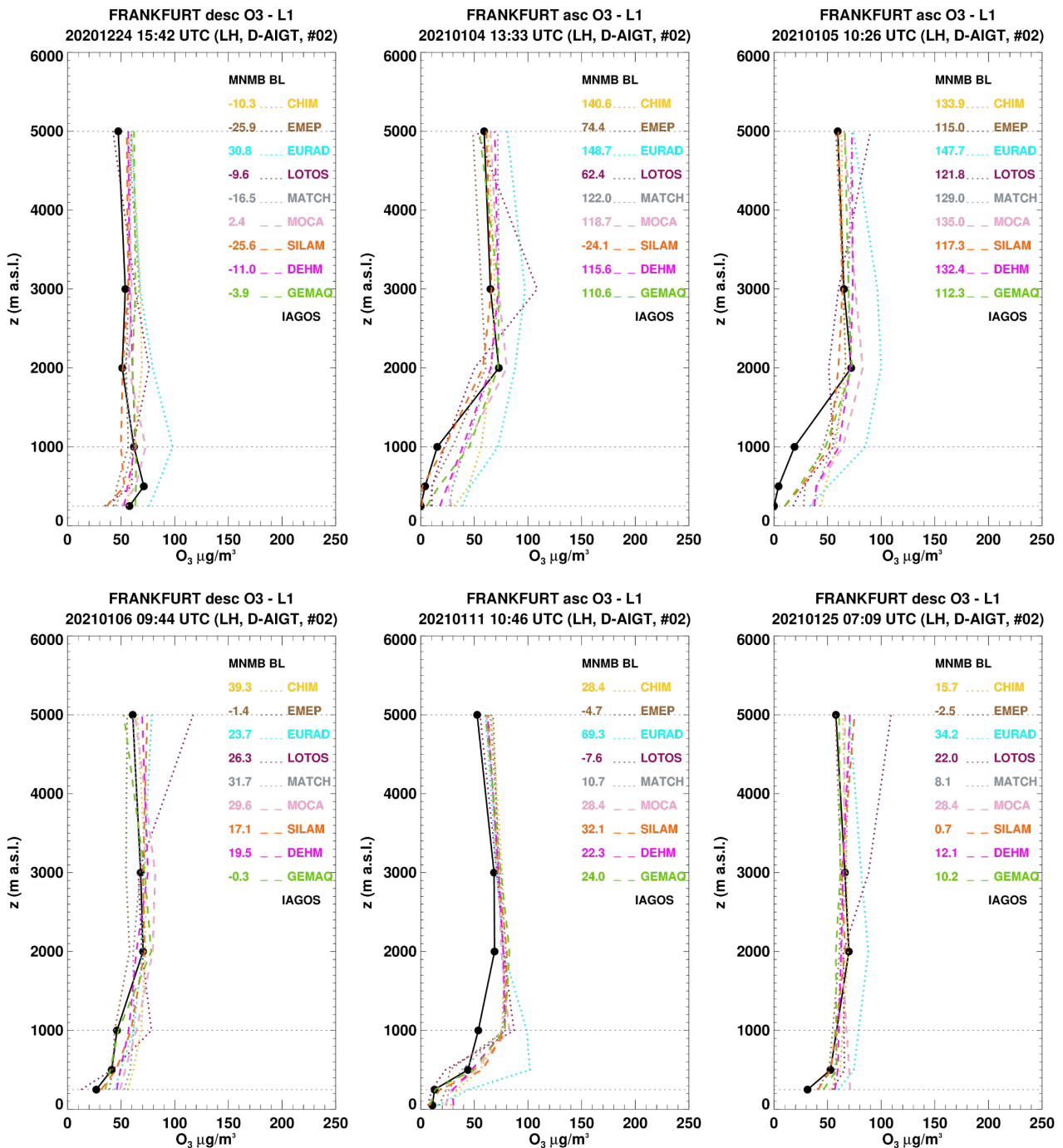


Figure 5.5.a: Ozone profiles at Frankfurt during the period DJF 2021. IAGOS is shown in black and each colour/Line style corresponds to one of the 9 models for the ensemble. In the legend, the models are CHIM=CHIMERE, EMEP=EMEP, SILAM=SILAM, LOTOS=LOTOS-EUROS, MOCA=MOCAGE, EURAD=EURAD, MATCH=MATCH, DEHM=DEHM, GEMAQ=GEM-AQ. (Units: $\mu\text{g m}^{-3}$).

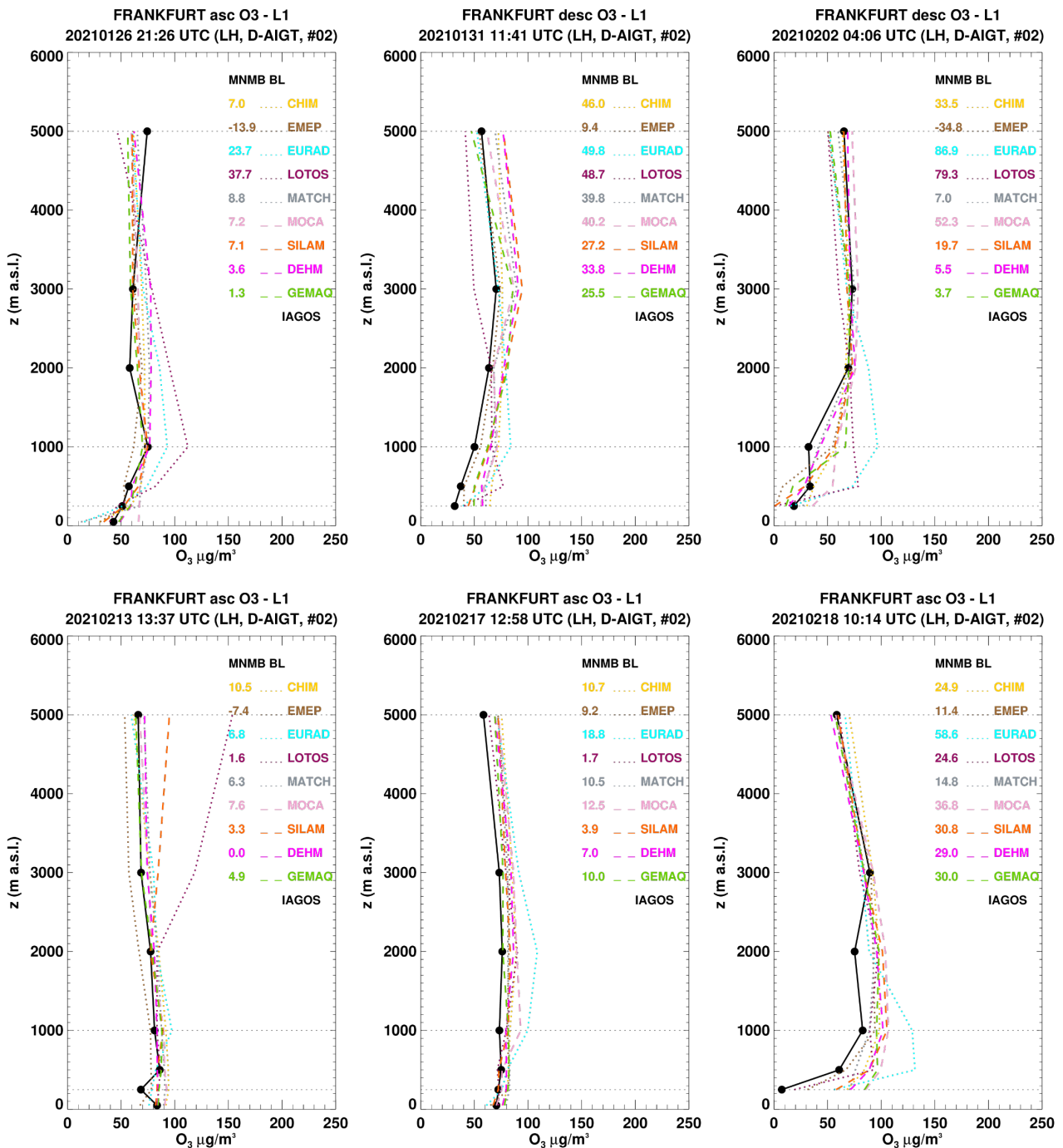


Figure 5.5.b: Ozone $\mu\text{g}/\text{m}^3$ profiles at Frankfurt during the period DJF 2021. IAGOS is shown in black and each colour/Line style corresponds to one of the 9 models for the ensemble. In the legend, the models are CHIM=CHIMERE, EMEP=EMEP, SILAM=SILAM, LOTOS=LOTOS-EUROS, MOCA=MOCAGE, EURAD=EURAD, MATCH=MATCH, DEHM=DEHM, GEMAQ=GEM-AQ. (Units: $\mu\text{g}/\text{m}^3$).

During this DJF period, IAGOS ozone observations in the surface and boundary layer are mostly in the range of minus one standard deviation from the climatological mean until about the end of the first week of February. Between 10 and 22 February several peaks in ozone between one and three standard deviations occur in the surface layer. In the boundary layer this increase is clearer as ozone values remain above one standard deviation during the entire aforementioned period. Compared to



the 1991-2020 reference average the month of February 2021 in Europe was substantially warmer over southern part and conversely colder over the northern (<https://climate.copernicus.eu/surface-air-temperature-february-2021>), however some parts of Europe have met important variations of temperatures during the month of February 2021 with a change in the prevailing atmospheric circulation pattern (<https://public.wmo.int/en/media/news/extreme-weather-hits-usa-europe>). Germany have experienced a rapid and record change from a cold and icy first half of February to an unusual spring warmth (https://gcos.dwd.de/EN/press/press_release/EN/2021/20210226_the_weather_in_germany_in_february_2021.html?nn=732034). In the second week of February, monthly records were broken with temperature falling below -20°C at many locations, and heavy snow over low elevations. Within a few days, this was followed by unusually mild weather in central Europe. Large and record temperature differences were recorded in the space of less than a week according to DWD. This observed increase in ozone is likely related to the increased production of ozone with this change to warm and sunny weather mentioned before. This increase is well captured by the regional and global models with slightly larger overestimations from the o-suite in the surface layer (Fig. 5.2). A number of profiles illustrating this increase are presented in Fig. 5.4.a between days 10 and 19 February, with nearly constant shape and ozone values of about $70 \mu\text{g}\cdot\text{m}^{-3}$ in the surface and boundary layer. It can be noted that all forecast times present similar results for these cases, while low ozone values sometimes but not systematically show disparities between the different forecasts (Fig. 5.4.b), with the best agreement provided by the closest forecast times. For most profiles of the period the results from all models are similar. The profiles in which the results of the individual models differ are presented in Fig. 5.5.a-b. The EURAD and LOTOS-EUROS sometimes present much larger bias than all other models in the boundary layer and in the free troposphere. In some cases these two models present similar behaviour with similarly shaped profiles presenting large overestimations in the same regions of the atmosphere (0126 21:26, 0131 11:41, 0202 4:06) but not systematically. The results of the LOTOS-EUROS model also show overestimations above 3000m which are not observed in any other model (0106 9:44, 0125 7:09 and 0213 13:37).

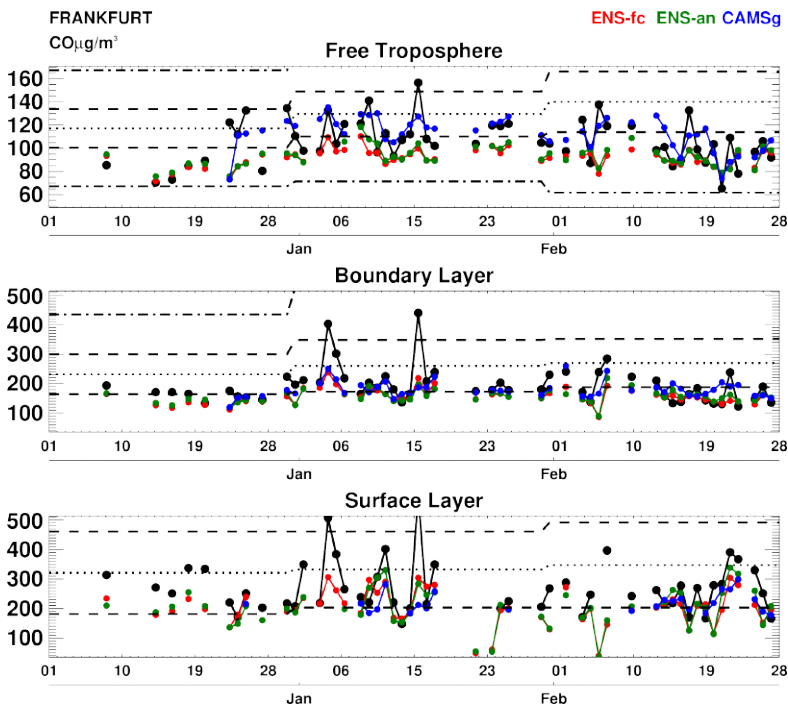


Figure 5.6: Daily time series of CO at Frankfurt for the period DJF 2021. IAGOS observations are shown in black, the regional ENSEMBLE and associated analysis are shown in red and green respectively, and CAMS-global (o-suite) is shown in blue. The black dotted line is the monthly mean of the observations over the period 2003-2016 (IAGOS/MOZAIC, Level 2 data), the black dashed line shows 1 standard deviation from the monthly mean and the black dot-dashed line shows 3 standard deviations from the monthly mean. (Units: $\mu\text{g}\cdot\text{m}^{-3}$).

5.3 IAGOS Carbon Monoxide

Like for the ozone section, the daily time series of CO and associated averaged profile at Frankfurt are presented in Fig. 5.6 and 5.7. The forecast and analysis of the regional ENSEMBLE is shown in red and green respectively, and CAMS-global is shown in blue. On the time series, available observations of CO in the lowest layers are mostly below the climatological values and often reaching values below one standard deviation from the mean. Only a number of values are close to the climatological mean in the surface layer, and two peaks are found in the surface layer where values are above one standard deviation above the climatology both in January around day 4 and 15 (Fig. 5.6 and Fig. 5.8).

In the surface and boundary layers CO is always underestimated by both regional and global models (Fig 5.6) with on average a better agreement of the regional close to the surface and more similar performance for both models above 200 m (Fig. 5.7). In the free troposphere, the agreement is better than in the lowest layers with smaller underestimations (Fig. 5.6 and 5.7), and from the time series CAMS-global is mostly performing better than the regional ENSEMBLE (Fig. 5.6).

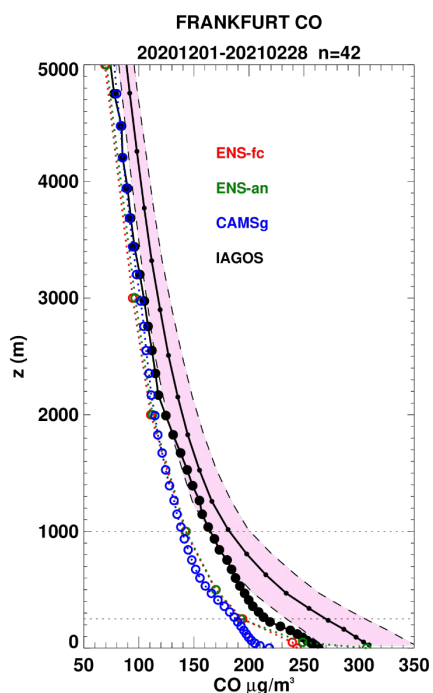


Figure 5.7: Mean profile of ozone at Frankfurt for the period DJF 2021. IAGOS observations are shown in black, the regional ENSEMBLE and associated analysis are shown in red and green respectively, and the global o-suite is shown in blue. The shaded area indicates the range of the mean climatology of the observations plus/minus one standard deviation during the same period for all years between 2003 and 2016 (IAGOS/MOZAIC, level 2 data). (Units: $\mu\text{g}\cdot\text{m}^{-3}$).

Some of the individual profiles at Frankfurt of the ENSEMBLE are presented in Fig. 5.8. These profiles correspond to the highest CO values observed in the low troposphere during this DJF period. The highest observed values of CO in the surface are roughly between 400 and $600 \mu\text{g}\cdot\text{m}^{-3}$ with the maximum reached on 15 January at 09:14 (Fig. 5.8.a). As shown on Fig. 5.8, these CO values are in most cases largely underestimated by the regional model in the lowest layers. In general, there is little difference between the different forecast times except for some cases in the boundary layer when CO values are the highest, which occurs for example on 15 January.

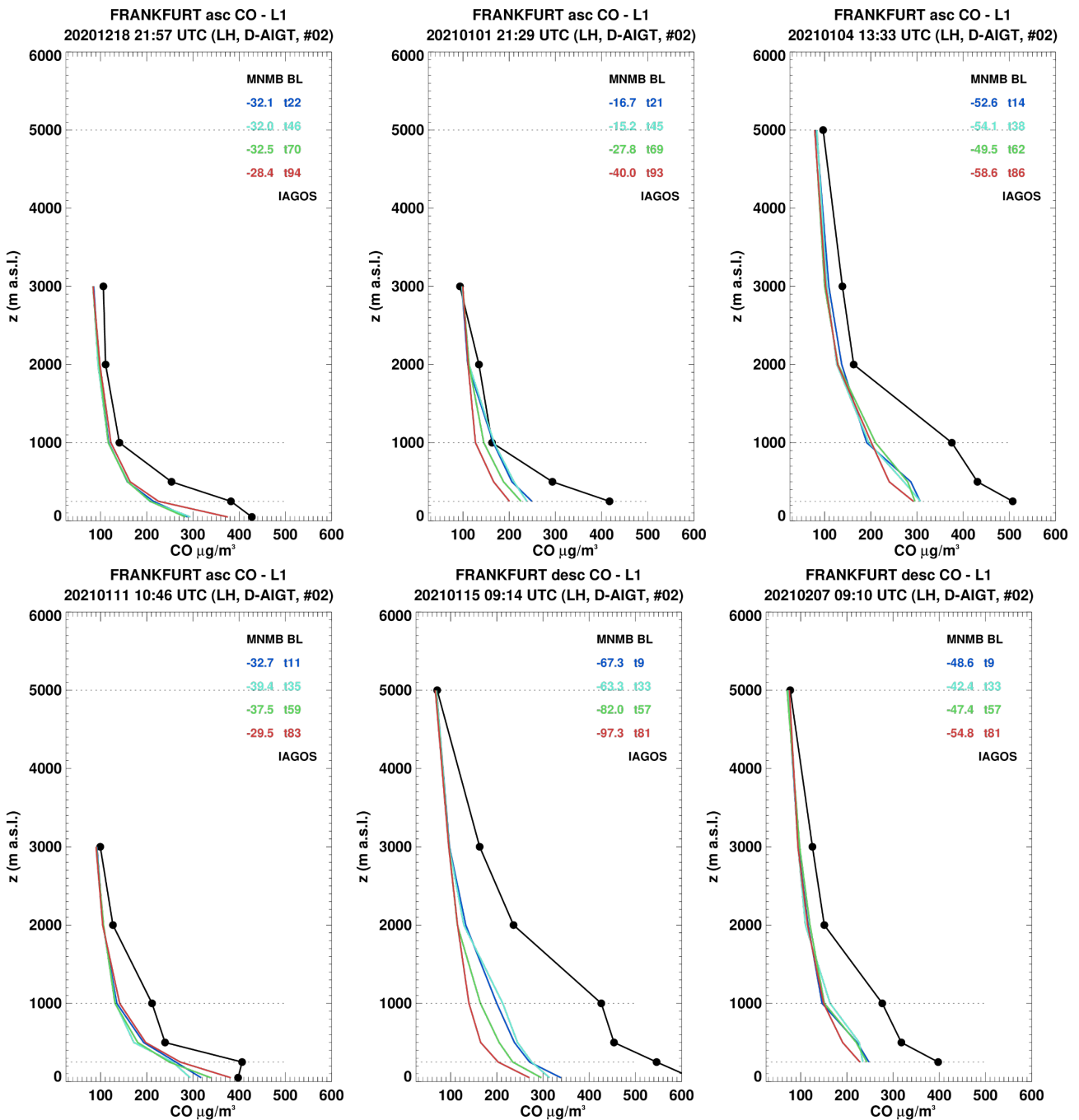


Figure 5.8.a: Selection of CO profiles at Frankfurt during the period DJF 2021. IAGOS is shown in black, and the ENSEMBLE is shown at 4 forecast times (blue: 1-day; cyan: 2-day; green: 3-day; red: 4-day). (Units: $\mu\text{g}/\text{m}^3$).

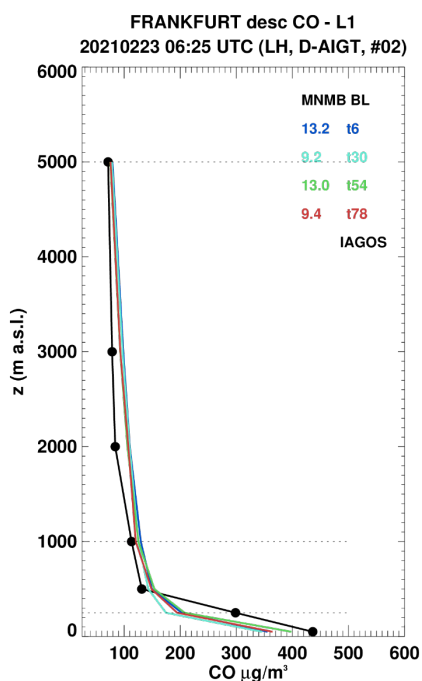


Figure 5.8.b: Selection of CO profiles at Frankfurt during the period DJF 2021. IAGOS is shown in black and the ENSEMBLE is shown at 4 forecast times (blue: 1-day; cyan: 2-day; green: 3-day; red: 4-day). (Units: $\mu\text{g}\cdot\text{m}^3$).

As regards the results of the individual models (Fig. 5.9.a-b), there are in general very similar for all profiles of DJF 2021. Most differences between models are found in the low to mid-troposphere when values of CO are high. High CO values close to the surface are in general better reproduced by the GEMAQ model compared to other models. Besides, the EURAD model often presents the largest underestimations in the boundary layer among all models. Nevertheless, when CO reaches extreme values (>400 ppbv) such as it is the case on 15 January 2021 all models underestimate the observations.

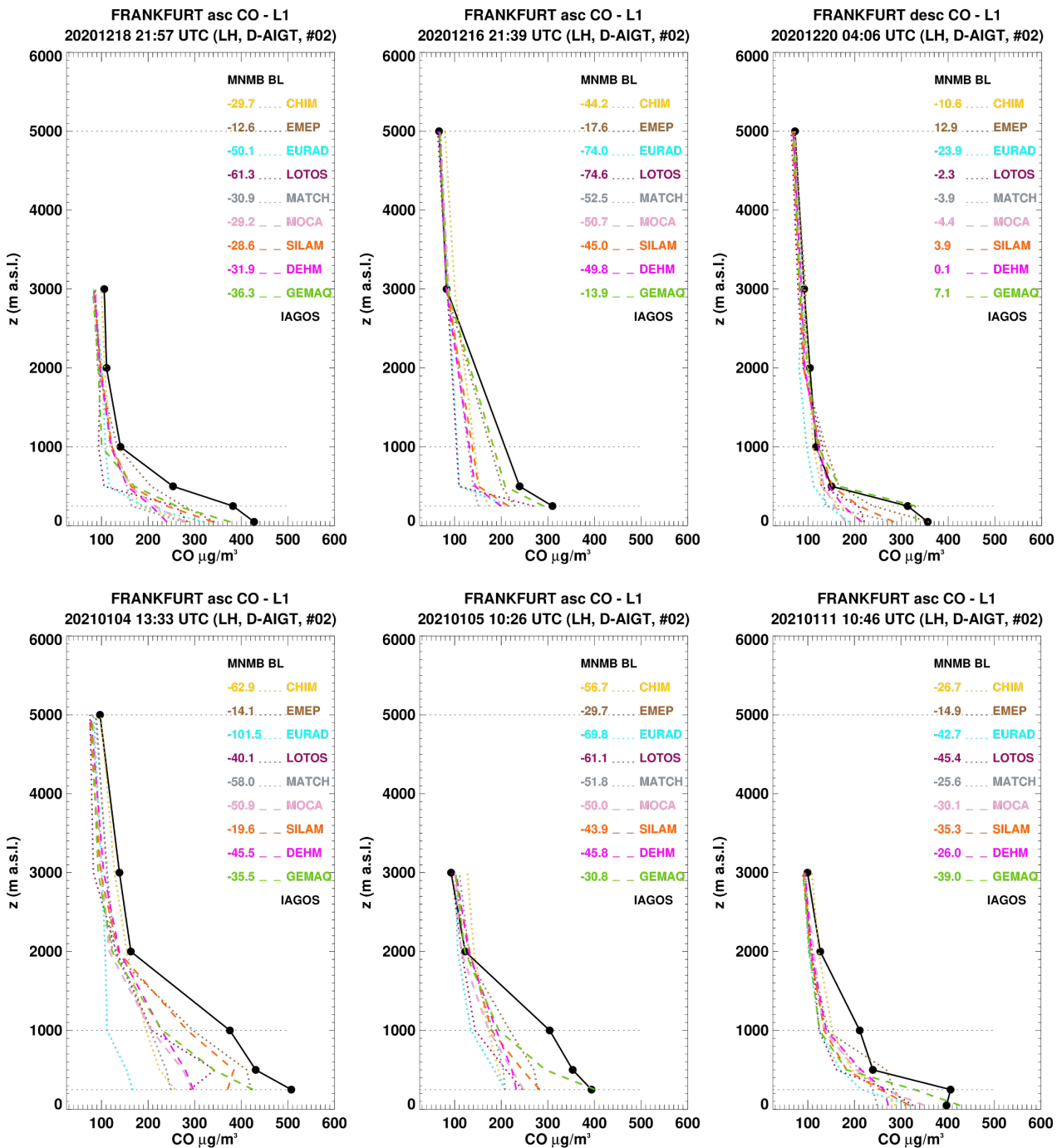


Figure 5.9.a: CO profiles at Frankfurt during the period DJF 2021. IAGOS is shown in black and each colour/ line style corresponds to one of the 9 models for the ensemble. In the legend, the models are CHIM=CHIMERE, EMEP=EMEP, SILAM=SILAM, LOTOS=LOTOS-EUROS, MOCA=MOCAGE, EURAD=EURAD, MATCH=MATCH, DEHM=DEHM, GEMAQ=GEM-AQ. (Units: $\mu\text{g}\cdot\text{m}^3$).

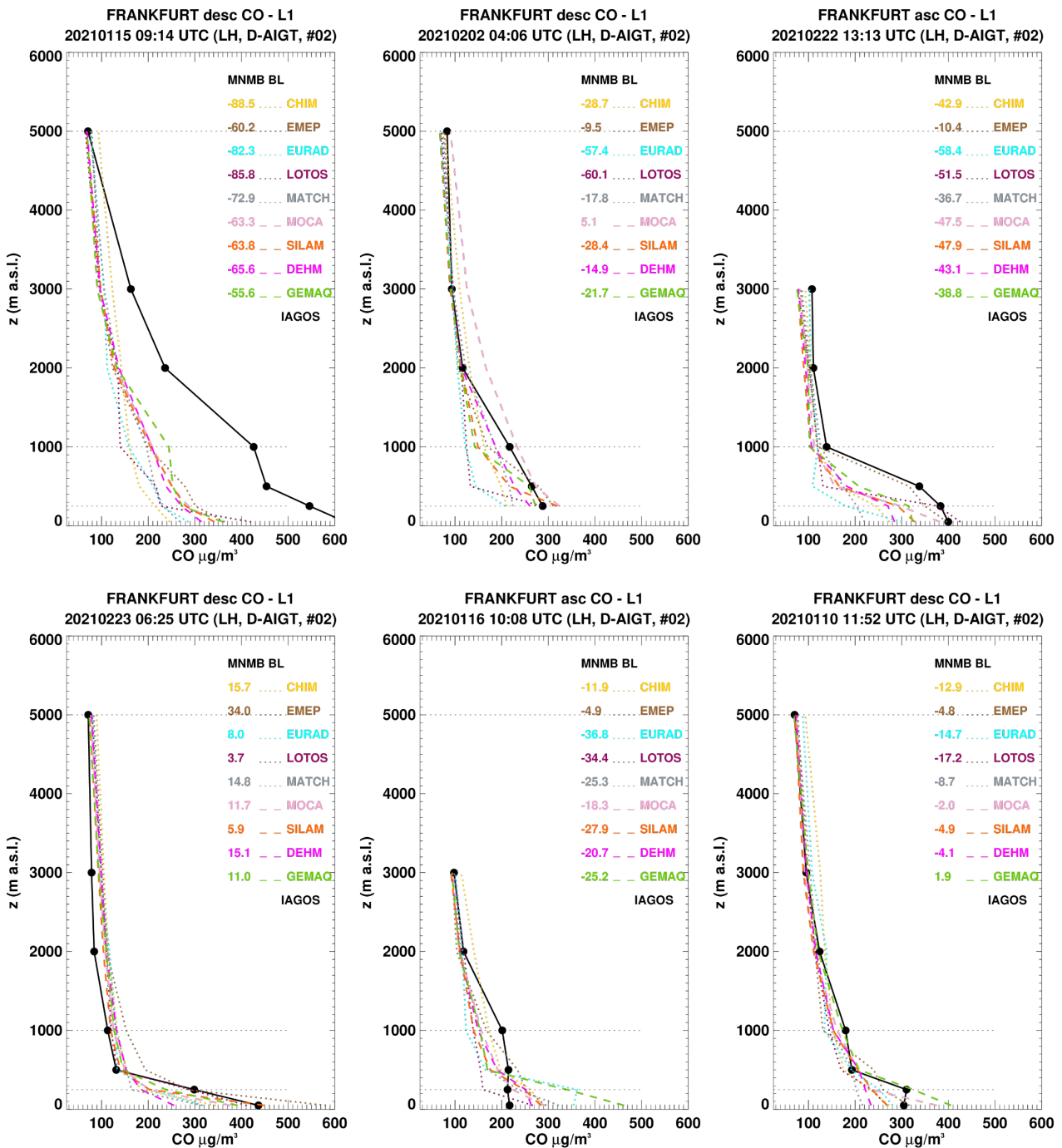


Figure 5.9.b: CO profiles at Frankfurt during the period DJF 2021. IAGOS is shown in black and each colour/ line style corresponds to one of the 9 models for the ensemble. In the legend, the models are CHIM=CHIMERE, EMEP=EMEP, SILAM=SILAM, LOTOS=LOTOS-EUROS, MOCA=MOCAGE, EURAD=EURAD, MATCH=MATCH, DEHM=DEHM, GEMAQ=GEM-AQ. (Units: $\mu\text{g}\cdot\text{m}^{-3}$).



6 Validation of regional model tropospheric NO₂ using MAX-DOAS

6.1 Summary

MAX-DOAS surface remote sensing observations provide tropospheric columns of NO₂, with the largest sensitivity in the boundary layer. While the magnitude of VCDs derived from the measurements for the urban stations De Bilt and Bremen are reproduced well by the model ENSEMBLE, the ENSEMBLE underestimates the values in Athens (bias $\sim -2 \times 10^{15}$ molec. cm⁻²). Large differences can be found between individual models depending on the station. Although many of the model simulated values probably fall within the uncertainty range of MAX-DOAS retrievals, the latter alone cannot explain differences between retrievals and simulations, especially those found for variations in time. For the regional ENSEMBLE, moderate correlations on the order of 30-65 % are found for each station. The regional ENSEMBLE performs significantly better than the global model in terms of correlation at all stations except for De Bilt (where correlations are comparable). However, some of the larger NO₂ values inside individual pollution plumes are underestimated by the models.

6.2 Introduction

MAX-DOAS observations of atmospheric composition are performed by taking measurements of the scattered sunlight at different elevation (and sometimes also azimuthal) angles. Depending on the viewing angle and solar position, the light path through the atmosphere is different, with the observation in the zenith direction usually providing the shortest light path through the lower troposphere. Therefore, using the zenith measurement as intensity of incident radiation and the observations in other angles as intensity of transmitted radiation, the total amount of molecules of a certain species along the light path difference, the so-called slant column densities, can be determined using Lambert Beer's law. Using radiative transfer modelling and Optimal Estimation techniques, this can be inverted to tropospheric columns and even lower altitude tropospheric profiles.

The advantage of MAX-DOAS measurements is their ability to observe several pollution related species at the same time (e.g., NO₂, HCHO, CHOCHO, SO₂, aerosols, potentially also O₃) and to provide data which is virtually free of interferences from other species such as PAN or NO_y for NO₂. Also, the fact that the observations integrate over a comparatively large volume can be an advantage for satellite and model validation as the observed quantity is relatively close to the modelled one. On the other hand, the uncertainty of the retrievals is considerable (on the order of 30% for NO₂ tropospheric columns and larger for individual layers) and depends on cloud occurrence and aerosol loading.

In this report, regional air quality model forecasts of tropospheric NO₂ columns are compared to MAX-DOAS retrievals from 3 urban stations (De Bilt – KNMI, Bremen – IUP-UB, Athens – IUP-UB). The reader is referred to previous reports for comparisons from the rural station OHP (BIRA-IASB) (which showed in general an underestimation by the model ENSEMBLE and an overall better performance for CAMS-global here) as the instrument at this site stopped working in March 2017. The MAX-DOAS instrument for the urban site in Uccle (BIRA-IASB) was dismantled in March 2020, and comparisons for this station are therefore not continued since this time. Since the MAM 2020 report, the MAX-DOAS data used in the comparisons is based on tropospheric column retrievals, meaning that no profile information is



incorporated in the comparisons since this time. A simple block profile is assumed in the MAX-DOAS retrievals and column averaging kernels are estimated based on the box air mass factor for each observation layer. An overview of the station data is given by Table 6.1. The MAX-DOAS instrument in Athens was not working correctly in August and September 2020 leading to gaps in the measurements during these months.

Note that the period investigated starts in June 2020 for consistency, as two new models were recently introduced to the ENSEMBLE.

6.3 Inter-comparison method

Model VCDs (vertical column densities) have been calculated based on regional model data interpolated to MAX-DOAS output altitudes. Column averaging kernels (AVKs) from the measurements were applied to model NO₂ partial columns before summing up NO₂ values in the vertical:

$$VCD_{method2}^{model} = \sum_{i=1}^{Nobs} AVK_i \cdot VCD_i^{model}$$

The averaging kernels are part of the profiling output and represent the sensitivity of the retrieved column to the NO₂ amount at different altitudes. As the sensitivity of MAX-DOAS retrievals is largest in the boundary layer, the application of averaging kernels from the measurements to model simulations can have a crucial influence on validation results.

Only those model values closest to the measurement time are used below. As the model output is given in hourly time steps, the maximum possible time difference between measurements and simulations shown here is 30 minutes.

6.4 Results

Figure 6.1 shows time series of tropospheric NO₂ VCDs derived from MAX-DOAS for the model ENSEMBLE. The magnitude of VCDs from the measurements for De Bilt and Bremen is reproduced by the models, an underestimation is found for Athens. The underestimation for Athens may be related to problems in simulating vertical transport of pollution within the boundary layer, as the instrument is placed on a small hillside above the city centre and the comparisons for Athens are therefore representative for altitudes larger than approximately 500 m absl. The retrievals show a larger variability of values compared to the models. Measurements and simulations don't agree very well for some of the time steps investigated. The models underestimate some of the larger NO₂ values inside individual pollution plumes. Models may fail to reproduce these peaks due to errors in NO_x emissions, transport of NO₂ towards the stations and chemistry.

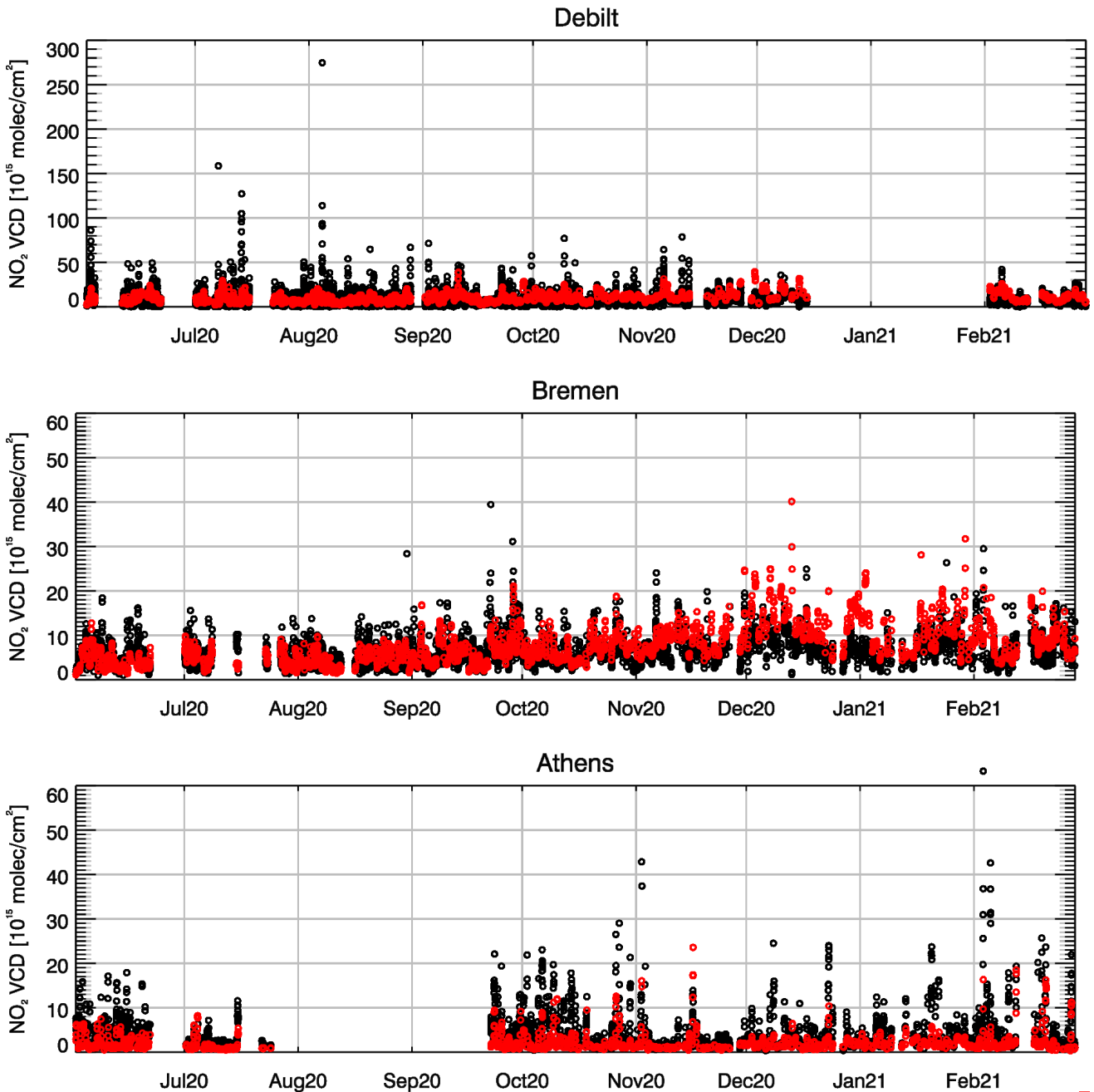


Figure 6.1. Time series of tropospheric NO₂ VCDs [10^{15} molec. cm^{-2}] from (black circles) MAX-DOAS and (red circles) the ENSEMBLE forecasts for (from top to bottom) De Bilt, Bremen, Athens.). Model results were calculated by multiplying NO₂ partial columns with averaging kernels for each observation layer followed by summing up resulting values in the vertical. Model data was interpolated to the averaging kernel layer altitudes prior to calculation of VCDs. Time period: June 2020 - February 2021. The MAX-DOAS instrument in Athens was not working correctly for a period in August and September 2020 leading to gaps in the time series.



Table 6.1. Overview of MAX-DOAS station data used for validation of regional air quality model simulations. The period covered in this report is June 2020 to February 2021.

Station	Latitude, longitude	Altitude above sea level	Institution	Quantity	Character
Bremen (Germany)	53.106°N, 8.86°E	21 m	IUP-UB	column	urban
De Bilt (Netherlands)	52.1° N, 5.18° E	23 m	KNMI	column	urban
Athens (Greece)	38° N, 23.7° E	527 m	IUP-UB	column	urban

Figure 6.2 shows comparisons of diurnal cycles. Again, the mean column amounts of the ENSEMBLE are comparable for De Bilt and Uccle, but values are generally underestimated in Athens. Although larger differences are found depending on the regional model, the model ENSEMBLE performs much better than CAMS-global for all stations, CAMS-global is negatively biased. Some regional models show different variations from one hour to another. The afternoon peak observed by MAX-DOAS for De Bilt is not reproduced by all models. This may be related to inadequate photochemistry, scaling of emissions in time or vertical distribution of NO₂ and errors in simulating pollution transport towards the station.

Comparisons of weekly cycles are shown in Figure 6.3. Weekly cycles are underestimated by all regional models, with a stronger decrease of NO₂ columns from workdays towards the weekend retrieved by MAX-DOAS for all urban stations. CAMS-global mainly fails to reproduce weekly cycles. Note that some variations of values from one day to another may just be coincidence due to data sampling. Large variability is found between the Monday averages by the individual regional models especially for Athens.

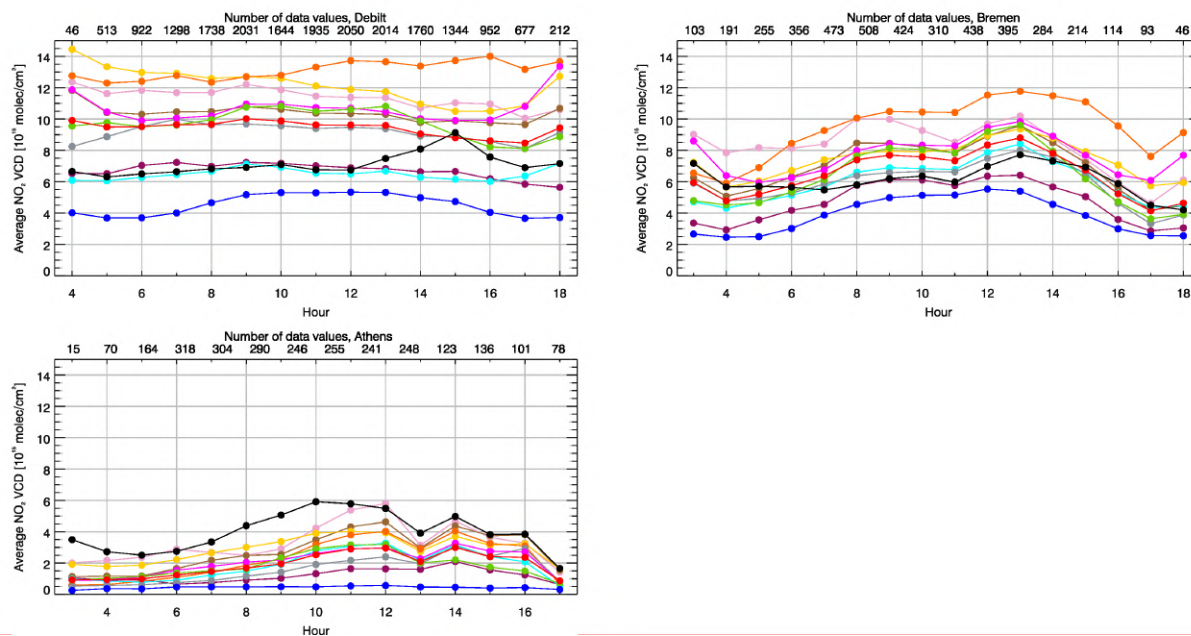


Figure 6.2. Diurnal cycles (averages over hourly bins) of tropospheric NO₂ VCDs [10¹⁵ molec. cm⁻²] for (top left) De Bilt, (top right) Bremen and (lower left) Athens. The black lines show the MAX-DOAS retrievals. All other lines refer to model data: (red) ENSEMBLE, (yellow) CHIMERE, (brown) EMEP, (orange) SILAM, (purple-blue) LOTOS-EUROS, (cyan) EURAD-IM, (pink) MOCAGE, (grey) MATCH, (fuchsia) DEHM, (light green) GEM-AQ and (blue) CAM5-global. Time period: June 2020 – February 2021.

Scatter density plots or heat maps of tropospheric NO₂ VCDs from MAX-DOAS against model ENSEMBLE values corresponding to the time series displayed in Figure 6.1 as well as statistical values (root mean squared error, bias, correlation) are given in Figure 6.4. Corresponding statistical values for all individual models are given in Table 6.2. Correlations on the order of 20-70 % are found for each station for all models. The ENSEMBLE reaches the highest correlation of about 65 % at Athens. Models tend to overestimate lower and underestimate higher NO₂ VCDs for the three urban stations. While the spread of values is quite large for individual data points, there is a fair agreement between models and retrievals for the majority of the measurements for urban stations (as shown by the high percentage of values close to the reference line). The ENSEMBLE performs significantly better than CAM5-global in terms of correlation for all stations except for De Bilt. CAM5-global has a rather strong negative bias at all stations and the correlation is nearly zero for Athens. All regional models are negatively biased in Athens.

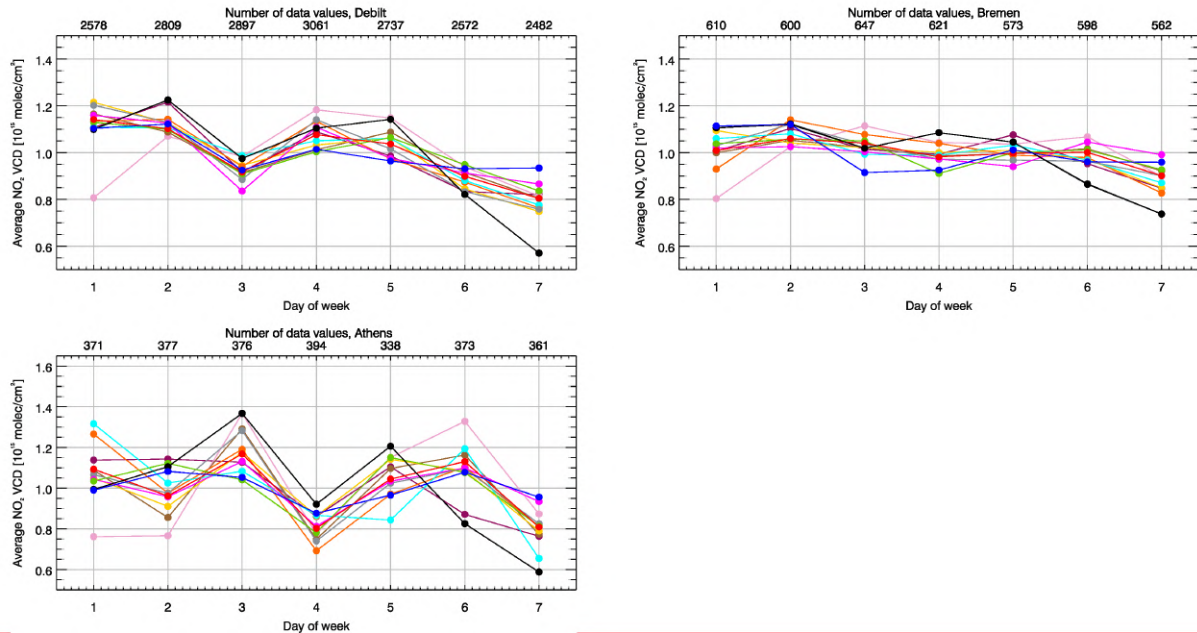


Figure 6.3. Weekly cycles (averages over daily bins divided by mean over whole week) of tropospheric NO₂ VCDs [10¹⁵ molec cm⁻²] for (top left) De Bilt, (top right) Bremen and (lower left) Athens. The black lines show the MAX-DOAS retrievals. All other lines refer to model data: (red) ENSEMBLE, (yellow) CHIMERE, (brown) EMEP, (orange) SILAM, (purple) LOTOS-EUROS, (cyan) EURAD-IM, (pink) MOCAGE, (grey) MATCH, (fuchsia) DEHM, (light green) GEM-AQ and (blue) CAMS-global. Time period: June 2020 - February 2021.

	De Bilt	Bremen	Athens
ENS	6.766/2.299/0.344	3.789/0.863/0.501	4.310/-2.296/0.621
CHIMERE	8.705/4.818/0.321	3.856/1.574/0.520	3.600/-1.211/0.665
EMEP	7.212/3.129/0.365	4.486/1.499/0.485	3.880/-1.403/0.641
SILAM	10.879/5.980/0.288	6.617/3.850/0.459	4.273/-1.896/0.557
LOTOS-EUROS	6.377/-0.313/0.255	3.897/-0.896/0.448	5.341/-3.129/0.291
EURAD-IM	8.615/4.257/0.308	6.339/2.642/0.382	5.254/-0.763/0.506
MOCAGE	5.984/-0.626/0.304	3.662/0.301/0.502	4.542/-2.332/0.530
MATCH	7.451/2.156/0.319	3.679/0.151/0.460	4.685/-2.736/0.566
DEHM	8.186/3.297/0.263	4.641/1.748/0.426	4.253/-2.091/0.614
GEMAQ	8.165/2.868/0.242	4.717/1.069/0.450	4.733/-2.249/0.393
OSUITE	6.159/-2.361/0.313	3.924/-1.851/0.343	5.943/-3.838/-0.010

Table 6.2: Statistics on how tropospheric NO₂ VCDs [10¹⁵ molec. cm⁻²] from models compare to MAX-DOAS retrievals at the three stations. Each column entry shows from left to right: root mean squared error [10¹⁵ molec. cm⁻²], bias [10¹⁵ molec. cm⁻²] and correlation coefficient (cor). Time period: June 2020 - February 2021.

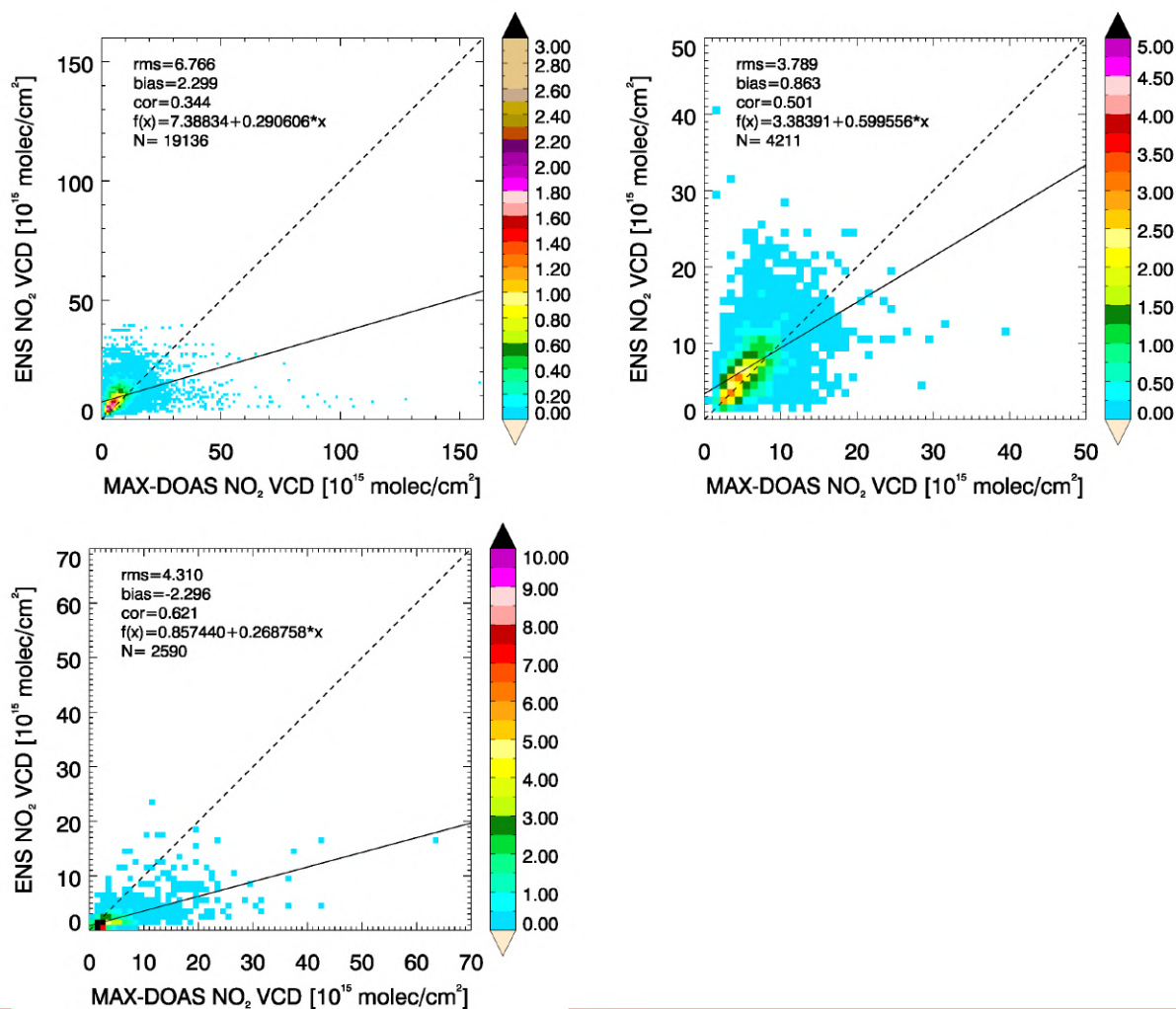


Figure 6.4. Scatter density plots of tropospheric NO₂ VCDs [10¹⁵ molec. cm⁻²] from MAX-DOAS against model ENSEMBLE hourly data for (top left) De Bilt, (top right) Bremen and (lower left) Athens. The data is shown with a bin size of 1 x 10¹⁵ molec. cm⁻² and colour according to the number of data points per bin [%]. The dashed line is the reference line (f(x)=x). The solid line is the regression line (see top left of each plot for f(x) of this line). The root mean squared error (rms) [10¹⁵ molec. cm⁻²], bias [10¹⁵ molec. cm⁻²], Pearson correlation coefficient (cor) as well as the number of data points N are given at the top left of each plot. Time period: June 2020 - February 2021.



7 Validation of tropospheric NO₂ columns against satellite retrievals

7.1 Summary

Regional air quality model columns of tropospheric NO₂, derived from the output provided on 8 levels with a top at 5km, are compared to 9:30 local time GOME-2/MetOp-A NO₂ satellite retrievals (IUP-UB v1.0 product). The overall spatial distribution of tropospheric NO₂ is reproduced by the ENSEMBLE, but values over central European emission hotspots are significantly underestimated by the majority of the models during winter, which results in a strong underestimation over these regions and of the seasonal cycle for the ENSEMBLE. There are stronger shipping signals compared to the satellite data. As a result of a major regional model upgrade in June (2019), which includes the use of an updated European emissions inventory with improved estimates for North African and Middle Eastern anthropogenic emissions, enhanced tropospheric columns of NO₂ are reproduced over these regions by all models. Differences between models and satellite observations may result from errors in anthropogenic emissions, photochemistry during winter months and from chemical processing inside ship plumes. In contrast to the analysis, the regional ENSEMBLE forecast shows a negative bias compared to the retrievals which is most pronounced for winter ($\sim 2-3 \times 10^{15}$ molec/cm²) but smaller during the rest of the year (overall $\sim 0.5 \times 10^{15}$ molec/cm²). The negative bias is even larger for CAMS-global, which agrees with the stronger underestimation of values for European emission hotspots compared to regional models, demonstrating the benefit of running models at higher horizontal resolution. A systematic negative bias is however not present in the analysis for seasons other than winter.

7.2 Comparison with GOME-2 NO₂

In this section, regional air quality model columns of tropospheric NO₂ are compared to GOME-2/MetOp-A NO₂ satellite retrievals (IUP-UB v1.0) [Richter et al., 2011]. This satellite data provides excellent coverage in space and time and very good statistics. However, only integrated tropospheric columns are available, and the satellite data is always taken at 09:30 LT for GOME-2A and at clear sky only. Therefore, model data are vertically integrated, interpolated in time and then sampled to match the satellite data. Uncertainties in NO₂ satellite retrievals are large and depend on the region and season. Winter values in mid and high latitudes are usually associated with larger error margins. As a rough estimate, systematic uncertainties in regions with significant pollution are on the order of 20% – 30%. Conclusions may differ for comparisons to other satellite NO₂ products (e.g., TEMIS GOME-2A, <http://www.temis.nl> shows lower retrieved NO₂ values for January). It should be noted here that model data is only available for altitudes up to 5000 m, meaning that (depending on tropopause height) tropospheric model columns may not be representative of the total amount of NO₂ in the troposphere. Note that since the CAMS-global upgrade of 26 June 2018, GOME-2A observations are assimilated by the global system. This is, however, a different retrieval product than what is used in the validation reported here (University of Bremen retrieval).

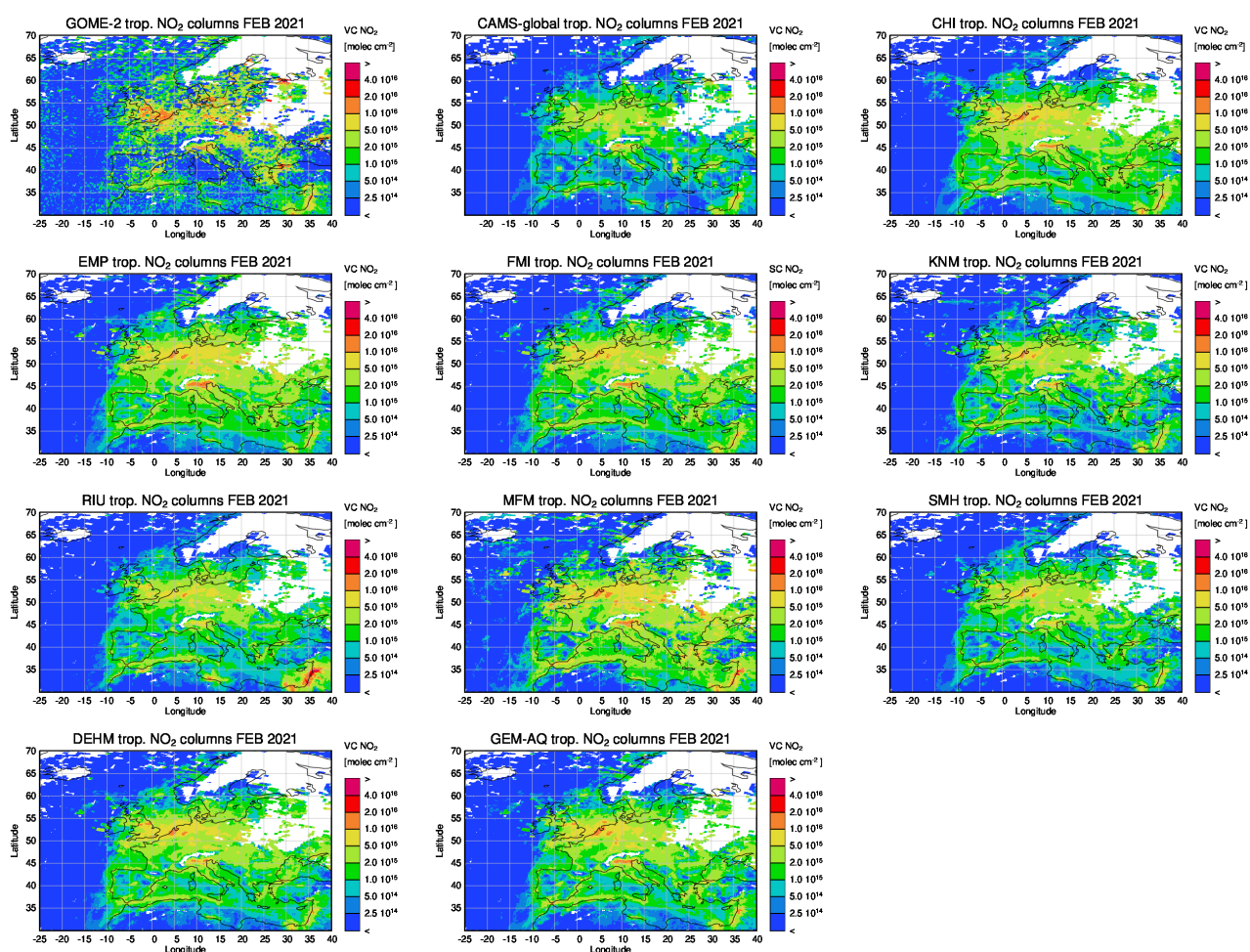


Figure 7.1. Maps of satellite-retrieved and model-simulated tropospheric NO₂ columns [molec cm⁻²] for February 2021. From left to right: (first row) GOME-2A, CAMS-global and CHIMERE; (second row) EMEP, SILAM and LOTOS-EUROS; (third row) EURAD-IM, MOCAGE and MATCH; (fourth row) DEHM and GEM-AQ. GOME-2A data were gridded to regional model resolution (i.e. 0.1° x 0.1°). Model data were treated with the same reference sector (25°W - 20°E) subtraction approach as the satellite data and linearly interpolated to the satellite overpass time (9:30 LT).

Figure 7.1 shows maps of monthly mean tropospheric NO₂ columns from GOME-2A, regional models and CAMS-global for February 2021. The overall spatial distribution and magnitude of tropospheric NO₂ is reproduced by the regional models in principle. There are generally stronger shipping signals in the models compared to the satellite data, which may result from errors in anthropogenic emissions or from chemical processing inside the ship exhaust plumes (see e.g., Vinken et al., 2014).

Compared to CAMS-global, most regional models perform better for Central European emission hotspots, showing the benefit of higher horizontal resolution runs. As a result of a major regional model upgrade in June 2019, which includes the use of an updated European emissions inventory with improved estimates for North African and Middle Eastern anthropogenic emissions, enhanced tropospheric columns of NO₂ are now reproduced over these regions (e.g., Lebanon, Israel) by all models.

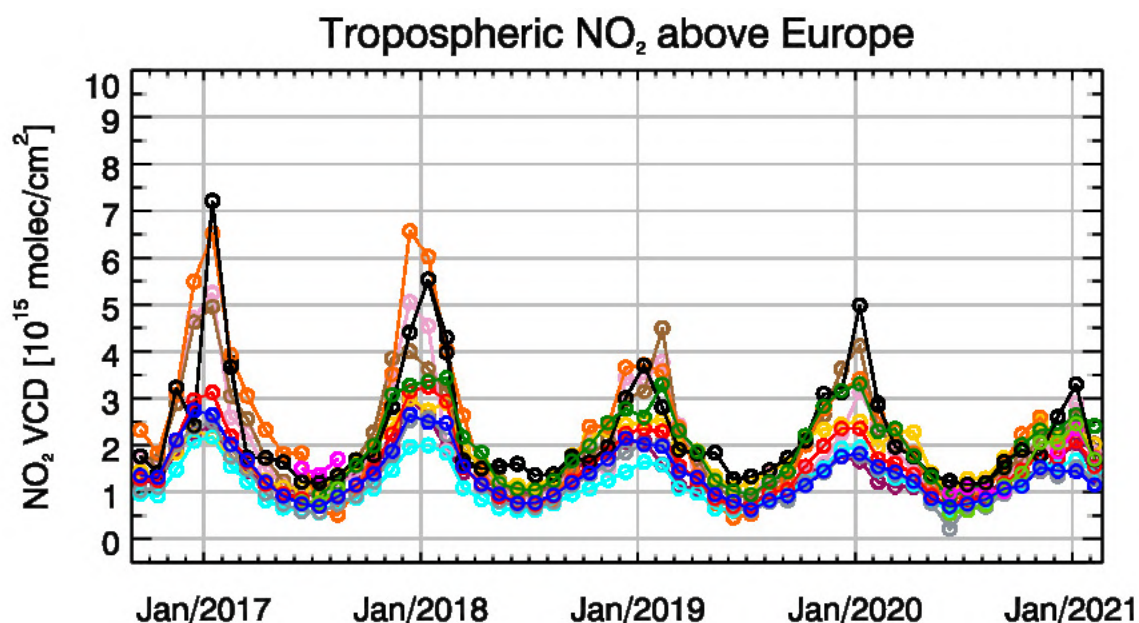


Figure 7.2. Time series of monthly averaged tropospheric NO₂ columns [10^{15} molec cm⁻²] retrieved by (black) GOME-2A and simulated by (red) ENSEMBLE forecast, (green) ENSEMBLE analyses, (yellow) CHIMERE, (brown) EMEP, (orange) SILAM, (purple) LOTOS-EUROS, (cyan) EURAD-IM, (pink) MOCAGE, (grey) MATCH, (fuchsia) DEHM, (light green) GEM-AQ and (blue) CAMS-global. GOME-2A data were gridded to regional model resolution (i.e., $0.1^\circ \times 0.1^\circ$). Model data were treated with the same reference sector ($25^\circ\text{W} - 20^\circ\text{E}$) subtraction approach as the satellite data and linearly interpolated to the satellite overpass time (9:30 LT). Time period: September 2016 – February 2021.

Figure 7.2 shows time series of monthly mean tropospheric NO₂ columns for GOME-2A and the models. The seasonal variation is better reproduced by SILAM, MOCAGE and EMEP than by the other models. The latter clearly underestimate the seasonal cycle over Europe due to the strong underestimation of values in winter described above. The regional ENSEMBLE forecast shows a negative bias compared to the retrievals which is most pronounced during winter ($\sim 2\text{-}3 \times 10^{15}$ molec/cm²) but smaller during the rest of the year (overall $\sim 0.5 \times 10^{15}$ molec/cm²). One of the reasons for this may be that the regional model output is limited to 5 km altitude. Compared to the ENSEMBLE forecast, the negative bias of CAMS-global is a bit larger, which agrees with the stronger underestimation over European emission hotspots for CAMS-global especially during winter. A systematic negative bias is not present in the regional model ENSEMBLE analysis for seasons other than winter. The decrease in the retrieved wintertime maxima from 2017 to 2019, and the increase in 2020 is not reproduced by most of regional models and CAMS-global.



8 Comparison with high-altitude EEA Air Quality e-reporting surface stations

8.1 Summary

European ozone EA Air Quality e-reporting measurements from high-altitude stations (above 1km) have been used to evaluate the regional models. Differences between the regional model orography and the true altitude of the station were used to select the model altitude level to compare with. The ENSEMBLE mostly overestimates ozone levels during the period December 2020 - February 2021. More specific, depending on the station the observed ozone levels are reproduced to within -15% and 50% by the ENSEMBLE D+0 forecast (1h-24h). Correlations observed were between 0.20 and 0.80 and the ENSEMBLE D+0 forecast has a performance better than any of the individual seven models. The EMEP, LOTOS-EUROS and SILAM models are deviating significantly from the ENSEMBLE in terms of MNMBs and LOTOS EUROS and EURAD are deviating significantly from the ENSEMBLE in terms of correlations with observations. Validation metrics are also given for the ENSEMBLE analysis. The ENSEMBLE analysis performs better than the ENSEMBLE D+0 forecast in terms of MNMBs (biases closer to zero) and correlations (significantly higher correlations).

8.2 Introduction

The seven models and their ENSEMBLE median (D+0 forecast as well as the analysis) have been compared against Background-Rural EA Air Quality e-reporting measurements for surface stations at elevation greater than 1000 m above mean sea level (<http://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-7>). Elevated stations were selected to fall within classes 1-2 in the O₃ Joly-Peuch (2012) classification for EA Air Quality e-reporting NRT stations. Table 8.1 shows the stations altitude above mean sea level together with the LOTOS-EUROS model altitude (i.e., from model's topography) pertaining to the nearest to the station grid point. Modelled gas mixing ratios were extracted at the model level, which is closest to the stations altitude as defined from the orography (see column 7 in Table 8.1).

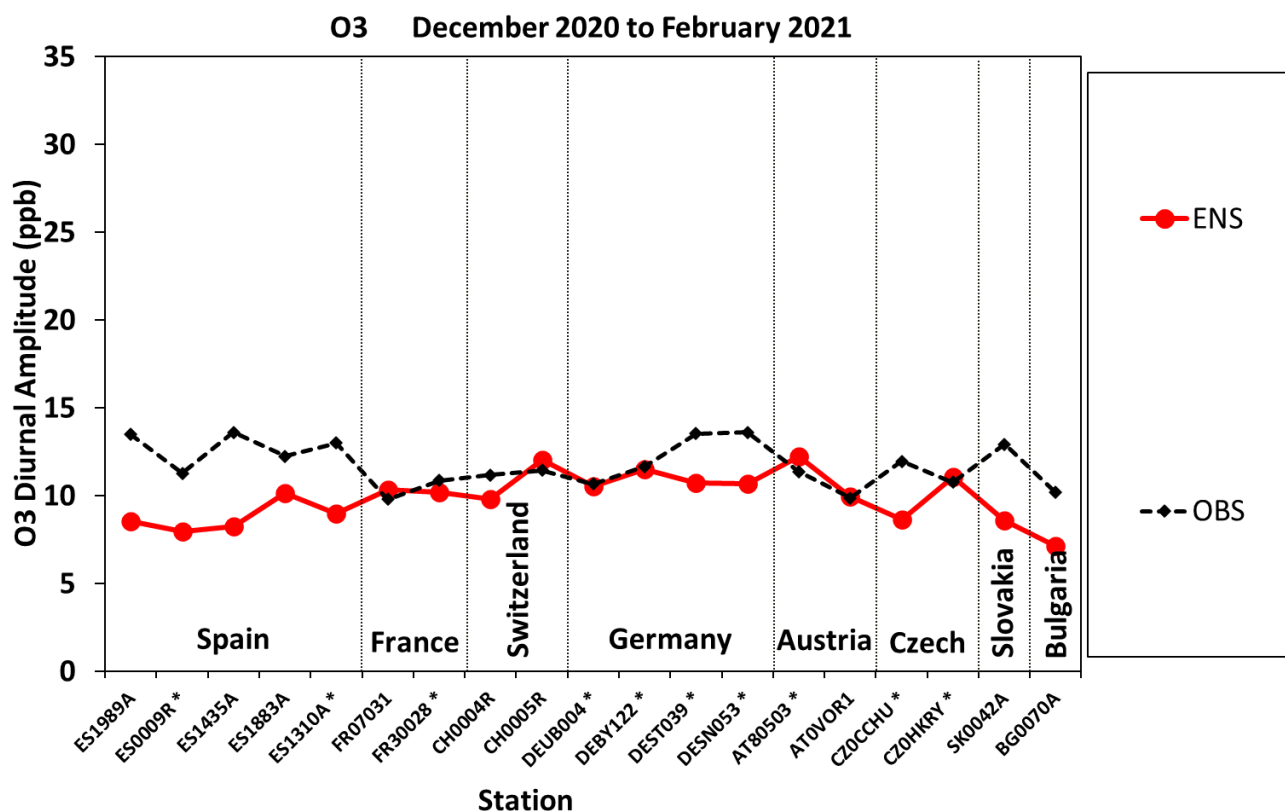


Figure 8.1. Amplitude of the diurnal cycle as measured (ppb) from stations observations (black diamonds) and as calculated from the ENSEMBLE forecast D+0 (red circles). With asterisks are denoted stations used in the assimilation process.

Table.8.1: Background-Rural EEA Air Quality e-reporting Stations (with NRT data) with elevation higher than 1000 m.

Station Name	Stat_id	Longitude	Latitude	real Altitude	model Altitude	nearest Level	use in CAMS50
Lario	ES1989A	-5.09	43.04	1140	1199	0	validation
Capmisabalos	ES0009R	-3.14	41.27	1360	1124	2	assimilation
Vilafranca	ES1435A	-0.25	40.42	1125	907	2	validation
Torrelisa	ES1883A	0.18	42.46	1005	1282	0	validation
Ak- Pardines	ES1310A	2.21	42.31	1226	1117	1	assimilation
Chaumont	CH0004R	6.98	47.05	1136	727	3	-
Rageade	FR07031	3.28	45.11	1040	944	2	validation
Schlucht	FR30028	7.01	48.05	1200	520	3	assimilation
Schauinsland	DEUB004	7.91	47.91	1205	554	3	assimilation
Rigi-Seebodentalp	CH0005R	8.46	47.07	1031	997	1	validation
Sulzberg im Bregenzerwald	AT80503	9.93	47.53	1020	961	1	assimilation
Bad Hindelang/Oberjoch	DEBY122	10.40	47.52	1169	1150	0	assimilation
Brocken	DEST039	10.62	51.80	1130	302	4	assimilation
Fichtelberg	DESN053	12.95	50.43	1214	555	3	assimilation
Vorhegg bei Kötschach-Mauthen	ATOVOR1	12.97	46.68	1020	1427	0	validation
Churanov	CZOCCHU	13.62	49.07	1118	739	4	assimilation
Krkonoše-Rychory	CZOHKRY	15.85	50.66	1001	530	4	assimilation
Bratislava - Jeseniova	SK0042A	20.99	48.78	1244	445	5	validation
Vitoshka mountain	BG0070A	23.24	42.64	1321	863	4	-

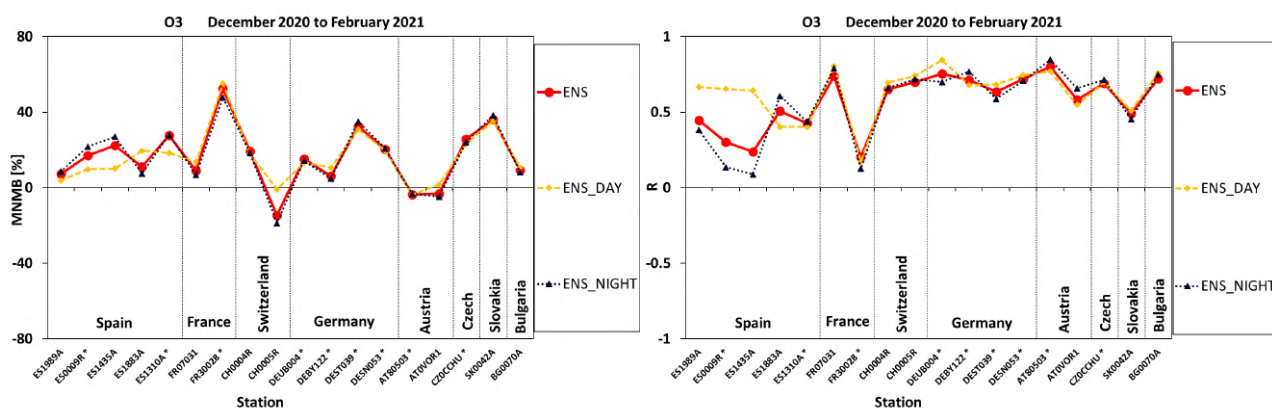


Figure 8.2. MNMBs [%] (left) and temporal correlations (right) calculated for the ENSEMBLE model for during daytime (orange) night-time (dark blue) as well as for the whole day (red) for the DJF2021 period. With asterisks are denoted stations used in the assimilation process.

For the validation, hourly O₃ concentration values (µg/m³) are extracted from the nine models as well as for the regional ENSEMBLE. It should be noted that, in the EEA Air Quality e-reporting network the O₃ measurements that were made by the instrument in ppb were converted from ppb to µg/m³, following the EU directive 2008/50, i.e., by multiplying by 2. This conversion is approximately correct for low altitude stations. However, at high altitude stations pressure and temperature should be considered when converting from ppb to µg/m³ and vice versa. As hourly pressure and temperature data were not available for all EEA Air Quality stations the comparison between observed and modelled ozone was done by re-converting both modelled and observed hourly O₃ concentration in ppb. For modelled ozone values the conversion was done by applying the following ideal gas equation with the model’s estimates of temperature (T) and pressure (P) (from CAMS- global):

$$O_3 \text{ (in ppb)} = O_3 \text{ (in mg/m}^3\text{)} * \left(\frac{R * T}{p_m * M_{O_3}} \right)$$

8.3 Regional ENSEMBLE results

In the previous report it was shown that comparing the observed and modelled amplitude of the diurnal variation of ozone at each high-altitude station could provide a criterion concerning the exposure suitability of the stations. We found out in this report that an additional criterion is needed to differentiate stations as to their suitability in exposure. The additional criterion is the correlation coefficients between the amplitude of the diurnal cycle as observed and modelled to be statistically significant roughly higher than 0.3. Figure 8.1 shows the observed and modelled diurnal amplitude of ozone at each station, moving from Spain to Bulgaria. Figure 8.2 shows the MNMBs and the correlation coefficients calculated for the ENSEMBLE model during daytime, at night-time as well as for the whole day. We can see that the 2 criteria of diurnal amplitude and day and night MNMBs and correlation coefficients differentiate the 3 stations in Spain from all other stations. For the above-mentioned findings these 3 high altitude stations will be excluded from our analysis. It should also be noted that daytime correlations have very small variations from stations to station and are higher than those calculated for night-time as well as for the 24-h daily means.

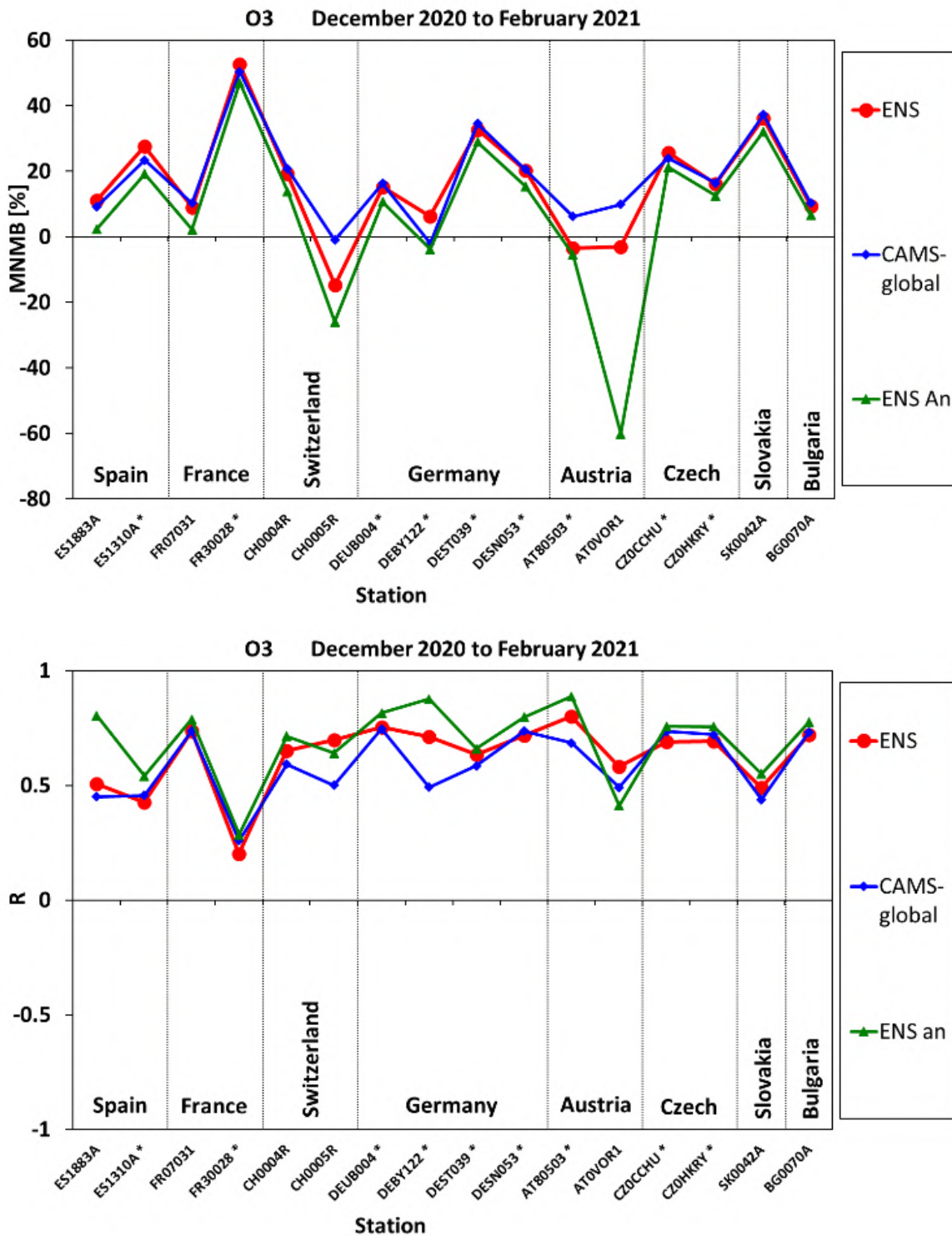


Figure 8.3. O₃ MNMBs [%] (top) and correlation coefficient (bottom) for the regional ENSEMBLE (forecast D+0; red circles and analysis; green triangles) as well as for CAMS-global (forecast D+0; blue diamonds) for the period DJF2021. With asterisks are denoted stations used in the assimilation process.

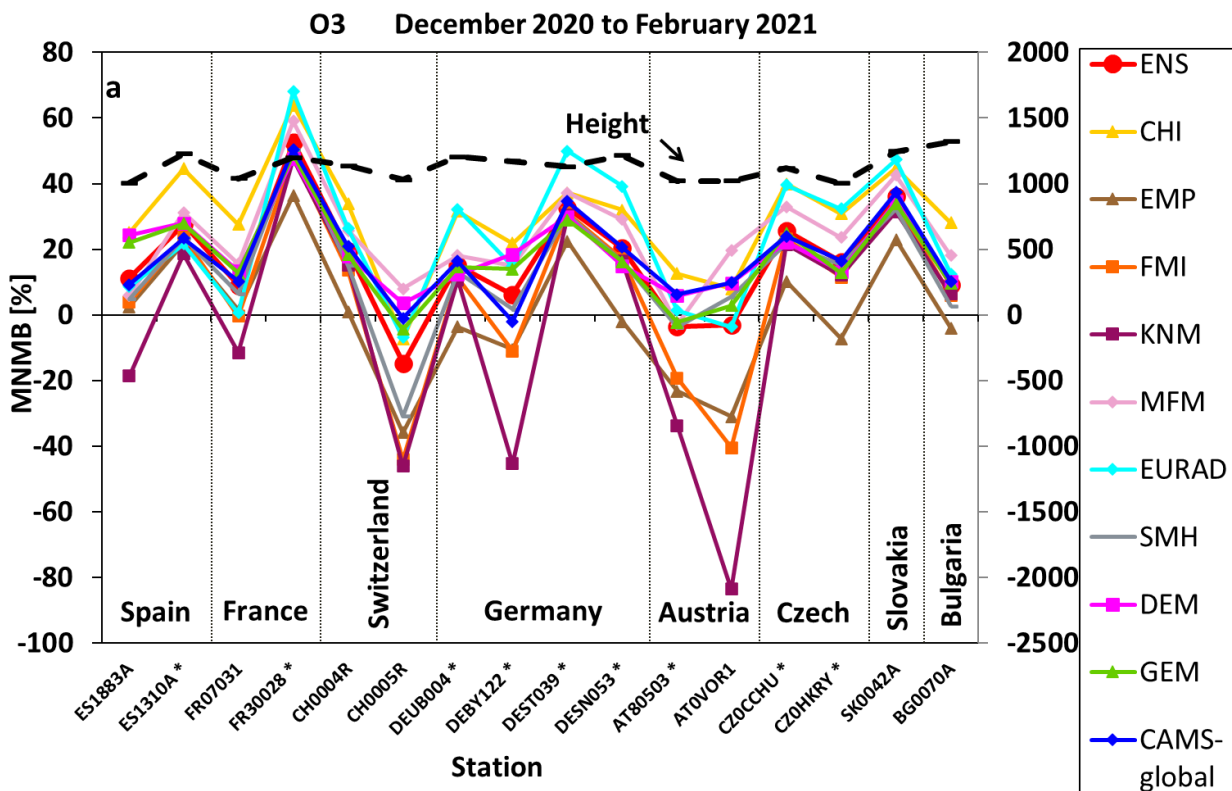


Fig. 8.4. Ozone MNMBs [%] for each of the 9 models forecasts (D+0), the ENSEMBLE forecast and CAMS-global (December 2020 to February 2021) for stations above 1000m altitude.

Figure 8.3 shows the Modified Normalized Mean biases (top) and correlation coefficients (bottom) at each of the remaining stations, moving from Spain to Bulgaria (i.e., from West to East) pertaining to the median of the ENSEMBLE forecast (D+0) and analysis (D+0) as well as CAMS-global (D+0). The ensemble median mostly overestimates ozone levels during the period December 2020 - February 2021. Depending on the station the range of MNMB for the ENSEMBLE D+0 forecast was found to be between -15% and 50%. From Figure 8.3 (bottom panel) it is obvious that the Ensemble Mean reproduces well the ozone variability. As it appears from Figs 8.3 (bottom panel) the correlation coefficients are highly significant ($0.45 < r < 0.80$; an exception is Schlucht station in France where correlation is lower $r=0.2$). It should be noted that the ENSEMBLE analysis performs better than ENSEMBLE D+0 forecast in terms of MNMBs (biases closer to zero) and correlations (significantly higher correlations) and that CAMS ENSEMBLE D+0 forecast shows higher correlations than CAMS-global D+0 forecast.

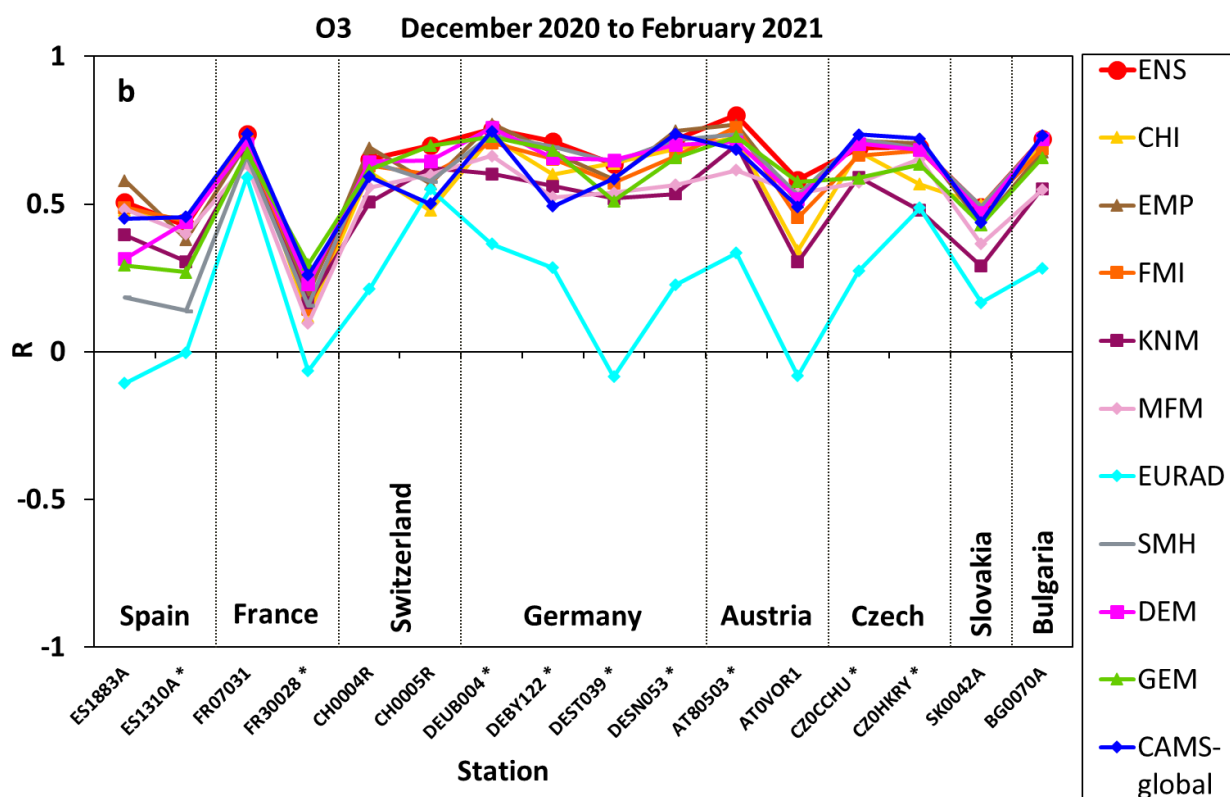


Fig. 8.5: Correlation coefficients (from hourly values) between observed and modelled ozone for the D+0 forecasts of all 9 models, the ENSEMBLE and CAMS-global (o-suite) for December 2020 to February 2021.

8.4 Results for the nine regional models

Figure 8.4 shows Modified Normalized Mean biases at each station with elevation greater than 1000 m above mean sea level, moving from Spain to Bulgaria (from West to East) pertaining to each one of the 9 model forecast calculations, the ENSEMBLE forecast as well as CAMS-global (o-suite). On top of the graph shown is the elevation of the station. Depending on the station the observed ozone levels are reproduced to within -15% and 50% from the ENSEMBLE. EMEP, LOTOS-EUROS and SILAM models deviate notably from the ENSEMBLE in several stations, while the remaining models show scores closer to the ENSEMBLE. The plot for the analysis (fig. 8.6) reveals that for all individual models (apart from the EMEP model) the analysis performs better than the forecast D+0 in terms of bias. A notable exception is the AT0VOR1 station where the ENSEMBLE analysis as well as most of the individual models (apart from the EURAD, GEMAQ and DEHM models) strongly underestimate ozone values and perform weaker than the forecast D+0 simulations.

Figure 8.5 shows the correlations between observations and each model for the forecasts. The ENSEMBLE reproduces well the ozone variability and has a better score than any of the individual models. LOTOS-EURO, and most prominently EURAD, exhibit a lower correlation while the remaining models show scores closer to the ENSEMBLE. The respective correlations for the analysis are shown in Figure 8.7. EURAD exhibits again significantly lower correlation, while the remaining models show scores closer to the ENSEMBLE.

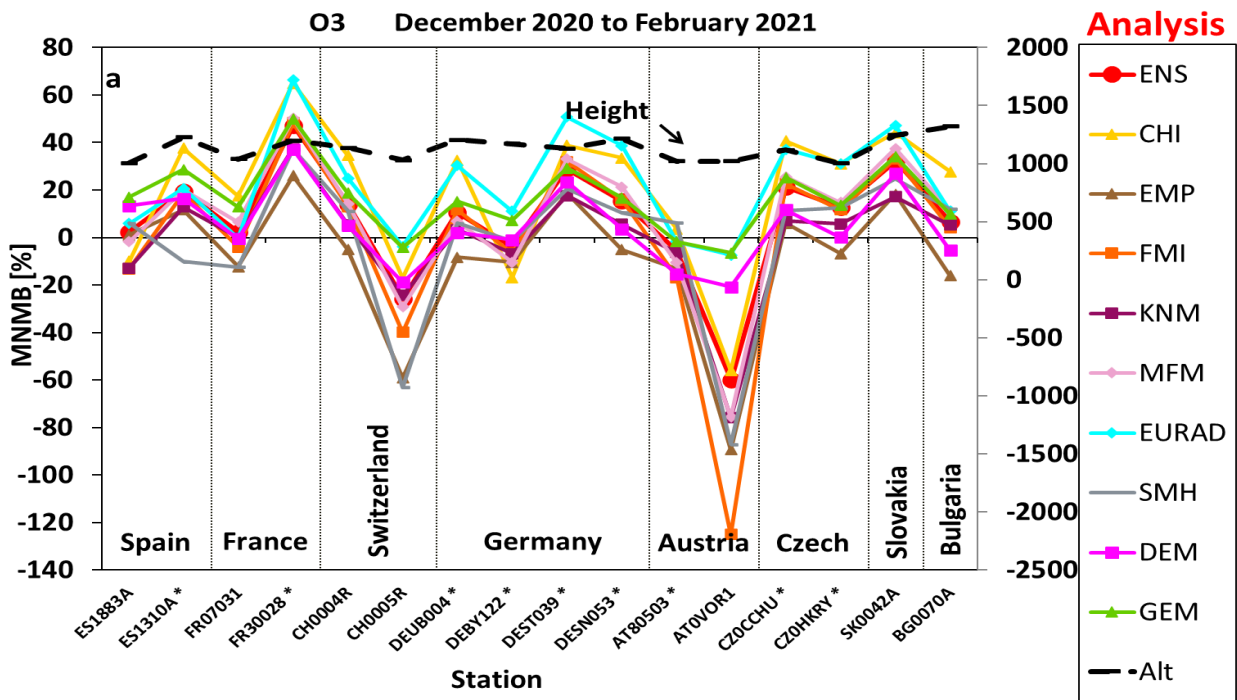


Fig. 8.6. Ozone MNMBs [%] for each of the 9 models analysis and the ENSEMBLE analysis (December 2020 to February 2021) for stations above 1000m altitud.. With asterisks are denoted stations used in the assimilation process.

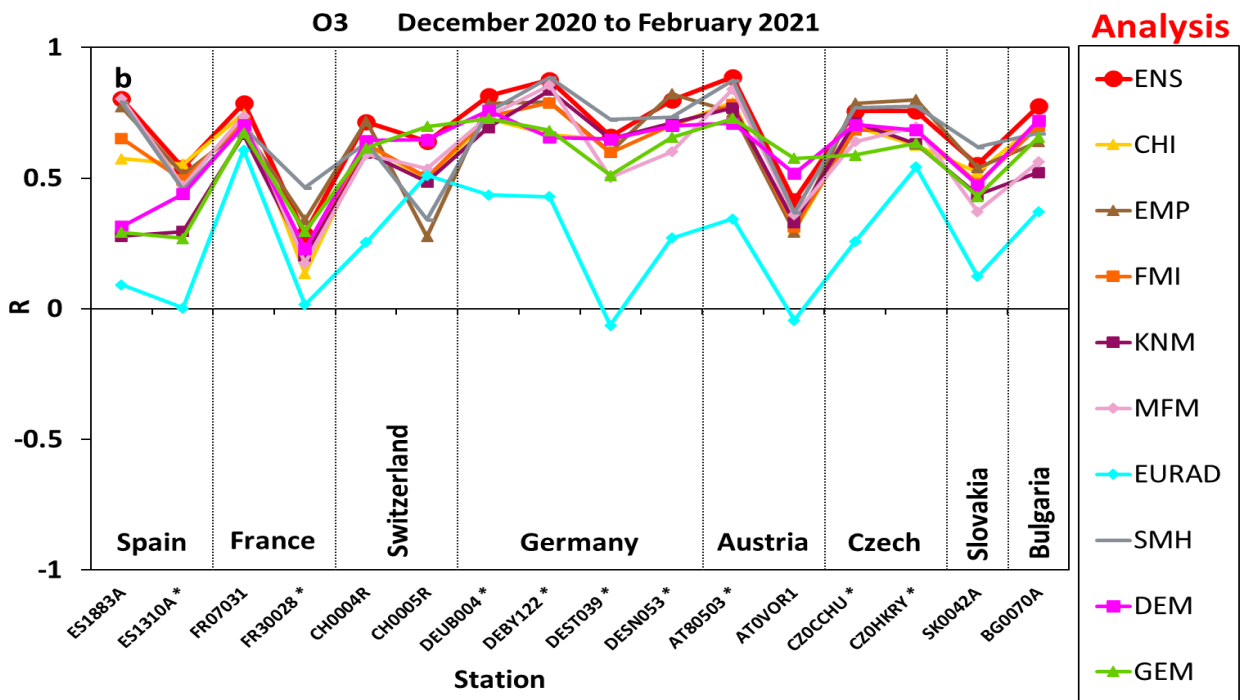


Fig. 8.7. Correlation coefficients (from hourly values) between observed and modelled ozone for the analysis of all 9 models and the ENSEMBLE for December 2020 to February 2021.. With asterisks are denoted stations used in the assimilation process.



9 Comparison with ozone sonde observations

9.1 Summary

Free tropospheric ozone (<850 hPa) could be reproduced by the ENSEMBLE forecast and analysis with MNMBs between 5% and 12% during DJF2021. The other models show MNMBs between -3% and 15% (forecasts and analysis).

9.2 Comparison approach

For the validation, the sonde profiles are compared to the model data closest in time. The model data is provided at the geographical coordinates of the sonde stations, the horizontal drift during the ascend of the sonde is considered negligible.

The model concentrations at the different height levels (0, 50, 250, 500, 1000, 2000, and 5000m above the ground) are matched to the respective sonde observations and are converted to mass mixing ratios. Pressure and temperature values needed for the conversion are taken from the sonde observations. For each station and all individual launches, the differences between observation and model are calculated. In order to be able to compare the profiles of different stations, this is done for fixed altitude levels between 0 and 6000m (interval for the surface 50m, above 100m, interval 100m). The sonde and model values are then aggregated to monthly means for each station and altitude level. For each month mean modified normalized biases (MNMB) are then calculated over all European stations for the free troposphere (<850 hPa).

9.3 Results for the ENSEMBLE

For the period February 2020 to February 2021, the ENSEMBLE forecast shows MNMBs between 3% and 15%. The ENSEMBLE analysis shows a similar behaviour with MNMBs between 3% and 14%, see Fig. 9.1.



Table 9.1: Sonde stations used in the validation for DJF2021

Station/location	Lat	lon	alt [m]
De Bilt	52.1	5.2	4
Hohenpeissenberg	47.8	11.2	976
Jokioinen	60.8	23.5	103
Legionow	52.4	20.97	96
Lerwik	60.14	-1.19	84
Madrid Spain	40.5	-3.8	631
Prag	50	14.4	302
Sodankyla	67	27	180
Uccle Belgium	51	4	100

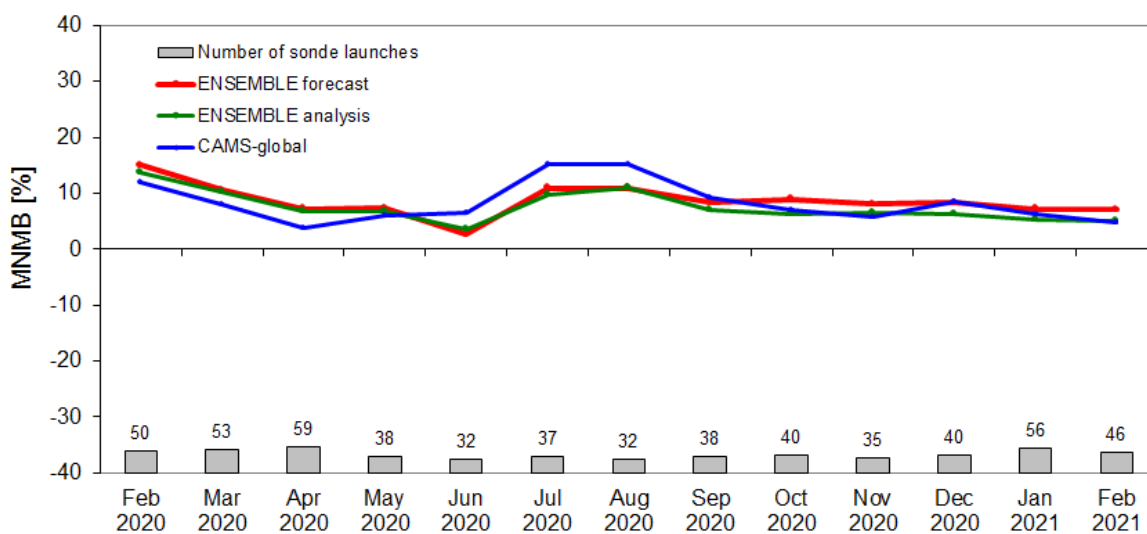


Figure 9.1 - MNMBs for the regional ENSEMBLE and CAMS-global between February 2020 and February 2021 for the free troposphere region (<850 hPa).

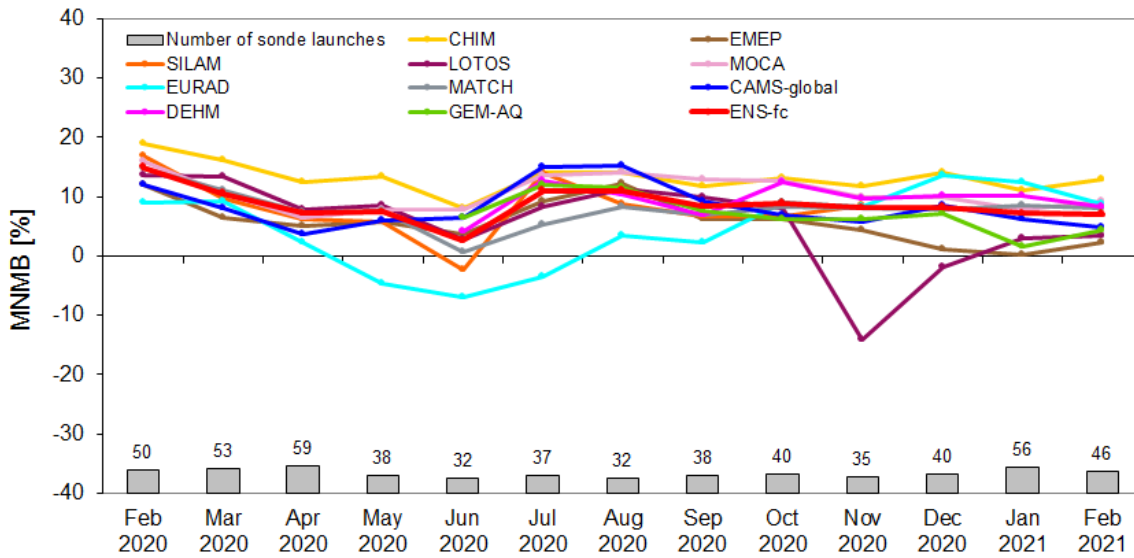
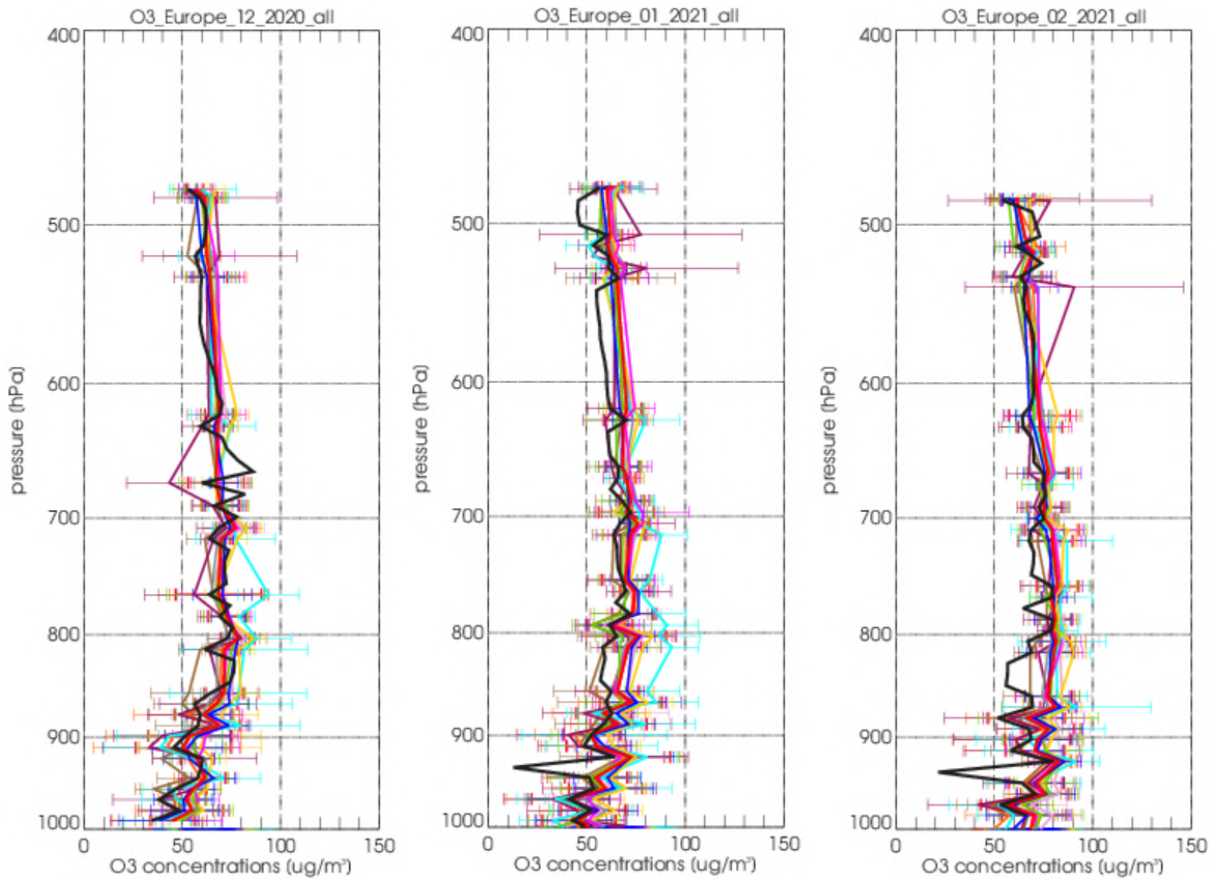


Figure 9.2. MNMBs for the individual regional model forecasts between February 2020 and February 2021 for the free troposphere region (pressure < 850 hPa).



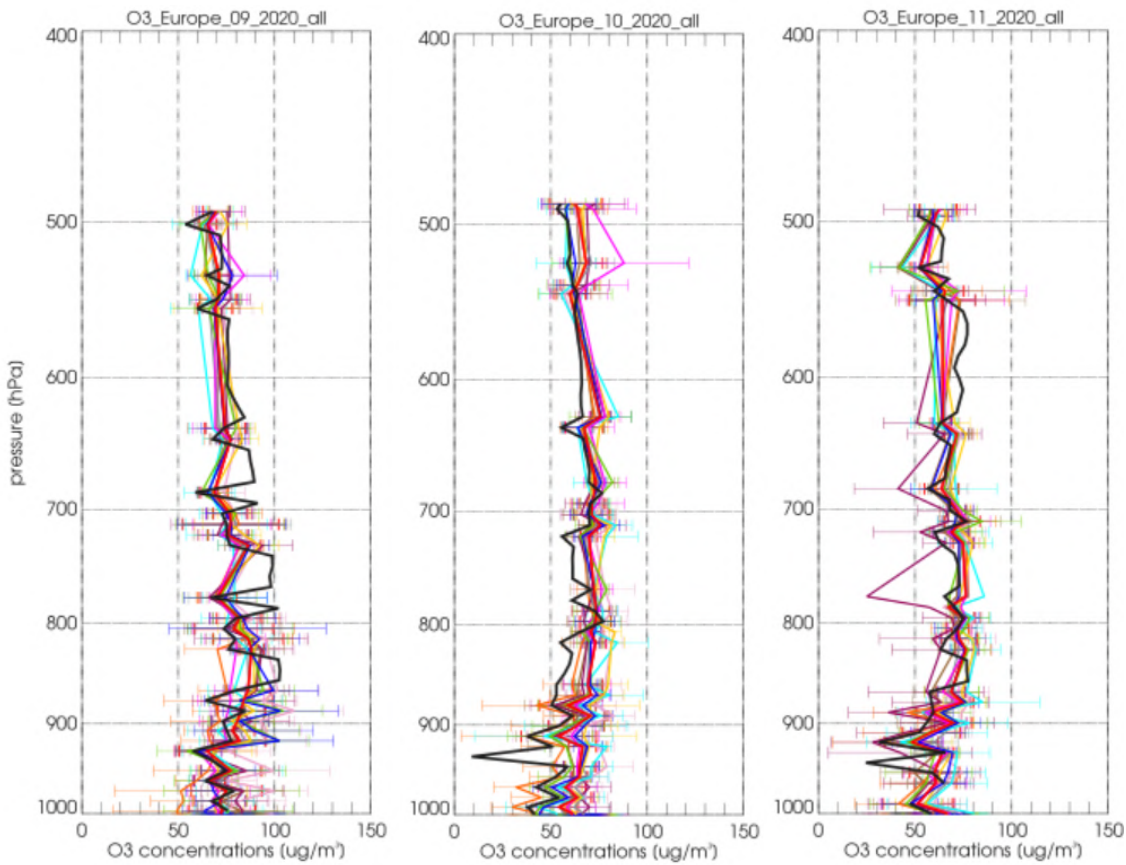


Figure 9.3. Averaged monthly profiles from the model forecast and sonde comparisons over European sonde stations for December 2020, January and February 2021 (red: ENSEMBLE, blue: CAMS-global, yellow: CHIMERE, brown: EMEP, orange: SILAM, purple: LOTOS-EUROS, cyan: EURAD-IM, pink: MOCAGE, grey: MATCH, fuchsia: DEHM, light green: GEM-AQ).

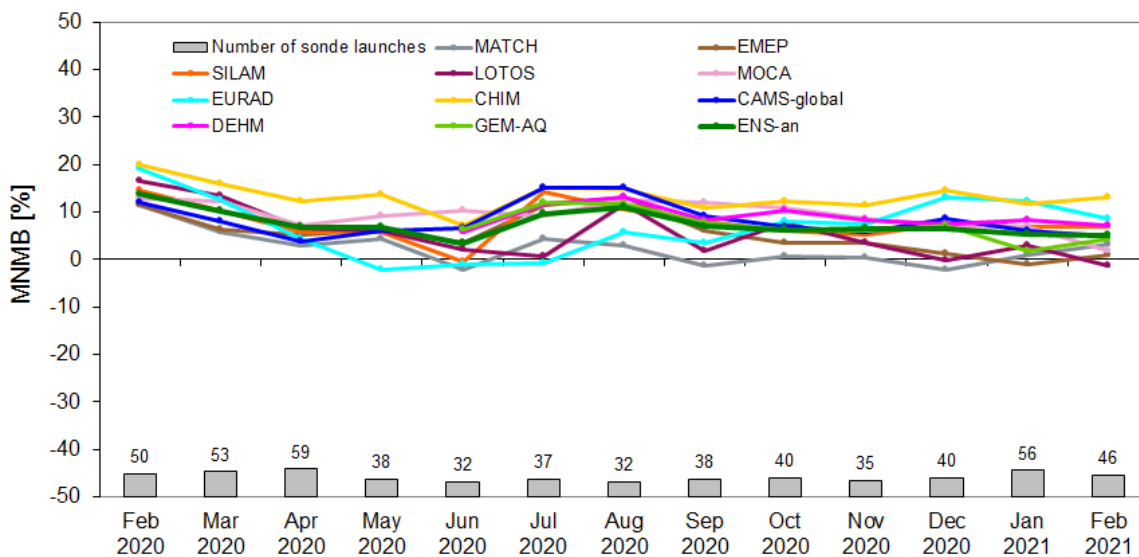


Figure 9.4. MNMBs for the regional model analyses between February 2020 and February 2021 for the free troposphere region (pressure < 850 hPa).



9.4 Results for individual regional models

Between February 2020 and February 2021 regional model forecasts show MNMBs in the range of -8 % and 20%, except for LOTOS-EUROS in November 2020 showing negative bias up to -15, see Fig. 9.2.

Results for the regional model analyses

The analyses show MNMBs between -3% and 20% (Fig. 9.4).

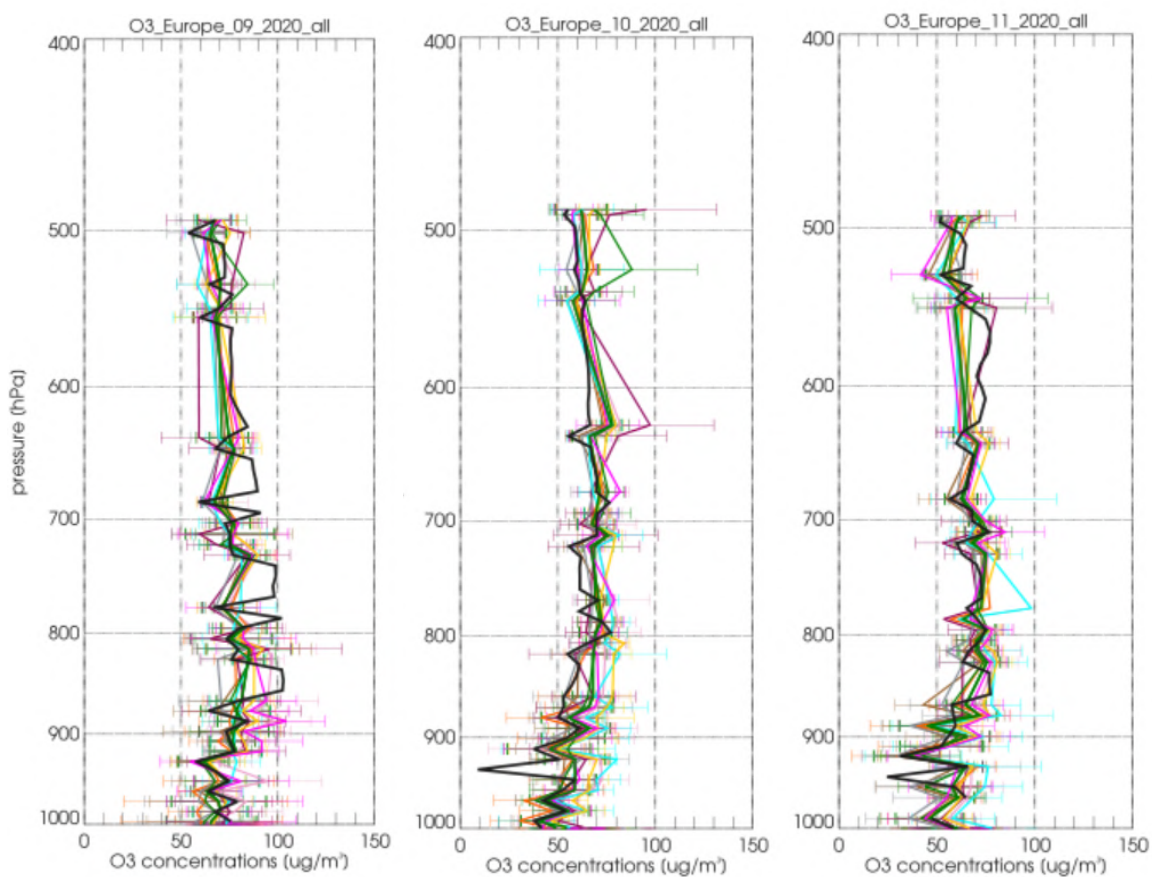


Figure 9.5. Averaged monthly profiles from the model analysis and sonde comparisons over European sonde stations for December 2020, January and February 2021 (green: ENSEMBLE, blue: CAMS-global, yellow: CHIMERE, brown: EMEP, orange: SILAM, purple: LOTOS-EUROS, cyan: EURAD-IM, pink: MOCAGE, grey: MATCH, fuchsia: DEHM, light green: GEM-AQ).



10 Comparison with GAW stations

10.1 Summary

The model concentrations of O₃ and CO at higher model levels were compared with five GAW high-altitude stations in mountainous terrain. As for the EEA air quality e-reporting stations, differences between the regional model orography and the true altitude of the station were used for this model level selection. The ENSEMBLE analysis shows slightly better performance reflecting in the smaller bias and higher correlation. The CHIMERE model shows slightly larger MNMBs in both the analysis and the forecast. The comparison with CO shows good agreement with the observations, especially the forecast data. The CHIMERE model shows largest positive bias.

10.2 Comparison method

Hourly O₃ and CO concentration values in µg/m³ are extracted from the seven models and are compared to the GAW measurements, which were converted from volume mixing ratios (ppb) into concentrations by using pressure and temperature values at the respective pressure levels from the CAMS-global model.

The altitude of the stations Hohenpeissenberg (HPB), Jungfrauoch (JFJ), Monte Cimone (CMN), Sonnblick (SNB) and Zugspitze (ZUG) in the model has been extracted from the orography as used in the LOTOS-EUROS model, see Table 10.1. For the level choice, the GAW stations' altitudes together with the best correlation of the corresponding levels were taken into account. Uncertainties due to the choice of level (calculated as mean differences between the chosen level and one up/down for the period DJF2021 for the ENSEMBLE) are up to ±11 µg/m³ for CO and up to ±9 µg/m³ for O₃.

Table 10.1 - Validation set-up for December 2020 – February 2021.

station	altitude station [m]	altitude model [m]	level choice (range 0-7)	altitude at level [m]
HPB	985	813	2	1063
JFJ	3580	1837	5	3837
CMN	2165	602	4	1602
SNB	3105	1687	3	2187
ZUG	2670	1348	4	2348

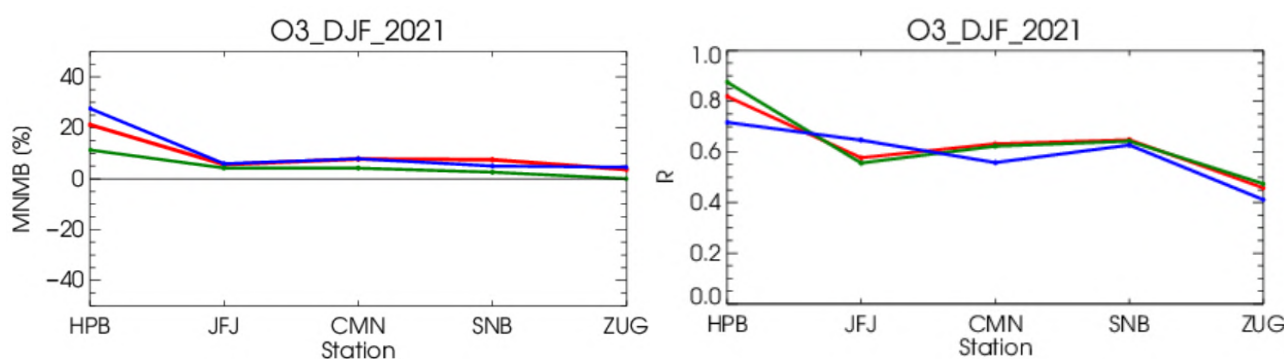


Figure 10.1. MNMBs [%] (left) and correlation coefficients (right) for ozone (red: ENSEMBLE forecast, green: ENSEMBLE analysis, blue: CAMS-global) for the period December 2020 to February 2021.

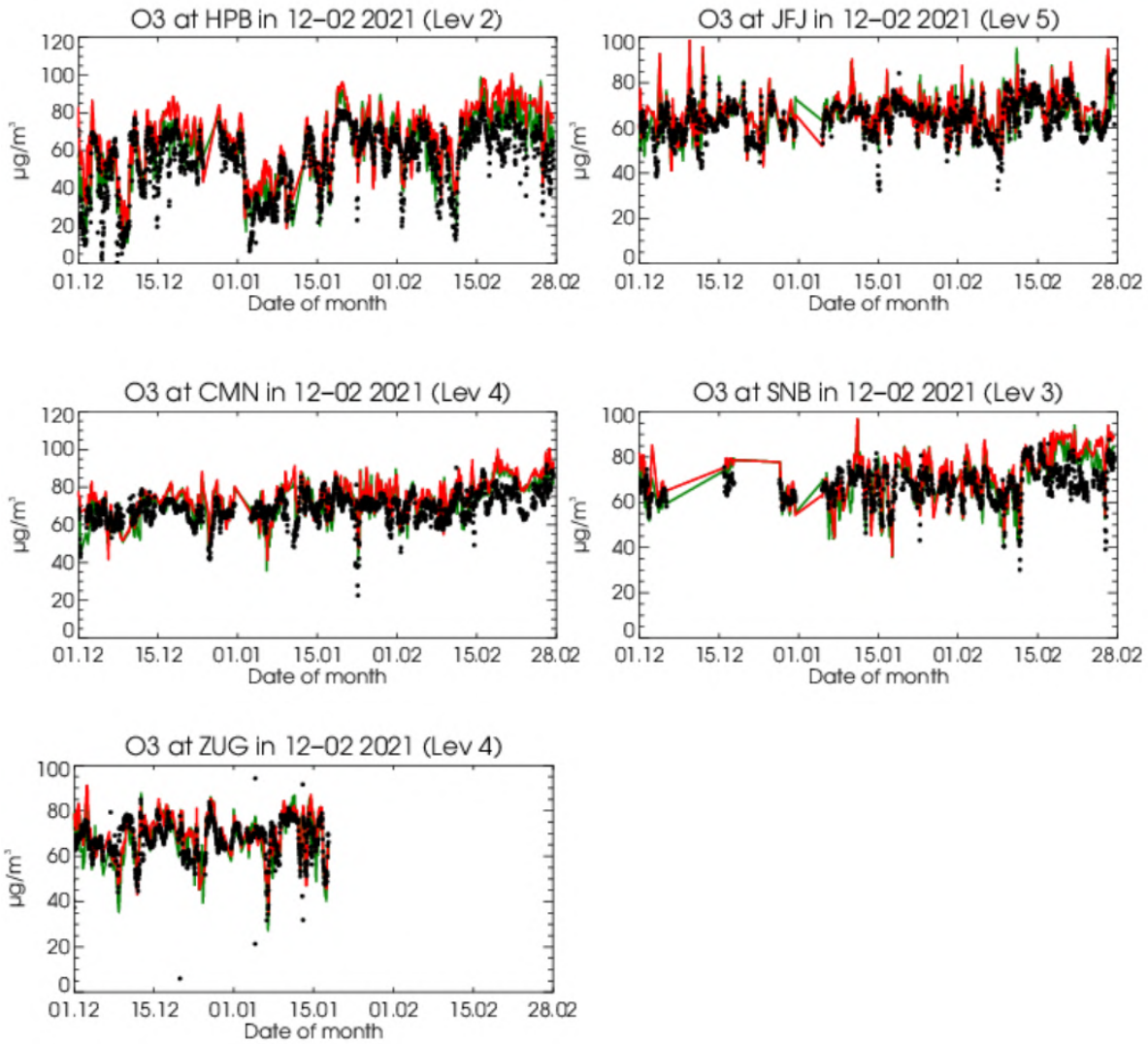


Figure 10.2. Time series plots for the ENSEMBLE forecast (red) and ENSEMBLE analysis (green) for surface O₃ in comparison with high altitude stations for the period December 2020 to February 2021.

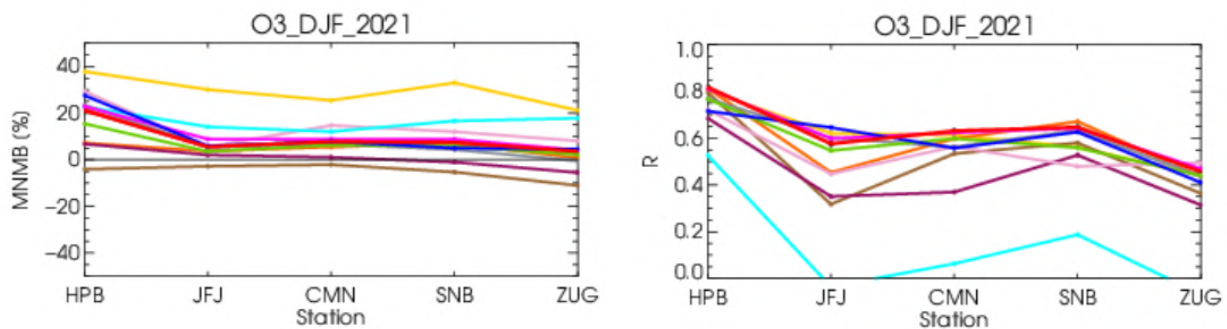


Figure 10.3. MNMBs [%] (left) and correlation coefficients (right) for all model forecasts for ozone (red: ENSEMBLE, blue: CAMS-global, yellow: CHIMERE, brown: EMEP, orange: SILAM, purple: LOTOS-EUROS, cyan: EURAD-IM, pink: MOCAGE, grey: MATCH). Altitudes are listed in Table 10.1.



10.3 Ozone

The ENSEMBLE forecast shows positive MNMBs between 5% and 20% and correlation coefficients ranging between 0.55 and 0.83 for the period December 2020 to February 2021. The ENSEMBLE analysis shows smaller MNMBs (between ~0% and 11%) and slightly better correlation coefficients ranging from 0.54 and 0.88 (see Fig. 10.1).

The time series plots show relatively good correspondence between model and observations (Fig. 10.2).

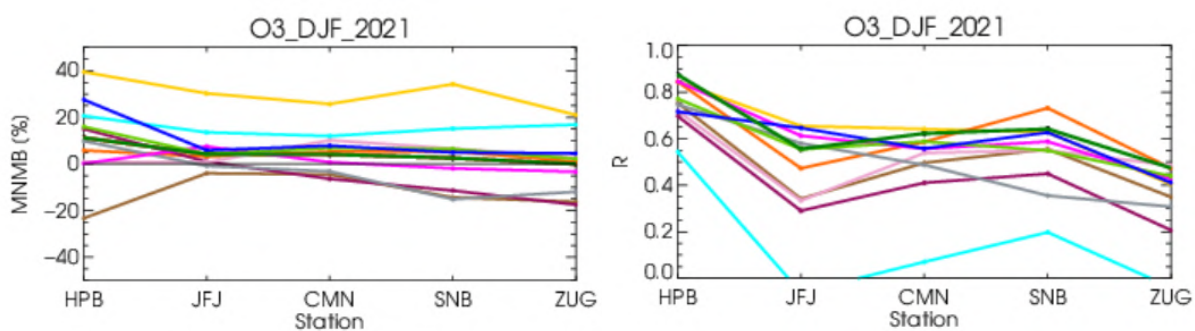


Figure 10.4. MNMBs [%] (left) and correlation coefficients (right) for all models analyses for ozone (green: ENSEMBLE, blue: CAMS-global, yellow: CHIMERE, brown: EMEP, orange: SILAM, purple: LOTOS-EUROS, cyan: EURAD-IM, pink: MOCAGE, grey: MATCH, fuchsia: DEHM, light green: GEM-AQ). Altitudes are listed in Table 10.1.

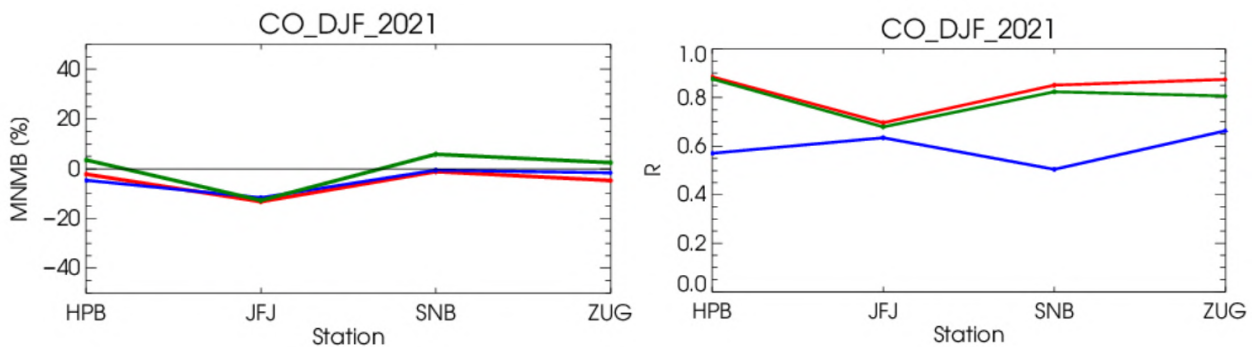


Figure 10.5. MNMBs [%] (left) and correlation coefficients (right) for the ENSEMBLE for CO (red: ENSEMBLE forecast, green: ENSEMBLE analysis, blue: CAMS-global).

Results for individual model forecasts:

The models show MNMBs in the range of 38% and -10% for the period December 2020 to January 2021 (Fig. 10.4). The CHIMERE model shows larger positive MNMBs than the other models. During DJF 2021, correlation coefficients vary between 0.3 and 0.82. An exception is the EURAD model with low correlation coefficients.

Results for the individual model analyses:



For the individual model analyses (Fig. 10.5), MNMBs range between 40% and -24%. The correlation coefficients range between 0.2 and 0.88, except for the EURAD model, which shows lower correlation.

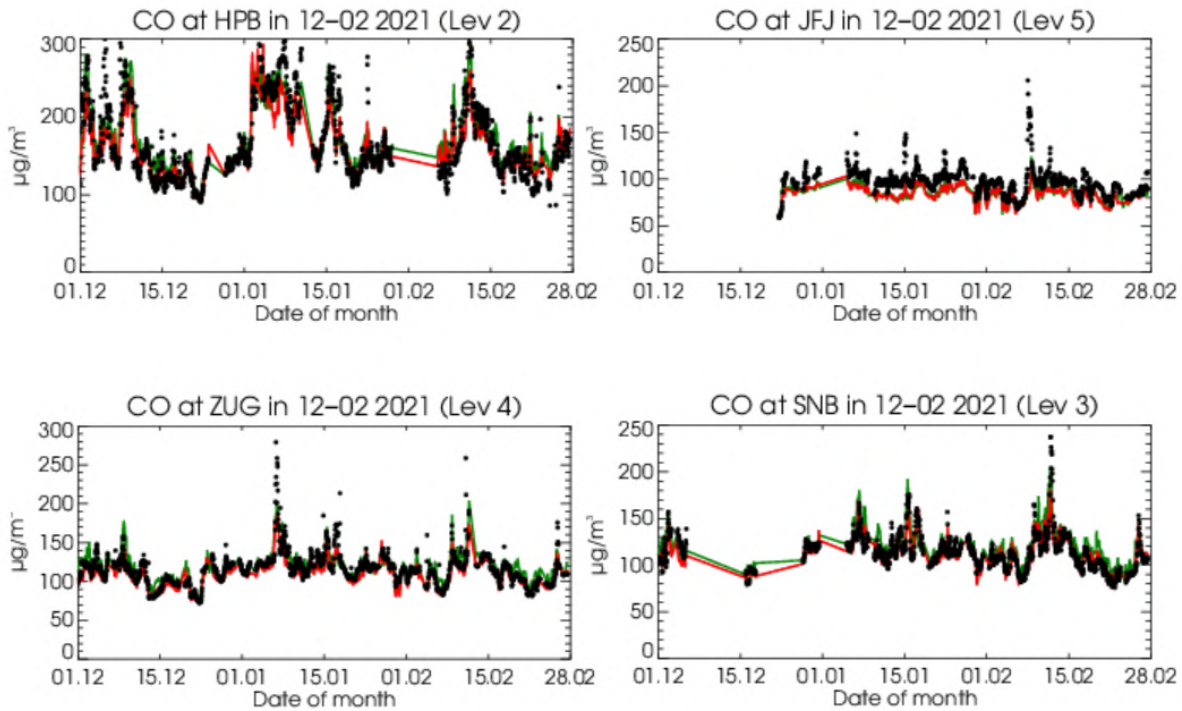


Figure 10.6. Time series plots for the ENSEMBLE forecast (red) and ENSEMBLE analysis (green) for surface CO in comparison with high altitude stations.

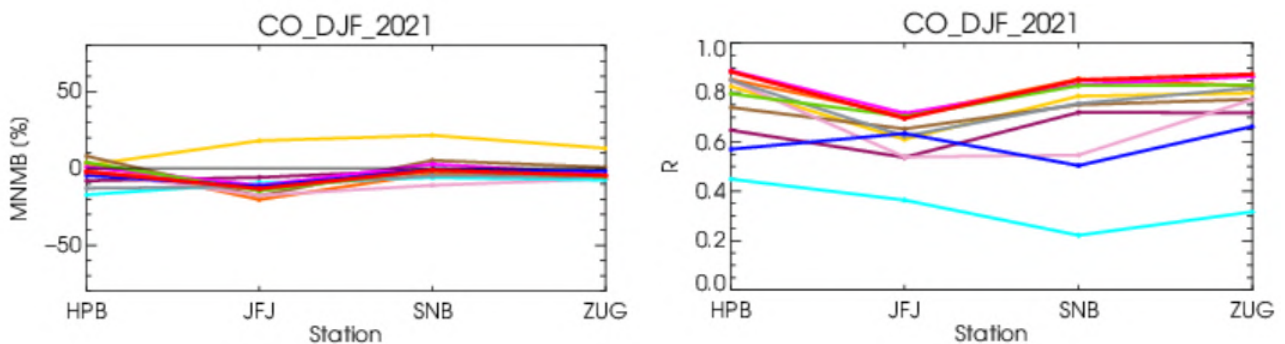


Figure 10.7. CO MNMBs [%] (left) and correlation coefficients (right) for all regional model forecasts, the ENSEMBLE and CAMS-global (red: ENSEMBLE forecast, blue: CAMS-global, yellow: CHIMERE, brown: EMEP, orange: SILAM, purple: LOTOS-EUROS, cyan: EURAD-IM, pink: MOCAGE, grey: MATCH, fuchsia: DEHM, light green: GEM-AQ). Altitude ranges are listed in Table 10.1.

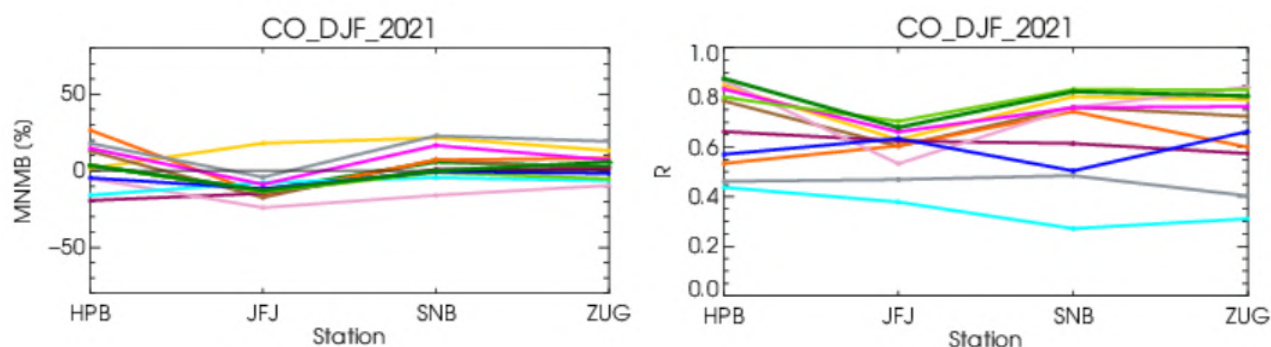


Figure 10.8. CO MNMBs [%] (left) and correlation coefficients (right) for all regional model analyses, the ENSEMBLE and CAMS-global (green: ENSEMBLE analysis, blue: CAMS-global, yellow: CHIMERE, brown: EMEP, orange: SILAM, purple: LOTOS-EUROS, cyan: EURAD-IM, pink: MOCAGE, grey: MATCH, fuchsia: DEHM, light green: GEM-AQ). Altitude ranges are listed in Table 10.1.

10.4 Carbon monoxide

For CO, the ENSEMBLE forecast and analysis MNMBs range between -14% and 8% for the analysis and between -14% and ~0% for the forecast during December 2020 - February 2021. Correlation coefficients are between 0.68 and 0.88 for the analysis and between 0.69 and 0.89 for the forecast (Fig. 10.6). The time series plots (Fig. 10.7) show mostly a very good agreement between model and observations, except for a small vertical offset for some stations.

CO results for individual model forecasts:

CO show MNMBs between -17% and 20% (Fig. 10.7). The CHIMERE model shows the largest positive MNMBs of all models. Correlation coefficients are between 0.23 and 0.8 for the forecasts. The EURAD model mostly shows the weakest performance.

Results for the individual model analyses:

CO mixing ratios range between 29% and -25% (Fig. 10.10). Correlation coefficients between the models vary significantly and range between 0.45 and 0.89.



11 Comparisons with MOPITT CO

11.1 Summary

In comparison with MOPITT v8 satellite data, the ENSEMBLE forecast data are in good agreement with the observations, showing slight negative bias within 10% (with some regional and temporal exceptions). The modelling data captured well both, relatively high CO values over the northern and middle parts of domain and relatively low values over the southern part. Analysis data are similar to the forecast data, showing slightly smaller negative bias.

11.2 Method

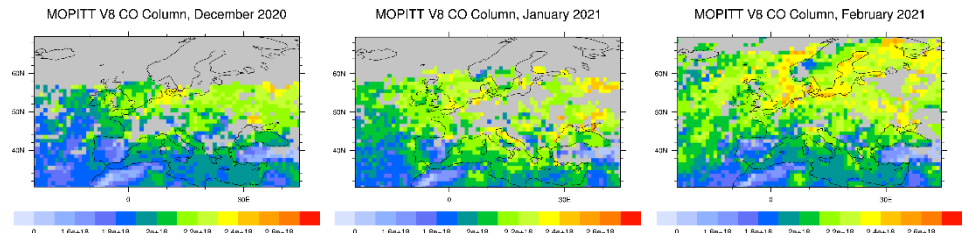
CO total column forecasts over Europe from seven regional models and the model ensemble are compared with CO total column retrievals from MOPITT Version 7 (thermal infrared radiances) (Emmons et. al., 2009). Modelled CO data were converted from $\mu\text{g}/\text{m}^3$ to VMR by using temperature obtained from CAMS-global (o-suite) model. Pressure at the middle of the layers was also interpolated from the global model. Regional model data are available from the surface up to altitude of 5 km. For the comparison with satellite retrievals, the averaging kernels were applied to the modelled data.

Regional model data up to 5 km were merged with CAMS-global data above 5 km in order to minimize uncertainty error. We performed several confidence tests to establish the method. To check the error due to coarse sampling of the profiles up to 5 km as provided by the regional models, CAMS-global data were sampled at the height levels of the regional models up to 5 km and merged with the CAMS-global original levels above 5 km. Comparison of this results with the original CAMS-global data showed that the errors due to coarse sampling of the profiles up to 5 km were very small. Both results showed slight underestimation of the MOPITT data. CAMS-global values up to 5 km sampled at the height levels as the regional models and ENSEMBLE data without merging with the levels above 5 km show overestimation of the satellite data over almost entire region. From this we concluded that error due to missing values above 5 km is significant and merging the regional data with CAMS-global values above 5 km is necessary for the proper comparison.

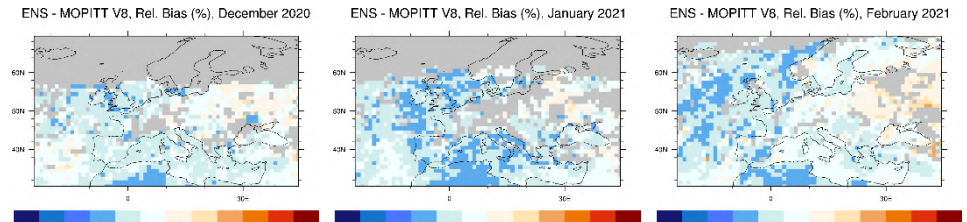
MOPITT shows relatively high CO values over the northern and middle parts of domain and relatively low values over the southern part. The ENSEMBLE forecast data show slight underestimation in general within 10% over entire domain with some regional exceptions. In February, the positive bias within 10% can be seen over the eastern part of domain. Almost all the models show very similar patterns and values. CHIMERE has a slight positive bias over the eastern part of domain (within 10%) and smaller negative bias during entire period. LOTOS-EUROS and SILAM have bigger negative biases in January and February 2021. Analysis data are similar to the forecast data, showing slightly smaller negative bias. MATCH and SILAM have slightly stronger positive biases during entire period. MOCAGE shows stronger negative bias during February 2021 over the western part of domain.



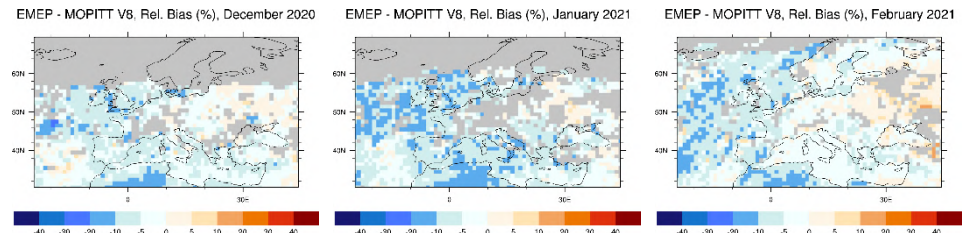
MOPITT



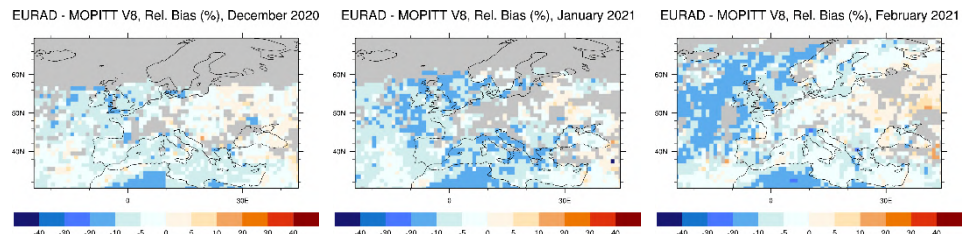
ENS-MOPITT
Forecast (0H24H)



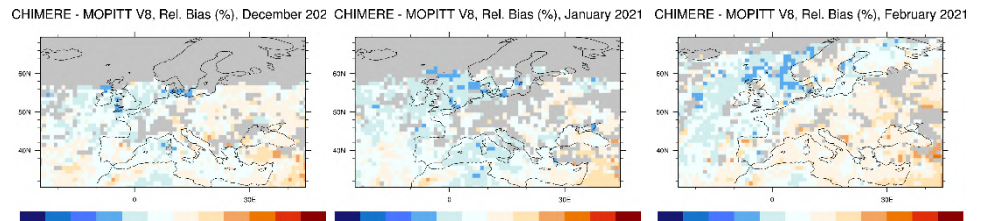
EMEP-MOPITT
Forecast (0H24H)



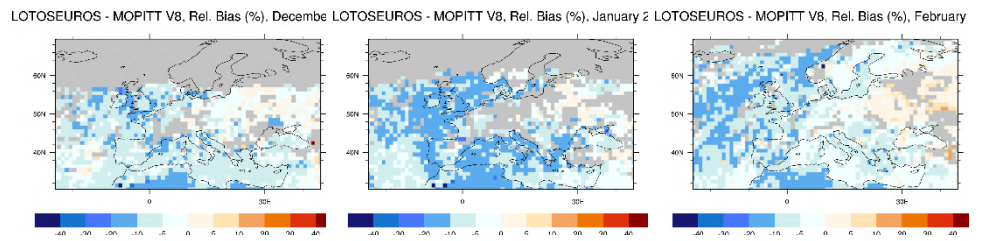
EURAD-MOPITT
Forecast (0H24H)



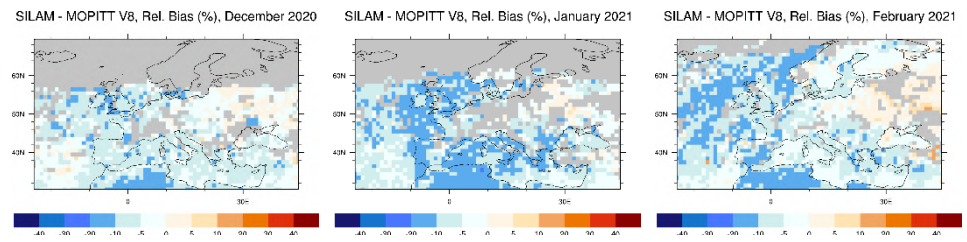
CHIMERE-MOPITT
Forecast (0H24H)



LOTOS-EUROS-MOPITT
Forecast (0H24H)

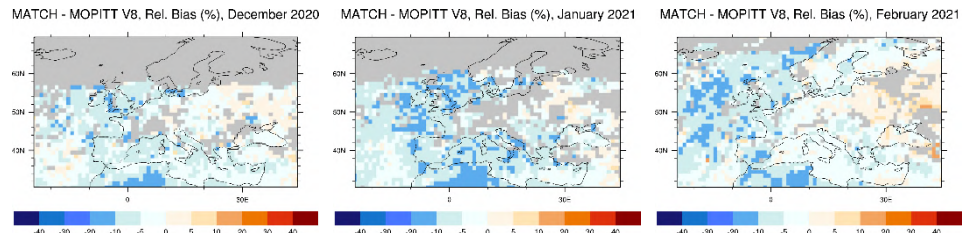


SILAM-MOPITT
Forecast (0H24H)

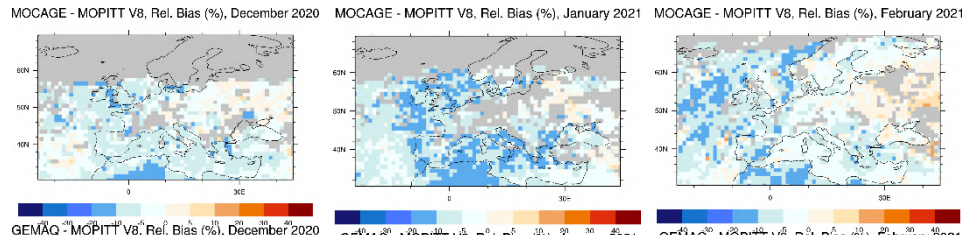




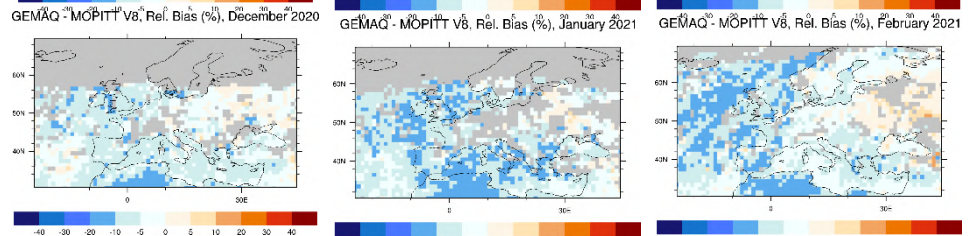
MATCH-MOPITT
Forecast (0H24H)



MOCAGE-MOPITT
Forecast (0H24H)



GEMAQ-MOPITT
Forecast (0H24H)



DEHM-MOPITT
Forecast (0H24H)

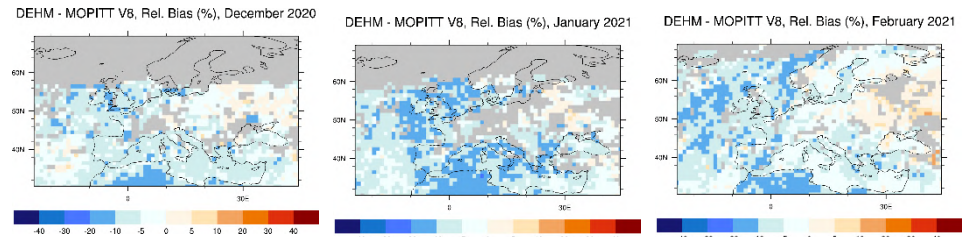
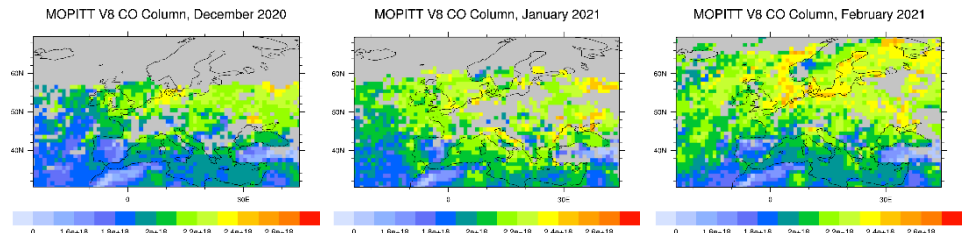


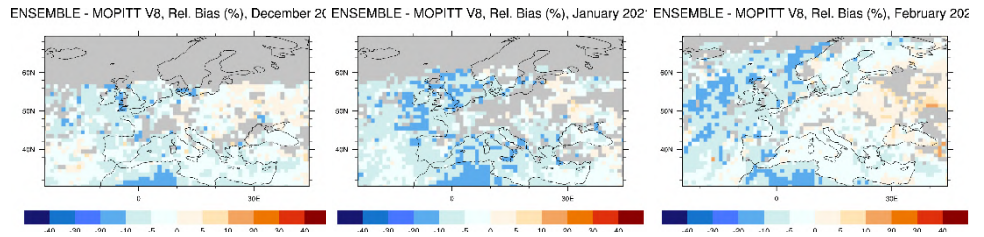
Figure 11.1. CO total column for MOPITT V8 satellite retrievals (top row, in molecules/cm²), relative difference between the regional forecasts of the nine models and the ENSEMBLE and MOPITT (other rows) for December 2020 (left column), January (middle column) and February 2021 (right column). Grey colour indicates missing values.



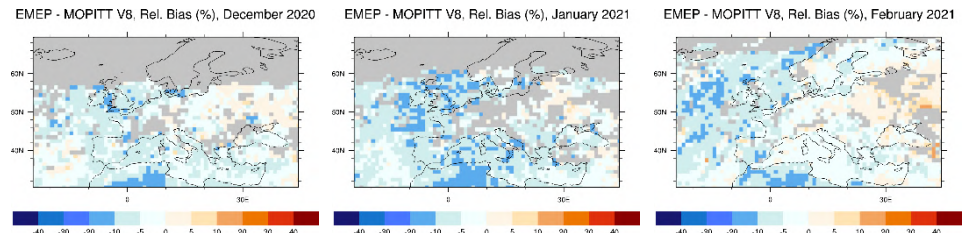
MOPITT



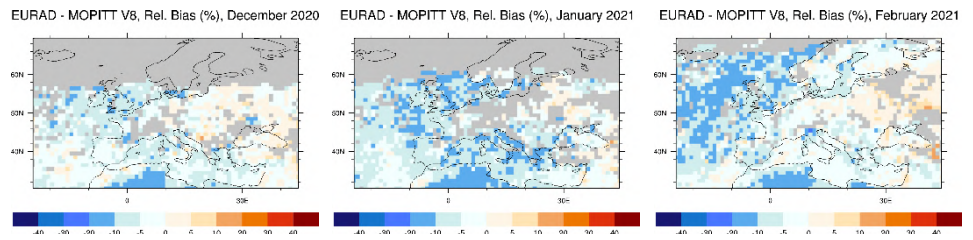
**ENS-MOPITT
Analysis (-24H-1H)**



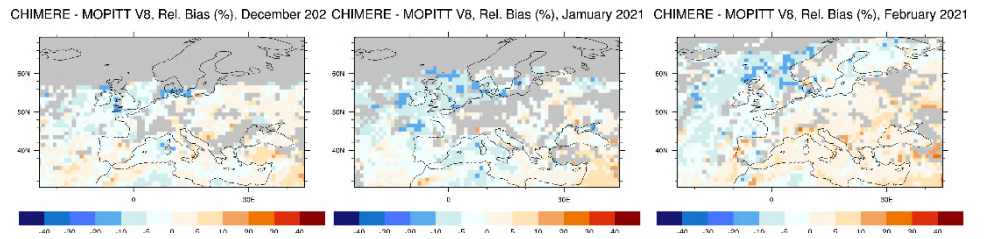
**EMEP-MOPITT
Analysis (-24H-1H)**



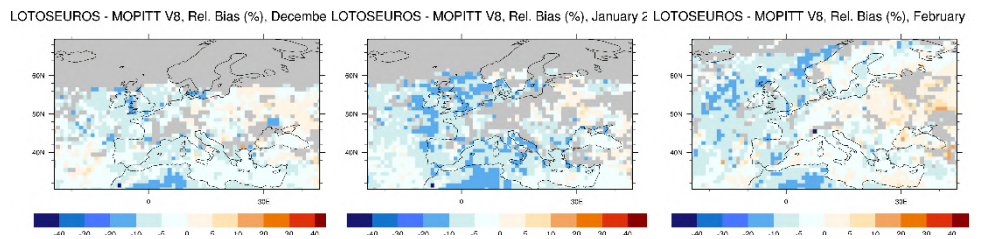
**EURAD-MOPITT
Analysis (-24H-1H)**



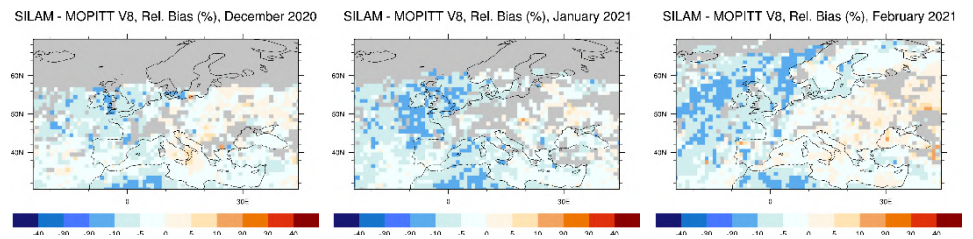
**CHIMERE-MOPITT
Analysis (-24H-1H)**



**LOTOS-EUROS-MOPITT
Analysis (-24H-1H)**

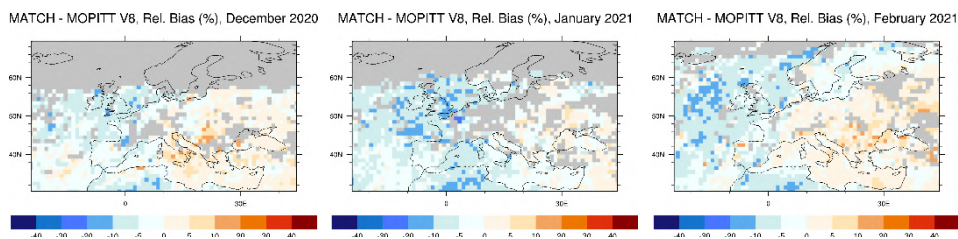


**SILAM-MOPITT
Analysis (-24H-1H)**

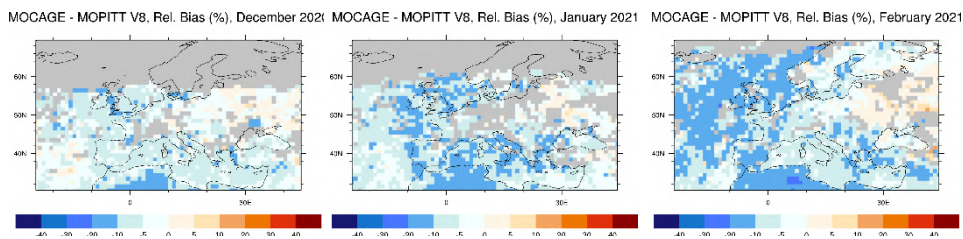




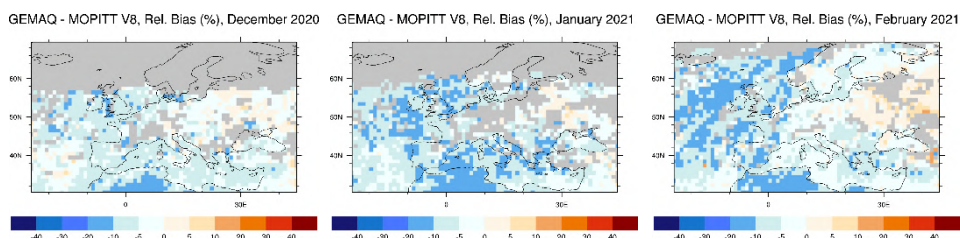
**MATCH-MOPITT
Analysis (-24H-1H)**



**MOCAGE-MOPITT
Analysis (-24H-1H)**



**GEMAQ-MOPITT
Analysis (-24H-1H)**



**DEHM-MOPITT
Analysis (-24H-1H)**

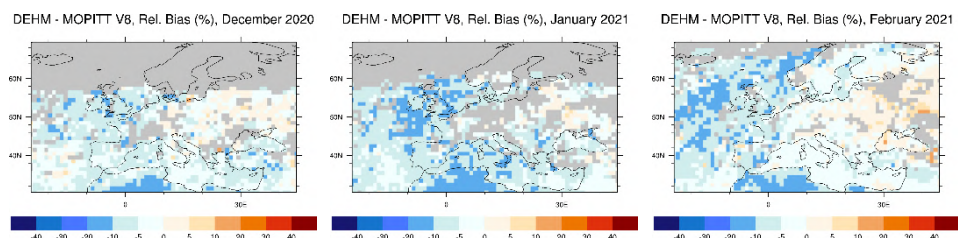


Figure 11.2. CO total column for MOPITT V8 satellite retrievals (top row, in molecules/cm²), relative difference between the regional analyses of the nine models and the ENSEMBLE and MOPITT (other rows) for December 2020 (left column), January (middle column) and February 2021 (right column). Grey colour indicates missing values.



12 Summary of issues identified in individual models

This section provides a short overview of the main, model specific issues of this report.

CHIMERE

Ozone and CO cross sections at the lateral boundaries show features that are not in line with what is seen in CAMS-global or the other models (figs 3.13 and 3.14). This indicates a possible issue with the CAMS-global boundary conditions implementation. Comparison with the GAW stations also indicates relatively high MNMBs for ozone for both the forecasts and the analyses (figs 10.3 and 10.4) and CO for the forecasts (fig 10.7).

EMEP

The model appears to be deviating from the ENSEMBLE in terms of ozone MNMB at several high-altitude stations (fig 8.4)

EURAD

Based on IAGOS observations, the model sometimes presents much higher bias than other models in the boundary layer and the free troposphere for ozone (fig. 5.5), while at the same time EURAD shows relatively large underestimations for CO. Ozone correlation coefficients at high altitude and GAW stations are much lower than for CAMS-global and the other models in both forecasts and analyses (figs 8.5, 10.3 and 10.4). Similar results are found in the correlation coefficients for CO based on comparisons at the GAW stations for the forecasts (fig 10.7).

LOTOS-EUROS

The model appears to have lower surface level values for CO compared to the rest of the models. Differences between forecasts and analyses for ozone seem to be large compared most other models and exhibit some unexpected spatial features (fig 3.18). Based on IAGOS ozone observations, the model sometimes presents higher biases than the other models, especially above 3000m (fig. 5.5). Moreover, ozone MNMBs and correlation coefficients at high altitude stations deviate considerably from the ENSEMBLE (figs 8.4 and 8.5). To some extent, these findings can be attributed to the increase, at the upgrade in November 2020, of the vertical resolution of the model for the forecasts.

MATCH

Similar to past findings, the Benelux area appears to be introducing a correction in the analysis for CO that is strongly to the opposite direction of that in the rest of the domain (fig 3.19). This may be indicative of issues in the pre-processing of the observations that are assimilated in the model

MOCAGE

High PM10 concentrations are spotted over the Atlantic, up to 250m above the surface (fig 3.9). The western boundary for PM10 is also quite high, while the southern boundary is low (fig 3.13), indicating possible issues with the implementation of the CAMS-global boundary conditions.

SILAM



Comparison with high-altitude EEA Air Quality e-reporting surface stations for ozone reveal relatively large negative biases at alpine stations (fig 8.4).



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