Investigating the impact of coupling HARMONIE-WINS50 (cy43) ~~and~~ meteorologie to LOTOS-EUROS (v2.2.002) ~~coupling~~ on simulation of NO_2 concentrations ~~in~~ over The Netherlands

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Abstract. Meteorological fields calculated by Numerical Weather Prediction (NWP) Models drive offline Chemical Transport Models (CTM) to solve the transport, chemical reactions, and atmospheric interaction over the geographical domain of interest. ~~In this way, forecasts and (re-)analyses provided by NWP can be used for air quality forecasting, climate modeling, and environmental studies. The more precise the meteorological input data represents the atmospheric dynamics, the better the CTM represents pollutant transport, mixing, and the subsequent impact on surface air quality.~~ HARMONIE (HIRLAM ALADIN Research on Mesoscale Operational NWP in Euromed) is a state-of-the-art non-hydrostatic NWP community model used at several European weather agencies to forecast weather at the local and/or regional scale. In this work, the HARMONIE WINS50 (cycle 43 cy43) reanalysis data set at a resolution of $0.025° \times 0.025°$ covering an area surrounding the North Sea for the years 2019-2021 was offline coupled to the ~~state-of-the-art model~~ LOTOS-EUROS (v2.2.002) ~~, which is a CTMthat is one of the members of the Copernicus Atmosphere Monitoring Service (CAMS), an ensemble of CTMs that is used to produce operational air quality forecasts over Europe and at a higher resolution also over the Netherlands~~CTM. The impact ~~on simulated NO_2 concentrations of using~~ of using either meteorological fields from HARMONIE ~~in~~ or from ECMWF on LOTOS-EUROS ~~compared to the use of fields from ECMWF (here used at $0.7° \times 0.7°$) is~~ simulations of NO_2 has been evaluated against ground-level ~~sensors~~ observations and TROPOMI tropospheric NO_2 vertical columns. Furthermore, the difference between crucial meteorological input parameters such as the boundary layer height and the vertical diffusion coefficient between the hydrostatic ~~(ECMWF)~~ ECMWF and non-hydrostatic ~~(HARMONIE) model fields is~~ HARMONIE data has been studied, and the vertical profiles of temperature, humidity, and wind are evaluated against meteorological ~~vertical profile~~ observations at Cabauw in The Netherlands. The results of these first evaluations of the LOTOS-EUROS model performance in both configurations are used to investigate current uncertainties in air quality forecasting in relation to driving meteorological parameters and to assess the potential for improvements in ~~high-resolution air quality forecasting episodes~~ forecasting pollution episodes at high-resolutions based on the HARMONIE NWP model.

Keywords: WINS50, LOTOS-EUROS, HARMONIE (cy43), ECMWF, Offline coupling, TROPOMI

1 Introduction

~~Meteorological fields calculated by~~ Numerical Weather Prediction Models (NWP) provide ~~necessary input to~~ the meteorological data required by Chemical Transport Models (~~CTM) to solve~~ CTMs) to resolve the emission, ~~transport~~transportation, chemical reactions ~~,~~ and other atmospheric interactions of pollutants ~~over the spatiotemporal domain~~ throughout the spatio-temporal field of interest (Chang, 1980; El-Harbawi, 2013; Khan and Hassan, 2020). In this way, forecasts and (re-)analyses provided by NWP can be used for air quality forecasting, climate modeling, and environmental studies. The more precise the meteorological input data represents the atmospheric dynamics, the better the CTM represents pollutant transport, mixing, and the subsequent impact on surface air quality. Meteorological parameters related to transport and mixing have a direct impact on the surface air quality simulated by ~~the~~ a CTM. A NWP model with a higher spatial resolution and better capabilities for resolving boundary layer turbulence dynamics and convective processes would provide ~~the~~ a CTM with more accurate input parameters to predict the ~~movement~~ transport of pollutants, especially in the lowest kilometer(s) of the troposphere (Pielke and Uliasz, 1998).

However, it is important to note that the spatial resolution of the NWP model is not the only factor. Other factors may include the model's ability to accurately represent small-scale phenomena, turbulence dynamics, and convective processes (non-hydrostatic), compared to models that replace the vertical momentum equation by hydrostatic equilibrium (SAITO et al., 2007). Also, the quality of (operational) meteorological input is constantly improved through the data assimilation applied in NWP (Marseille and Stoffelen, 2017; Bengtsson et al., 2017; Lorenc and Jardak, 2018) which can reduce ~~the model uncertaintyrepresentation~~model uncertainty. Overall, it is important to carefully consider the uncertainty of the meteorological driving parameters in a CTM, as these parameters can significantly affect the accuracy and reliability of the ~~simulated~~ air quality predictions.

HARMONIE (HIRLAM ALADIN Research on Mesoscale Operational NWP in Euromed) ~~, (Bengtsson et al., 2017) is the operational~~ pertains to a script system and model configuration in meteorological modelling (Bengtsson et al., 2017; van Stratum et al., 202 . It is named for the scripting system utilized for data assimilation, observation handling, and operational processes in the Applications of Research to Operations at Mesoscale (AROME) model within the countries utilizing the High-Resolution Limited Area Model (HIRLAM). Additionally, "HARMONIE" denotes a particular setup of the AROME model. This updated configuration includes physical parameterizations specifically adapted for European conditions, particularly at northern latitudes. The operational high-resolution NWP model that is used in The Netherlands is generated with the HARMONIE model configuration (Haakenstad et al., 2021). The ~~WINS50 is the~~ dataset that is used in this work ~~, it is an~~ is denoted as WINS50, which is a homogeneous HARMONIE reanalysis focusing on the North Sea region, developed by a consortium of Whiffle, TU Delft, and KNMI. The dataset covers the years 2019 to 2021 and has been created using HARMONIE cycle 43. It was evaluated for one year by (van Stratum et al., 2022), to show how and to what extent current wind farm structures in the north sea can cause effects on the meteorology at local to regional scales (Verzijlbergh, 2021; Kalverla et al., 2019; Baas et al., 2022).

LOTOS-EUROS (LOng Term Ozone Simulation-EURopean Operational Smog model) is a chemical transport model CTM that simulates the formation and transport of pollutants and trace gases in the atmosphere (Manders et al., 2017). The processes in the model include emission, advective transport, turbulent mixing, chemical reactions, wet- and dry deposition, and sedimentation. It is a CTM that is one of the members of the Copernicus Atmosphere Monitoring Service (CAMS) regional ensemble of CTMs that is used to produce operational air quality forecasts over Europe and at a higher spatial resolution also over the Netherlands. In most applications, the model is driven by meteorological input from ECMWFthe European Centre for Medium-Range Weather Forecasts (ECMWF), but in this study, it has also been coupled with the HARMONIE NWP to provide a more comprehensive understanding of the formation and transport of air pollutants in the BeNeLux countries and North Sea region. In earlier studies, other meteorological drivers have been offline one-way directional coupled to the LOTOS-EUROS model in one-way direction, including WRF (Escudero et al., 2019), COSMO (Thürkow et al., 2021), and, in RACMO (Manders-Groot et al., 2011)a and COSMO (Thürkow et al., 2021). A two-way coupling was implemented with frequent coupling between NWP and air quality simulations to provide insight in between the RACMO climate model and the CTM to provide information on the impact of meteorological conditions on air pollutants, and vice versa the impact of trace gasses gases and aerosol on weather and climate via for example the radiation budget (Manders-Groot et al., 2011).

In a previous study by (Ding, 2013), the impact of using HARMONIE (cy36) as a meteorological driver for LOTOS-EUROS (v1.8) was compared with using European Centre for Medium-Range Weather Forecasts (ECMWF) meteorology the standard ECMWF meteorology as driver. That study found large differences in the meteorological variables obtained from the two drivers, especially at the coast, over forest regions, and in urban areas. However, the surface temperature, relative humidity, and wind patterns were found to be very similar between the models. Since this previous study, various updates and improvements have been made to both the HARMONIE NWP model and the LOTOS-EUROS CTM, which have involved into cycle 43 and version v2.2002 .2.002, respectively. Therefore, conducting a new assessment and reassessing reassessment of their coupled performance is valuable.

Section 2 of this paper introduces the methodology used in the study. It includes a description of the two meteorological input fields with in the configurations made for the coupling with the state-of-the-art version of LOTOS-EUROS used in this study. The coupling procedure between the meteorological driver and the CTM is explained in this section, along with the list of variables taken into account and any necessary calculations or assumptions for their correct ingestion by into the CTM. Section 3 presents the results of the model simulations and their evaluation against ground-base ground-based observations and satellite-observed trace gas plumes. The comparison with observations is important to better assess provide an independent assessment of the differences between the model simulations. The paper's final section, Section 4, discusses our results and provides the conclusions on the coupling of HARMONIE WINS50 NWP to LOTOS-EUROS as to the extent these can be drawn from this study. Additionally, the potential for improvements improvement in high-resolution air quality forecast forecasts that are offline driven by high-resolution non-hydrostatic meteorological parameter fields data is assessed.

2 Methodology: Coupling of Meteorological Drivers to the Chemical Transport model Model

2.1 LOTOS-EUROS driven by ECMWF meteorology

LOTOS-EUROS is a large-scale three-dimensional CTM that simulates air pollution in the lower troposphere by solving a differential equation involving different operators, such as the transport operator, the chemical reaction operator, and the emissions/deposition operator. This These operators are executed sequentially on a 3D set of grid cells covering the troposphere over the domain of interest. The horizontal advection is driven by horizontal winds (U, V) that are part of the meteorological input. When driven by ECMWF meteorology, the model calculates the vertical wind component (W) through the convergence and divergence of the horizontal winds. Turbulence driven vertical diffusion is modelled with a seperate separate operator. The chemistry operator simulates the chemical production and loss terms from the different chemical reactions in the atmosphere. A Carbon Bond Mechanism with 81 reactions (Schaap et al., 2008) is used to describe the gas-phase chemistry, and interaction with aerosols follows the ISORROPIA parameterization parameterisation (Fountoukis and Nenes, 2007). The dry deposition operator is parameterized parameterised following the resistance approach (Wichink Kruit et al., 2012). The wet deposition operator includes the below-cloud scavenging for gases (Schaap et al., 2004).

LOTOS-EUROS receives the ECMWF Integrated Forecasting System (IFS) meteorological fields on a regular longitude-latitude grid, which is then interpolated to the target grid that is either regular longitude-latitude too or uses a different projection. The vertical layers of the model are defined as a coarsening of the ECMWF hybrid sigma-pressure layers. The meteorological fields received from the ECMWF data include 3D fields of pressure, wind vectors, temperature, and humidity, as well as 2D fields of mixing layer height, precipitation rates, cloud cover, and other boundary layer and surface variables, among others, listed in table 1 . A full overview of the meteorological fields is listed in Table 1 and described in the following section, are used to drive the transport and concentration rates of pollutants in the atmosphere. A simulation with LOTOS-EUROS driven by ECMWF meteorology has been performed to serve as a reference for other simulations, and this will be referred to as "EC_LE".

2.2 LOTOS-EUROS driven by HARMONIE meteorology

The HARMONIE (HIRLAM ALADIN Research on Mesoscale Operational NWP in Euromed) is a non-hydrostatic convection-permitting Numerical Weather Prediction model (Engdahl et al., 2020; Clark et al., 2016). In a non-hydrostatic model, the vertical momentum equation is solved directly instead of applying the hydrostatic approximation, which frequently fails during extreme weather events (Gibbon and Holm, 2011). HARMONIE incorporates various dedicated sub-models to describe atmospheric processes. One of these models is SURFEX, which simulates processes such as temperature and water balance, radiation balance, and heat transport at the surface and in the soil (Viana Jiménez and Díez Muyo, 2019). The model accounts for various types of land surfaces and processes at and below the surface to describe the interaction between the atmosphere and the surface.

Similar as to the ECMWF model, the HARMONIE model uses terrain-following hybrid sigma-pressure layers that are defined by surface pressure and hybrid level coefficients provided in the data files; Although the HARMONIE model could provide non-hydrostatic vertical advective fluxes, it was decided for this study to perform a coupling with HARMONIE based on the same approach as used for ECMWF variables (see ~~our~~ also the discussion in Section 4).

The particular HARMONIE simulation for this ~~project~~ study comes from the "WINS50" project. TUDelft, Whiffle, and KNMI have formulated the WINS50 project in the framework of the TKI Wind op Zee R&D 2019 (www.wins50.nl). The WINS50 model was run for 2019-2021 to produce winds undisturbed by wake effects (extension of the Dutch Offshore Wind Atlas DOWA) and disturbed winds (wake-DOWA). ~~The simulation was performed with the LOTOS-EUROS driven with ECMWF meteorology (EC_LE) and the LOTOS-EUROS driven with the HARMONIE meteorology (HA_LE). One recent comparison~~ Kalverla et al. (2019) compared the simulations of the HARMONIE model ~~for~~ over the North Sea with other models and ~~also observation~~ with observations from a mast~~to compare a couple of vertical levels can be found in (Kalverla et al., 2019)~~.

~~First, the data was moved from ECGATE to SNELLIUS.~~

2.2.1 Coupling choices

To ensure successful coupling of the HARMONIE/LOTOS-EUROS system, a systematic approach was taken comparing the available ECMWF and HARMONIE fields. This involved classifying the variables into three categories: static, surface, and 3D fields as shown in Table 1. The table was created to compare the variables' acronyms, units, and availability between the two systems. A simulation with LOTOS-EUROS driven by HARMONIE meteorology has been performed and this will be referred to as "HA_LE". LOTOS-EUROS ingested the variables selected from ~~the HARMONIE WINS50 correspondent to the~~ HARMONIE that correspond to equivalent ECMWF variables based on the coupling choices specified in the ~~following~~ next section. Second, ~~the decision~~ decisions are taken about whether direct or indirect mapping should be done and what to do with missing variables~~is taken. Third~~. Third, the labeling and timestamp frequency and time bounds were corrected and the direct paths to find the data and meteorological files were generated for the LOTOS-EUROS files. Mapping ~~Halflevel altitudes with Half level~~ half-level altitudes with half-level pressures with coefficients ~~calculation~~ calculations was done using specific routines ~~generated~~ that additionally flip the order of some ~~needed~~ of the required variables. Additionally, determining and converting ~~the~~ variables needed in either accumulated or instantaneous ~~formats~~ format was another task~~that was paid attention to.~~

2.2.2 ~~Coupling choices~~

~~To ensure successful coupling in the system (HA_LE), a systematic approach was taken comparing the available ECMWF and HARMONIE fields. This involved classifying the variables into three categories: static, surface, and 3D fields in Table 1. The table was created to compare the variables' acronyms, units, and availability between the two systems. The resulting comparison helped identify which variables could be used immediately, which required further calculations, and which needed to be excluded due to unavailability. The coupling strategy was built under the assumption we wanted to emulate how currently, the LOTOS-EUROS ingest datafrom the ECMWF fields (EC_LE). This table represents the static variables in purple, the dynamical two-dimensional in red, and the dynamical three-dimensional fields in green. This~~ . The coupling strategy for

HARMONIE data in this study was defined with goal to emulate the coupling with ECMWF data, thus so far without using variables that are only available from HARMONIE. This thorough approach ensured that the (HA_LE) system is technically coupled, allowing for the generation of accurate and comprehensive CTM fields driven by this new source of ~~meteorology information.~~ meteorological information. Surface dewpoint and friction velocity for grass were not available in the HARMONIE data. For the surface dewpoint the approximation in Lawrence (2005) was used. The friction velocity for grass was calculated by dividing the surface stress by air density and taking its square root.

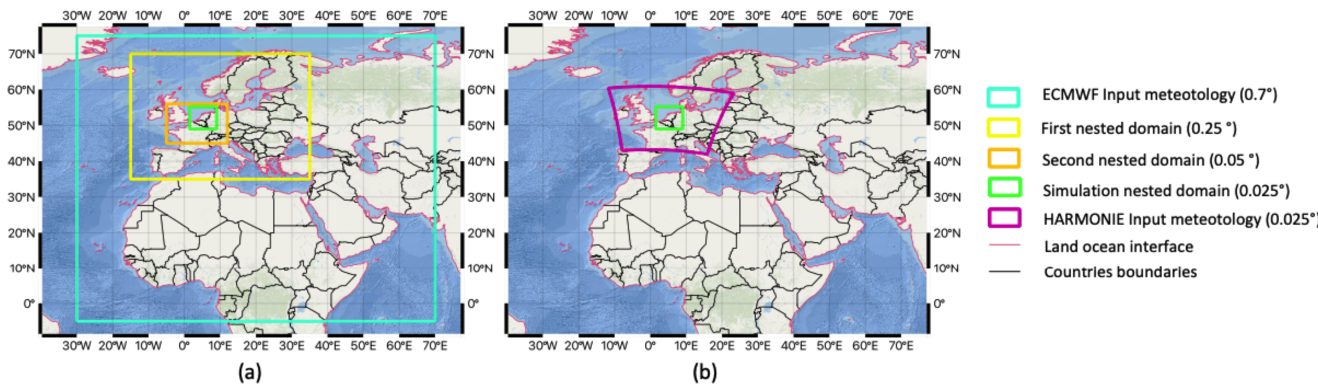

Figure 1. ~~Configurations of the two meteorology drivers~~ Simulation domains used for the LOTOS-EUROS ~~CTM. On the left LOTOS-EUROS nested domains~~ simulations using ECMWF ~~meteorology, and on the right the LOTOS-EUROS domain using~~ (left) or HARMONIE (right) meteorology. Both configurations use boundary conditions from CAMS. (map from Natural Earth collection (https://www.naturalearthdata.com/ ~~1:50m Natural Earth I with Shaded Relief and Water)~~

Table (~~??)~~ 2) and (3) shows the LOTOS-EUROS configuration settings for the simulations performed in this study. ~~Those configuration settings are essential for understanding the methods used in this study and for interpreting the results, with the main difference between the system from the meteorology input. The other parameters were kept equal to isolate the effects of the meteorology changes and attribute any discrepancies to this factor. Using different meteorological models allows for comparing the resulting NO_2 concentrations while keeping the other parameters constant, allowing for a more accurate assessment of the effects of the meteorologychanges on the simulations~~The simulations differ from each other in the meteorological driver. The rest of the parameters were not touched to attribute the discrepancies only to the change in meteorology. The table lists the different parameters used in the two LOTOS-EUROS configurations, including the meteorological data source, the chemical boundary conditions, ~~the~~ emissions, land use, ~~the~~ horizontal resolution for the objective domain and for the nested domains, and the time step used for the simulations.

Table 1. ~~Comparison between~~ Overview of the ~~ECMWF~~ meteorological input data of the IFS (~~fields from ERA5 Levels 137 converted to levels 42~~Integrated Forecasting System) provided by the ECMWF and the HARMONIE ~~WINS 50 (cy 43)~~ WINS50 meteorological variables ~~, their acronyms, and units~~used for the coupling to LOTOS-EUROS. The variables are divided into ~~static (purple)~~1-Static surface fields, ~~dynamical two (red),~~ 2-Surface and other dynamic 2D variable, 3- three ~~dimensions (green)~~dimensional variables. ~~Variables~~ HARMONIE variables with the symbol (*) were converted from instantaneous to accumulated.~~The variables underlined were calculated with other available variables~~

ECMWF		HARMONIE		Units
Acronym	**Long name**	**Acronym**	**Long name**	
1- Static surface fields				
lsm	Land sea mask	*lsm*	Sea area fraction	[0,1]
orog	Orography	*orog*	Surface altitude	[m]
slt	Soil type	*slt*	Soil type	
2- Surface and other dynamic 2D model				
blh	Boundary layer height	*zmla*	Atmosphere boundary layer thickness	[m]
tsurf	Surface temperature	*ts*	Surface temperature	[K]
dsurf	Surface dewpoint		Calculated from hus and ts using Lawrence approximation	[K]
u10	10 meter wind vector	*uas*	Eastward Near-Surface Wind Velocity	[m s^{-1}]
v10	10 meter wind vector	*vas*	Northward Near-Surface Wind Velocity	[m/s]
sd	Snowdepth	*snw*	Surface snow amount	[m]
sstk	Sea surface temperature	*sst*	Sea surface temperature	[K]
swvl1	Volumetric soil water layer N	*wsa_L01.P01*	Volume Fraction Of Liquid Water In Soil Layer 1	[m^3 m^{-3}]
swvl2	Volumetric soil water layer N	*wsa_L02.P02*	Volume Fraction Of Liquid Water In Soil Layer 2	[m^3 m^{-3}]
swvl3	Volumetric soil water layer N	*wsa_L03.P03*	Volume Fraction Of Liquid Water In Soil Layer 3	[m^3 m^{-3}]
swvl4	Volumetric soil water layer N	*wsa_L04.P04*	Volume Fraction Of Liquid Water In Soil Layer 4	[m^3 m^{-3}]
tcc	Total cloud coverage	*clt*	Total cloud fraction	[0 1]
zust	Friction velocity grass		Calculated from wind with square(Tauu+Tauv)/density	
sshf	Surface sensible heat flux	*hfss*	Accumulated Surface Upward Sensible Heat Flux	[J m^{-2}]
slhf	Surface latent heat flux	*hfls_eva*	Accumulated Upward latent flux of evaporation (*)	[J m^{-2}]
cp	Convective precipitation	*prrain*	Accumulated rain (*)	[kg m^{-2}]
lsp	Large scale precipitation	*prrain*	Accumulated rain (*)	[kg m^{-2}]
sf	Snowfall	*prsn*	Snowfall amount (*)	[kg m^{-2}]
ssrd	Surface solar radiation downwards	*rsds*	Accumulated Surface Downwelling Shortwave Radiation (*)	[J m^{-2}]
sp	Surface pressure	*ps*	Surface air pressure	[Pa]
3- Dynamic model 3D fields				
hp	pressure at layer interfaces	*hp*	pressure at layer interfaces	[Pa]
t	Temperature	*ta*	Air temperature	[K]
q	Specific humidity	*hus*	Specific humidity	[kg kg^{-1}]
v	v component of wind	*va*	Northward wind velocity	[m s^{-1}]
u	u component of wind	*ua*	Eastward wind velocity	[m s^{-1}]
cc	Cloud cover	*clt*	Total Cloud Fraction	[0-1] [kg kg^{-1}]
clwc	Specific cloud liquid water content	*clw*	Cloud water	[kg kg^{-1}]

Table 2. LOTOS-EUROS configuration settings for the simulations in this work . The principal difference is the input of the with HARMONIE meteorology input. The rest Coordinates of the parameters were not touched to attribute the discrepancies only to the change in meteorology. coordinates of the domain presented in [Lat N, Lon E].

Simulation periodsCharacteristic	HARMONIE Details
Simulation Periods	1 April to 30 April 2019
Meteorology	ECMWF; Temp.res: 1h; Spat.res: 0.7° **Meteorology** HARMONIE WINS50; Temptemp.res: 1h; Spatspat.res: 0.025°
Initial and boundary Boundary Conditions	CAMS (D1). Temp.; temp.res: 1h. **conditions** ; Spat.Res: 0.9°
Anthropogenic emissionsEmissions	CAMSSpat; spat.res: 0.1°
Biogenic emissionsEmissions	MEGANSpat; spat.res: 0.1°
Fire emissionsEmissions	MACC/CAMS GFASSpatCAMS GFAS; spat.res: 0.1°
Land useUse	CLC 2012. Spat2012; spat.res: 0.01°
Topography	GMTED2010. Spat.res: 0.002°
HARMONIE WINS50 (Lagrangian projection)	[-8.5°, 43°] x [16°, 42°]x[23°, 59°] x [-12°, 61°]
Objective simulation grid [Lat] x [Lon] (Both configurations)	[49°, 13.27°] x [1.5°, -65.94°]

2.2.2 About the computational Computational aspects

The Figure (1) shows two the spatial configurations of the LOTOS-EUROS CTM that use different used with a specific meteorology drivers. The configuration on the left has three nested domains and uses ECMWF meteorology , while the configuration on the right has one domain and uses HARMONIE meteorology. Both configurations use boundary conditions from CAMS. Using nested domainsin the first configuration allows for using ECMWF meteorology on panel (a) has three nested domains, which is common practice for more precise modeling modelling of atmospheric conditions in areas with coarse boundary information. In contrast, the second configuration has a high-resolution meteorology information. meteorological information. The configuration for the HARMONIE meteorology on the panel (b) only uses one domain; in this case there is no nesting needed because the input resolution of the meteorology already has the intended resolution for the CTM; the green box is the actual domain for the "final" simulations which is the same for both configurations.

Using a nested domain simulation that reduces from three nested simulations in the configuration (EC_LE) to only one in the configuration (HA_LE) to reach the concentration simulations The objective of the simulations is to obtain CTM simulations at 0.025° as the objective can provide over The Netherlands and the North Sea. For this, the HARMONIE/LOTOS-EUROS coupling uses only one nested simulation, while the ECMWF/LOTOS-EUROS coupling uses three nested simulations. The single nested configuration provides significant computational benefits. By comparing the performance of the new approach with the traditional three-nesting method, we found that the computational cost was reduced by a factor of four The computational resources required for the single-level approach are a factor four lower than the costs of the three-level nesting approach, while

Table 3. LOTOS-EUROS configuration settings for the simulations in this work with the ECMWF meteorology input. Coordinates of the domain presented in [Lat N, Lon E].

Characteristic	**ECMWF Details**
Simulation Periods	1 April to 30 April 2019
Meteorology	ECMWF; temp.res: 1h; spat.res: 0.7°
Initial and Boundary Conditions	CAMS (D1); temp.res: 1h; Spat.Res: 0.9°
Anthropogenic Emissions	CAMS; spat.res: 0.1°
Biogenic Emissions	MEGAN; spat.res: 0.1°
Fire Emissions	CAMS GFAS; spat.res: 0.1°
Land Use	CLC 2012; spat.res: 0.01°
Topography	GMTED2010. Spat.res: 0.002°
ECMWF [Lat N x Lon E]	[-5°, 75°] x [-30°, 70°]
First ECMWF nested domain Nested Domain [Lat] x [Lon]	[35°, 70°] x [-15°, -35°]
Second ECMWF nested domainNested Domain [Lat] x [Lon]	[45°, 18°] x [5°, -60°]
Objective simulation grid [Lat] x [Lon] (Both configurations)	[49°, 13.27°] x [1.5°, -65.94°]

maintaining comparable accuracy in the results. This was achieved because the resolution of HARMONIE ensured that the boundary conditions were more input meteorology were comparable in terms of spatial resolution and was doable to go directly to the simulation objective of the simulation domain objective, and could be used directly at the target grid. The reduction in the number of nested domains led to a substantial reduction in the computational resources required for the simulation, enabling us to tackle larger and more complex problems with the same resources. Overall, the results of our study highlight the significant benefits of using a nested domain simulation with fewer levels of nesting and demonstrate its potential as a powerful tool for numerical simulations. HARMONIE operational data files are provided in 'grib' format. Standard and freely available. Each hourly gribfile has a file size of 200 Mb. Over two days, 16 runs are performed for each hour. Only 1/16th of the data volume provided will be needed to drive a CTM (5 Gb / day) for a given forecast lead time and time window.

2.3 Cabauw meteorology information

2.3 Observations

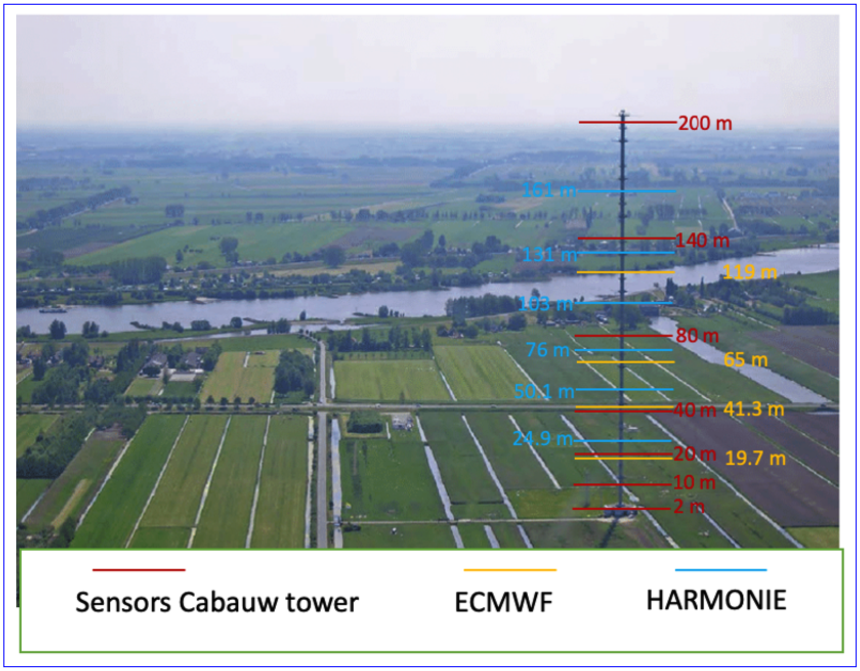

Figure 2. ~~(a) Time series~~ Image of the ~~temperature from the ECMWF meteorology compared with the~~ Cabauw ~~observations compared for different levels and (b) the image from the Cabauw~~ tower (lat 51.96° N, lon 4.89°W) with ~~three colors for~~ in red the locations of the meteorological sensors, and in yellow and blue the interfaces between the ECMWF and HARMONIE model ~~levels for comparison, aerial~~ layers. Aerial photo image modified from (Apituley et al., 2008).

During April 2019, we observed two distinct weather patterns which changed the atmospheric conditions within the month and could be attributed to variations in wind speed and direction. Evidence for the change in meteorological conditions is provided by the directionality of plumes captured by satellite instruments, as illustrated in the subsequent figures 1 and 2 in the appendix section. We compared the model simulations with ground-based observations derived from the air quality network during these periods in April 2019.

2.3.1 Cabauw meteorology observations

Meteorological observations from the Cabauw site have been used to validate the meteorological data sets used in this study. The 213-meter tall KNMI-mast ~~Cabauw generates continuum~~ in Cabauw (Figure 2) generates continuous and stable meteorological ~~measurements for~~ observations at a location with homogeneous characteristics in a central part of ~~the~~ The Netherlands. This site is located in a flat terrain with an elevation of 0 meters above sea level and has been used to validate models, satellite ~~information~~instruments, and other meteorological sensors (Bosveld et al., 2020). The surrounding area is mainly used for agriculture purposes; although the Cabauw tower is located in a rural area, small towns and villages are nearby. ~~The data for this experiment was downloaded from https://dataplatform.knmi.nl/dataset/cesar-tower-meteo-lb1-t10-v1-2 for the months~~

April-May-June-July-August For this study observations were downloaded from KNMI for the period April-August 2019. The data comes in 10 minutes Interval of sampling and contains the following variables: Air temperature, Dew 10-minute sampling intervals and contains air temperature, dew point temperature, Specific humidity, Wind specific humidity, wind speed, and wind direction.

2.3.2 Surface concentration pollutants information

Surface observations NO_2 have been used to validate the LOTOS-EUROS simulations. The NO_2 data was downloaded from the ground base sensor stations of different from (for the ground stations at different places in the Netherlands from www.luchtmeetnet.nl). The different locations along . Different locations in the country were chosen to compare the two NO_2 in the LOTOS-EUROS model configuration to cover the more representation possible systems with the different meteorological data sets in a representative way. This data is provided by *Rijksinstituut voor Volksgezondheid en Milieu* (RIVM). The RIVM is accredited for air quality measurements of SO_2, NO, NO_2, O_3, $PM_{2.5}$ and PM_{10} by the Dutch "Raad voor Accreditatie (RvA)" according to NEN-EN-ISO/IEC 17025:2018.

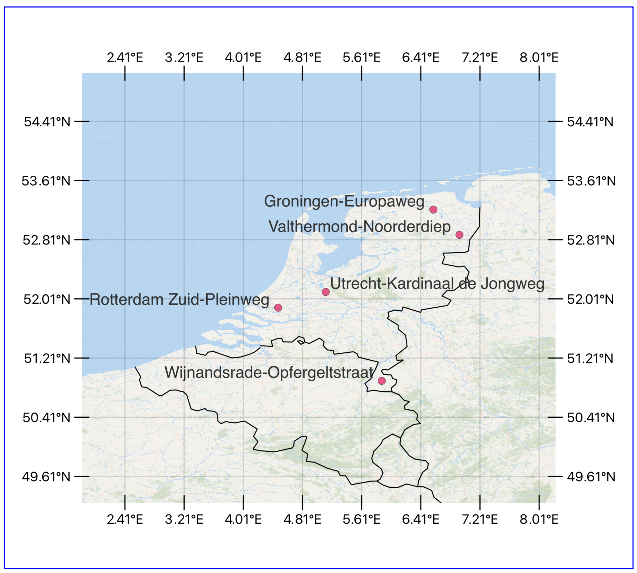

Figure 3. Map of the Netherlands with the air quality locations of the selected points to compare the simulations

2.4 TROPOMI

2.3.1 TROPOMI

The TROPOMI information was explored qualitatively because we wanted to establish a period from which we can have some TROPOspheric Monitoring Instrument (TROPOMI) is the satellite instrument on board the Copernicus Sentinel-5 Precursor (S5p) satellite. S5P is a low-Earth polar orbit satellite. The polar orbit and wide coverage of the scanner provide almost daily global coverage, with a spatial pixel resolution of 5.5 x 3.5 km^2. The TROPOMI instrument is a spectrometer sensing ultraviolet (UV), visible (VIS), near (NIR), and short-wavelength infrared (SWIR) wavelengths to monitor Ozone (O_3), Methane (CH_4), Formaldehyde (CH_2), Aerosol, Carbon Monoxide (CO), Nitrogen Dioxide (NO_2), and Sulfur Dioxide (SO_2). The NO_2 retrievals used in this study are retrieved from a wavelength range of 405–465 nm (spectral band 4). The Royal Netherlands Meteorological Institute (KNMI) created the TROPOMI NO_2 retrieval method based on the DOMINO NO_2 retrieval algorithm employed on the Ozone Monitoring Instrument (OMI) precursor instrument (Boersma et al., 2011). In this work, the NO_2 retrievals from TROPOMI were used to select a simulation period with well-defined characteristics to have a priory knowledge of the concentration state at the tropospheric and total column level, at least for the daily satellite snapshot. characteristics of the tropospheric NO_2 concentrations and to see if the different model simulations are able to represent this.

3 Results

3.1 Meteorology fields evaluation

Figure (2)compares the temperature ECMWF meteorology and the temperature Cabauw observations at different levels, as well as an image from Figure 2 provides an illustration of the Cabauw tower illustrating the sensor positions for comparison with the ECMWF and HARMONIE models. Panel (a) of the figure displays the time series of temperature from the ECMWF meteorology compared with the temperature Cabauw observations at different levels. The comparison shows some differences between the two datasets at certain levels, particularly during nighttime; the daily cycle is in phase, but there are differences in magnitudes. This suggests the importance of validating model outputs with ground-based observations.

The daily temperature cycle from ECMWF (a) and HARMONIE (b) models and Cabauw observations at different LOTOS-EUROS simulation levels. The RMSE for different levels is shown for the two input meteorological value compared against the sensors in the tower

Panel (b) of the figure provides an image from the Cabauw tower, with the positions of the sensors and the interfaces between ECMWF and HARMONIE models overlaid in three different colors model layers to illustrate the height of the varying model levels for comparison. This information is essential for validating the models' height levels and identifying potential sources of discrepancies between the model outputs and the observations in the height structurevertical domain.

Overall, the results in Figure (2)demonstrate the importance of validating model outputs with ground-based observations and the value of visualizing sensor positions and model outputs together for comparison. These findings can inform improvements to the models and ultimately lead to more accurate temperature and other meteorological variables predictions. A comparison between the observed and simulated temperatures at different levels is shown in Figure 4. Panel (a)displays the time series of temperature from the ECMWF meteorology compared withobservations at Cabauw at different levels. The comparison shows that there are some differences between the two datasets at certain levels, particularly during nighttime. The daily cycle is in phase, but there are minor differences in magnitude.

Figure (4) shows the daily cycle for three levels of the two meteorology input information to the data sets as provided to LOTOS-EUROS model compared with the respective height sensor in the compared to observations at the Cabauw tower. The

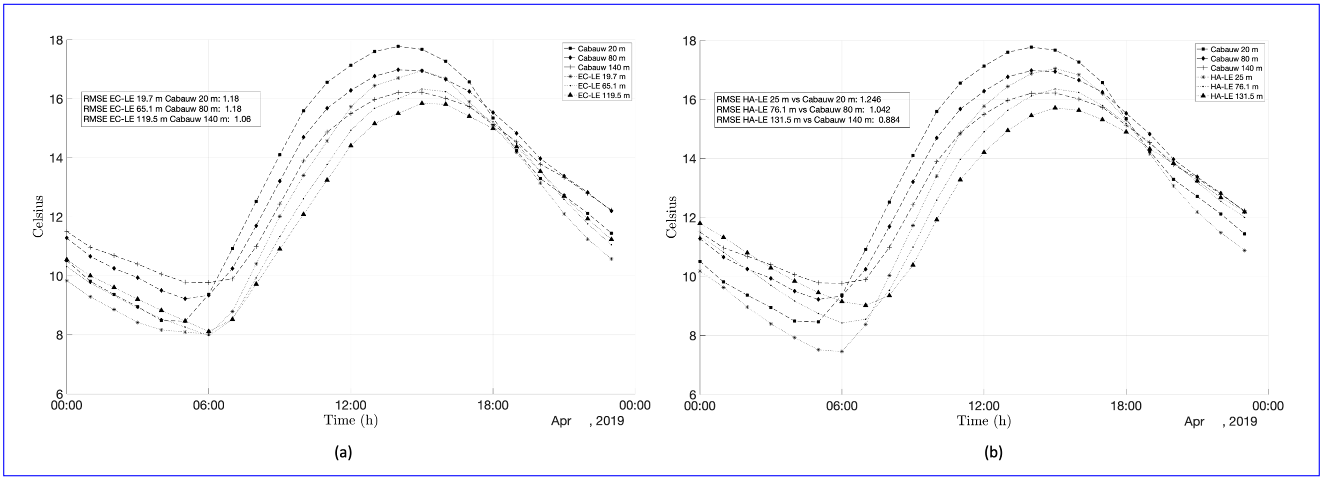

Figure 4. The daily temperature cycle from ECMWF (a) and HARMONIE (b) meteorological data and the Cabauw observations at different LOTOS-EUROS simulation levels. The RMSE for different levels is shown for the two input meteorological data sets in comparison to the temperature from the sensors in the tower

~~comparable~~ values show minor differences, which gives technical trust in the model ~~configuration~~configurations. For the height of 140 m from the Cabauw tower, the HARMONIE meteorology ~~got a~~ shows a bit lower RMSE, showing a bit better agreement with the ~~measurements in the extreme part of the day~~observations.

~~In~~ Figure (5) ~~can we see a spatial~~ shows a comparison of the wind ~~direction and magnitudes~~ speed and wind direction at the resolution of LOTOS-EUROS for the two meteorological input data sets. For the chosen time, the ~~model simulation, and on the right side of the image, some more statistical comparisons based on different metrics of this variable comparison over the red square over there map. When comparing the results, we found that the overall performance was comparable. However, there were~~ fields are very similar, although there are some differences in the details~~of these fields. These differences may have contributed to variations in the results observed between studies.~~

~~Despite the performance similarities, further investigation is needed to determine the most effective approach for achieving accurate results. Other results of~~ . Although small, such differences may contribute to differences in air quality at specific locations and variations in wind (direction) could be important when comparing model simulations with air quality observations from measurement sites. An extensive validation of the meteorological variables of the HARMONIE model ~~but in this case from the Dutch Offshore Wind Atlas (DOWA)~~ against Cabauw vertical ~~measurements,~~ profile observations can be found in (Knoop et al., 2020).

3.2 ~~Concentration~~ Comparison of concentration fields~~validation~~

~~We compared the surface concentration~~ Figure (6) compares the simulated surface concentrations of NO_2 for the ~~(~~EC_LE ~~)~~ ~~and (~~(a and d) and HA_LE ~~) configurations and visualized the results in Figure (6) . Panel (a) of the figure shows the surface~~

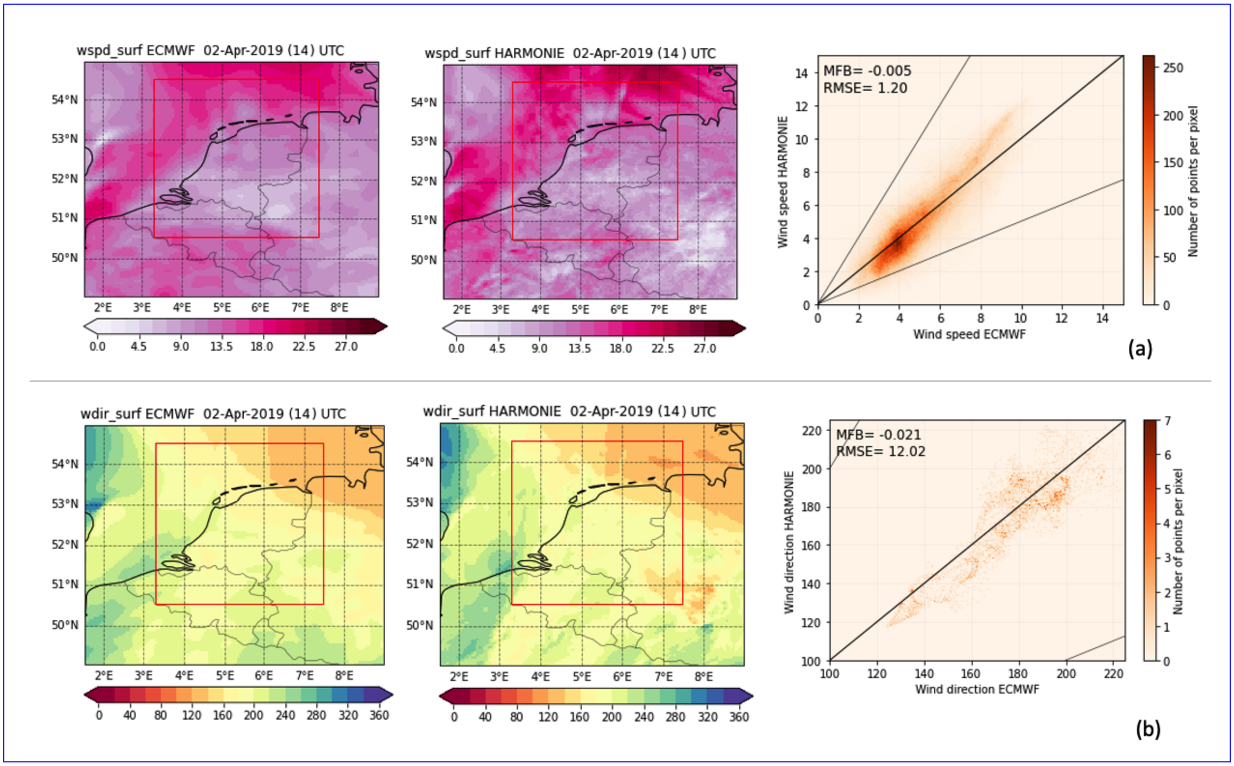

Figure 5. Instantaneous ~~spatial comparison between the~~ of surface wind speed [m/s] (wspd_surf) and direction [°] (wdir_surf) from ECMWF or HARMONIE meteorological fields interpolated to the ~~simulation resolution~~ LOTOS-EUROS grid~~, and in~~. The scatter density plots on the right ~~image, a quantitative comparison in~~ compare the values enclosed by the red square~~demarcated over The Netherlands where the RMSE and the MFB scores are shown~~. Base maps from http://www.gadm.org/

~~concentration of NO_2 for the (EC_LE)configuration, while panel (c) shows the surface concentration of NO_2 for the (HA_LE) configuration.~~

~~Air masses distinctions from the comparisons for the system configurations in volume mixing ratio of surface NO_2 mol mol^{-1}from (a) (EC_LE) and (c) (HA_LE). The middle panel (b) shows the fractional difference. Base maps from~~

(c and f) model configurations at two different moments: April 3, a day without a dominant wind direction (upper panels), and April 12, a day with a clear westward directed wind field. To gain further ~~insights~~ insight into the differences between the two configurations, ~~we included a difference comparison in panel (b). The difference comparison (~~the fractional difference ((EC_LE)-(HA_LE))/(~~HA~~EC_LE) ~~clearly shows~~ is shown in panels (b and e). These fractional differences clearly show that the (HA_LE) ~~configuration produces~~ model configuration produces similar but different NO_2 concentrations ~~than~~ compared to the (EC_LE) configuration at the ~~air mass of specific locations, revealing a wind direction difference indicated by the bias observed in the plumes depending on the meteorology uses to drive each model which can impact the time series~~ chosen time. This reveals a difference in wind direction in the meteorological drivers which could impact the simulated time series at any location. This finding suggests that wind direction can play a crucial role in the transport and diffusion of NO_2 in the atmosphere and can affect the accuracy of the modeled concentrations. This experiment shows could affect the simulated concentrations.

The experiment demonstrates that air mass characterizationbased on , e.g. informed by NO_2 concentration plume structures. The statistical metric lets us quantify the areas where the , may reveal significant discrepancies between HA_LE overestimates the and EC_LE , indicating the discrepancy between the two sources of information.

The fractional difference Specifically, the direction of the wind can influence the transport of simulations. Evidence is provided by the statistical metrics, quantifying areas of over and underestimation. Clearly, small-scale wind direction changes have an impact on NO_2 emissions from their sources to other areas, leading pollutant transport, and contribute to variations in the concentrations of the pollutant .

Our results provide insights into the factors contributing to variations in pollutant concentrations across different regions. Accurate meteorological inputs are invaluable for LOTOS-EUROS simulations, particularly for the understanding of time series of NO_2 concentrations in the Netherlands and underscore the need to carefully consider model configurations with meteorological input in atmospheric chemistry modeling. The a the stations of the gound network in the Netherlands.

An example of the tropospheric column of NO_2 for the (EC_LE) and (HA_LE) configurations, as well as the TROPOMI satellite retrieved information for this pollutant for the troposphere, are tropospheric column retrieved from the TROPOMI satellite instrument is shown in Figure (7) . for a single snapshot for 22 April 2019. Panel (a) of the figure shows the tropospheric column of NO_2 for the (EC_LE) configuration, while panel (b) shows the tropospheric column of NO_2 for the (HA_LE) configuration. Panel (c) shows the tropospheric column of NO_2 obtained from the TROPOMI satellite retrieval.

Table 4. Names and labels are displayed in Figure 7 for the largest emitters in the Netherlands, categorized by factories, refineries, and power plants using coal and gas as sources.

Factories and Refineries					
Tata Steel	1	Terneuzen	5	Gunvor Petroleum	9
Chemelot	2	Yara Sluiskil	6	Vitol/Koch/VPR Energy	10
DOW Benelux	3	Exxon Mobile Rotterdam	7		
Shell Rotterdam	4	BP	8		
Power plant (coal)					
Maaslavkte	11	Hemweg 8	13	Eemshaven	15
Maaslavkte MPP3	12	Gelderlan	14		
Power plant (gas)					
Sloe	16	Ijmond	20	HARCULO	24
Rijnmond II	17	Centrale Merwedekanaal	21	Magnum	25
ELSTA	18	Maxima	22	Eems	26
Diemen 33	19	Flevo	23	Delesto	27

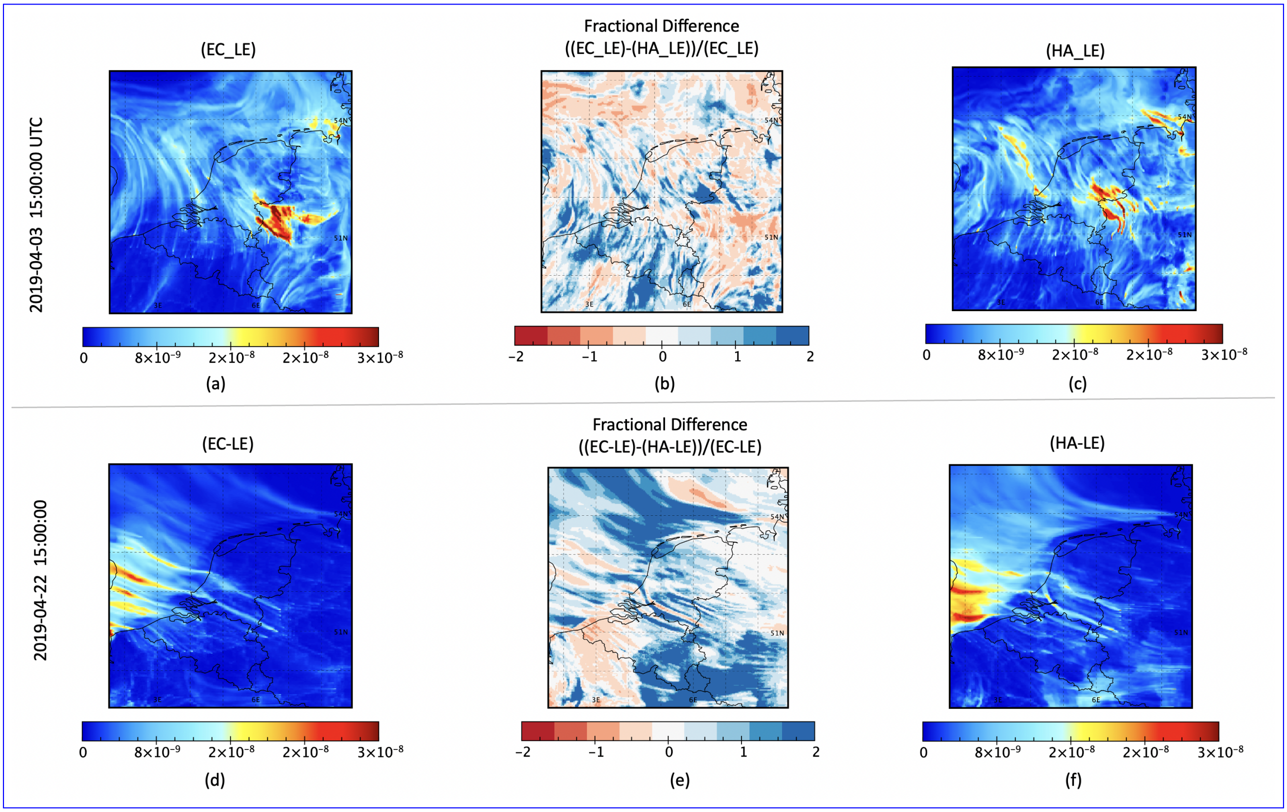

Figure 6. Maps of volume mixing ratio of surface NO_2 [mol mol^{-1}] at 2019-04-03 (top) and 2019-04-12 (bottom) at 15:00 UTC, from either EC_LE (a and d) or HA_LE (c and f) configurations. The middle panels (b and e) shows the fractional difference. Base maps from http://www.gadm.org/

The comparison reveals that the (HA_LE) configuration produces a tropospheric column of NO_2 that is slightly more similar to the TROPOMI satellite retrieval, particularly in regions with high NO_2 concentrations. This similarity is likely The difference with the EC_LE simulation is due to a slight change difference in wind direction in the HARMONIE configuration, which affects the transport and diffusion of NO_2 emissions in the atmosphere. In addition to revealing differences in NO_2 concentrations between the two model configurations and the satellite retrieval, the images in Figure (7) show different details over the maps. Specifically, the maps illustrate the locations of coal and gas power energy stations, oil rigs and pipelines, principal airports, and roads across the Netherlands. These details are important to consider in atmospheric chemistry modeling, as they can help to identify potential sources of NO_2 emissions and inform policy decisions related to air quality management.

Figure 8 presents a comprehensive analysis of air quality measurements obtained from Figure 8 shows a comparison between the NO_2 simulations and observations at three stations within the luchtmeetnet.nl national air quality network. The stations, namely locations compared are marked in Figure 7 with a red star: Station Utrecht Kardinaal de Jongweg (a) , is located in a central part of the country; Rotterdam Zuid-Pleinweg (b) , and is located in the city of Rotterdam and is characterized by high

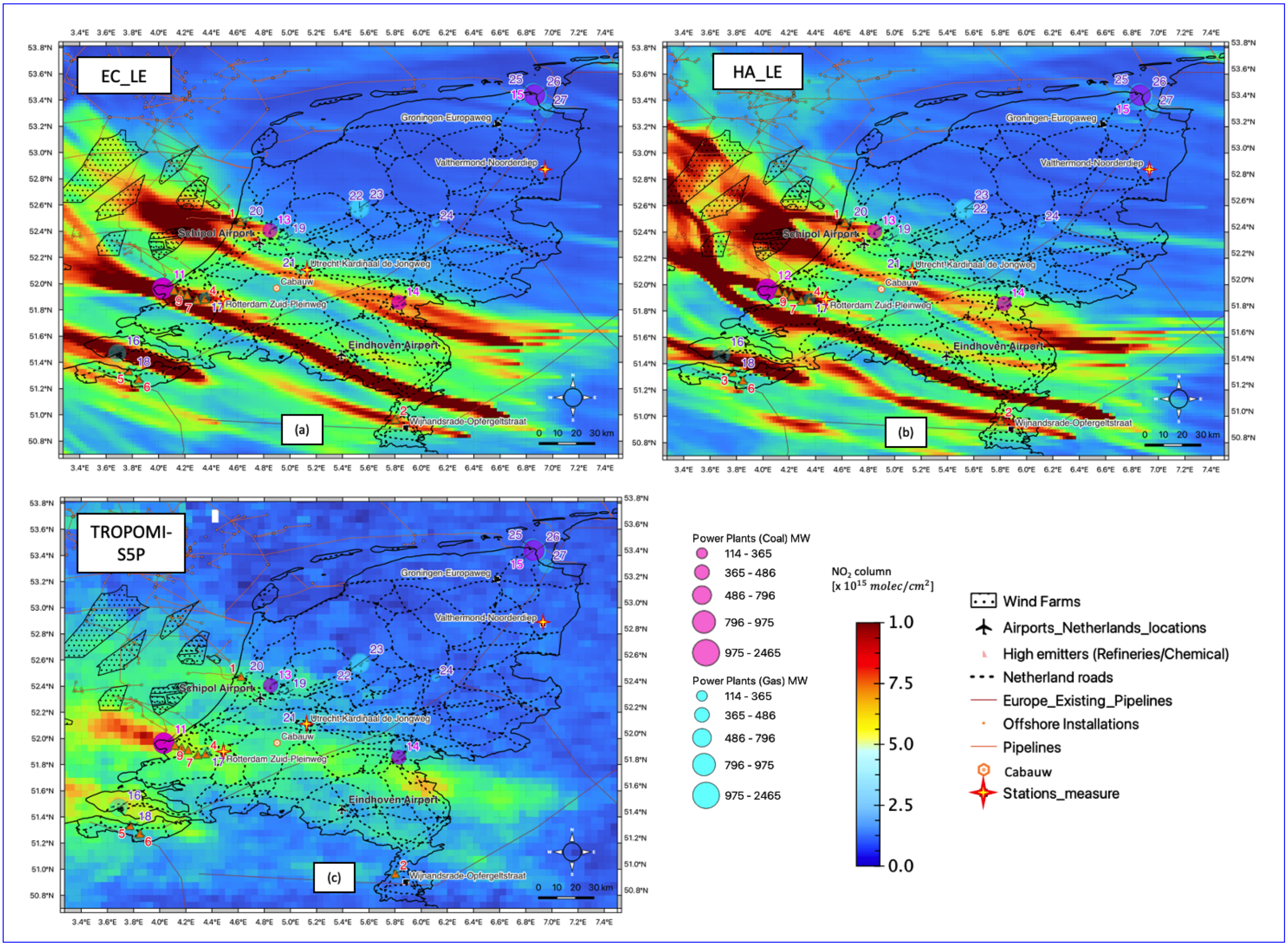

Figure 7. Comparison between the tropospheric ~~column~~ columns of NO_2 (EC_LE) (a) and (HA_LE) (b) for the ~~TROPOMI tropospheric column at the~~ overpass time ~~(c). Different characteristics~~ at 14:00 local of TROPOMI of which the retrieved tropospheric columns are shown in ~~the figures~~ (c) for 22 April 2019. Large sources of NO_x such as ~~the~~ power plants, principal airports, and main roads are marked. The ground ~~measurement station~~ observation stations depicted with a star are the stations shown in ~~the next figure~~Figure 8. ~~Units are different in the model and satellite column concentration shown but for the purpose of the comparisons the plume structure and direction is the intended.~~ Base maps from (http://www.gadm.org/) and information from (https://emodnet.ec.europa.eu/en/human-activities).

levels of pollutants, also due to the nearby presence of the harbor and refineries activities, and Valthermond Noorderlep (c) ~~, are compared against two model configurations depicted in the upper panel. The first configuration, ECMWF->LOTOS-EUROS, is visualized in orange, while the second configuration, HARMONIE->LOTOS-EUROS, is depicted in blue. The evaluation focuses on the representative error in dispersion, specifically examining the deviation of~~ is located in a more rural area. The

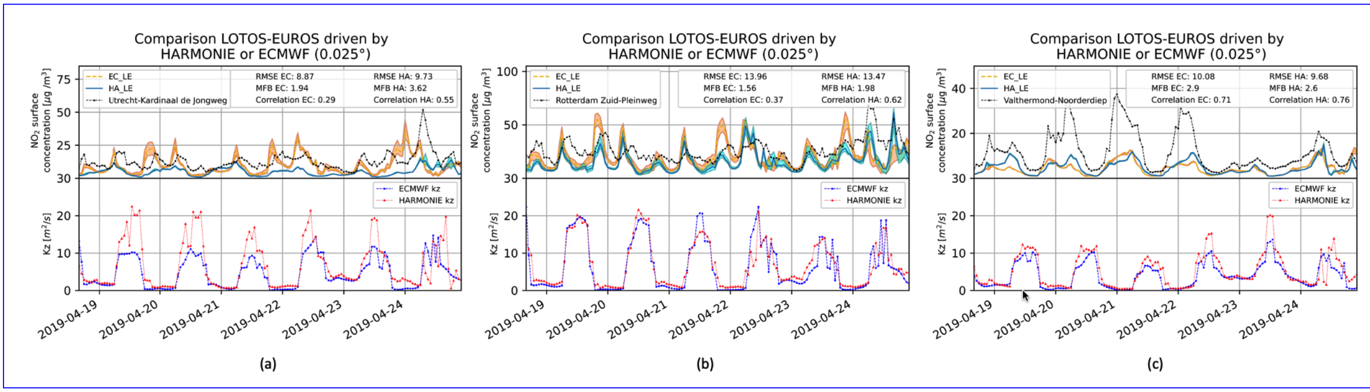

Figure 8. ~~Three~~ Comparison between NO_2 observations and simulations at 3 air quality stations from the ~~(www.luchtmeetnet.nl)~~ national network: (a) Utrecht Kardinaal de Jongweg, (b) Rotterdam Zuid=-Pleinweg, and (c) Valthermond Noorderlep~~compared with~~. Shaded area represents the ~~two model configurations in the upper panel (ECMWF->LOTOS-EUROS in orange and HARMONIE->LOTOS-EUROS in blue) taking the representative error in the dispersion such as the~~ standard deviation ~~of~~ between the grid cell where the station is located and the ~~immediate~~ surrounding cells~~around~~. The below panel shows ~~a comparison for~~ the ~~surface Kz~~ simulations of the vertical diffusion coefficient K_z between the surface and second LOTOS-EUROS model layer.

standard deviation between the grid cell where each station is located and its ~~immediate neighboring cells . The~~ neighbouring cells is added to the simulation time series to have a notion of the representative error of these site comparisons. Three statistics (RMSE, MFB, Correlation) are presented for each configuration in each location. The highest correlations with the observations over the full time period are obtained using the HARMONIE high-resolution meteorology. The lower panel of ~~the figure compares the surface Kz coefficient , offering~~ this figure shows the vertical diffusion coefficient K_z between the surface and the second LOTOS-EUROS model layer which offers additional insights into the analysis of ~~air quality data.~~ surface air quality observations. Kz values are high over the Rotterdam Zuid-Pleinweg station; for the other two stations, Utrecht Kardinaal de Jongweg and Valthermond Noorderlep, lower Kz values are found but with relatively higher values in the HARMONIE model configuration which suggest a locally higher vertical mixing in this model configuration.

The transversal cut over the Netherlands in Figure (9) shows a comparison between the (EC_LE) configuration in the upper panel and the (HA_LE) NO_2 fields in the panel below. The figure indicates notable differences in the NO_2 concentration fields produced by the two model configurations in both the NO_2 columns and the value of the K_z diffusion coefficient at the layer interfaces. The planetary boundary layer height is shown in all pictures with a shaded blue line. Here, the HARMONIE provides a more complex structure that impacts the modelled vertical mixing. In panel (a), the (EC_LE) configuration shows lower NO_2 concentrations ~~in some areas~~ compared to panel (b), where the (HA_LE) configuration produces higher NO_2 concentrations~~in the same regions. These differences~~. Note that the differences with the observations may be attributed to ~~using differentmeteorological and~~ the (different) meteorological drivers as well as to the (identical, but uncertain) emission data in the two ~~configurations, which~~ model configurations. Both meteorology and emission and chemistry uncertainties can affect the ~~model's ability to simulate atmospheric chemistry accurately~~ability of a CTM to simulate observations of atmospheric polutants.

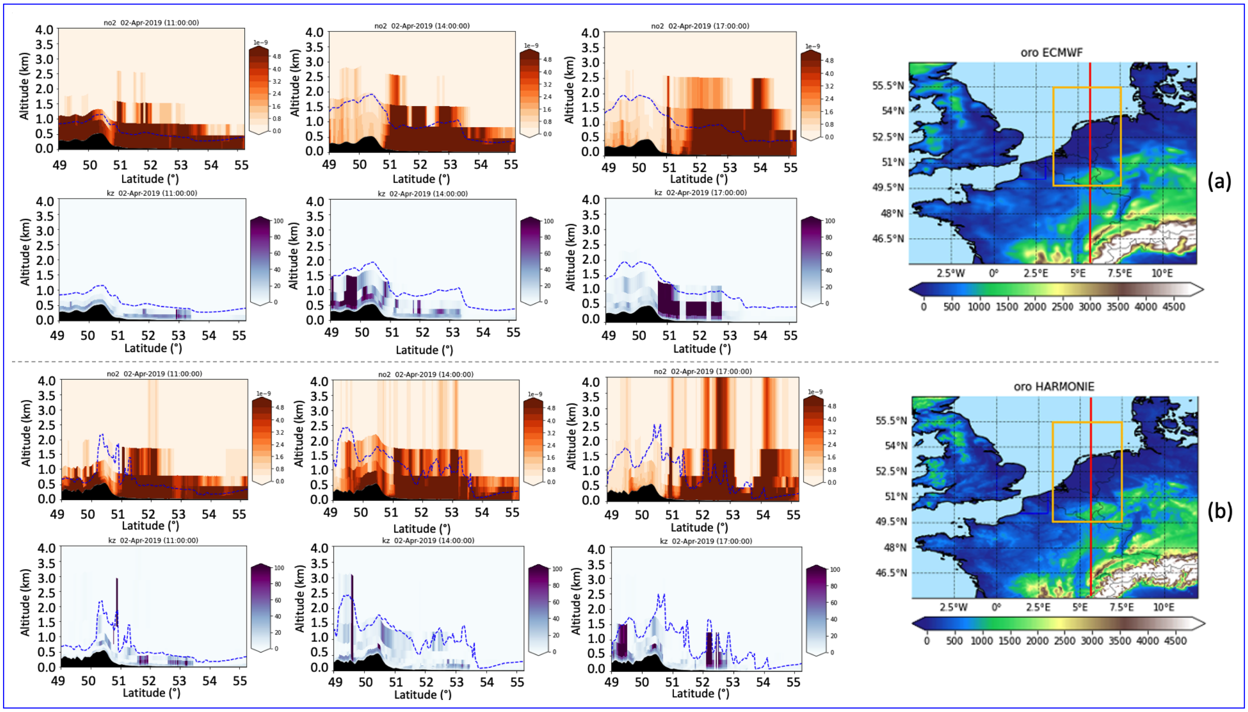

Figure 9. (a) Transversal cuts on longitude (6.2 ° E) over the Netherlands comparison between the (EC_LE) configuration ~~,~~ and (b) the (HA_LE) NO_2 concentration fields. The dashed blue lines correspond to the planetary boundary layer in the models. The panels on the right show each of the transversal cuts. Base maps from (http://www.gadm.org/).

~~Overall, comparing the two~~

Overall, our comparison of the two model configurations highlights the importance of ~~carefully selecting appropriate model configurations~~ an appropriate model configuration when evaluating NO_2 ~~2~~ concentrations in a given region ~~. Further~~ with a CTM at a given spatial resolution. More research is needed to investigate the specific factors ~~contributing~~ that contribute to the differences between the two ~~configurations and~~ model configurations for LOTOS-EUROS and to determine which configuration is more accurate for ~~NO_2 concentration modeling~~ simulating NO_2 concentration in the Netherlands. ~~The transversal cut over the Netherlands in Figure (9) shows a comparison between the (EC_LE) configuration in the upper panel and the (HA_LE) NO_2 fields in the panel below. The figure indicates notable differences in the NO_2 concentration fields produced by the two configurations in the columns and the value of the K_z diffusion coefficient at the layer interfaces. The planet boundary layer is shown in all pictures with a shaded blue line. Here, the HARMONIE provides a more complex structure that must prevail in the impact of vertical modeled transport.~~

Figure 10 compares both configurations for a mean of April for 4 levels of the NO_2 concentration and the diffusion coefficient.

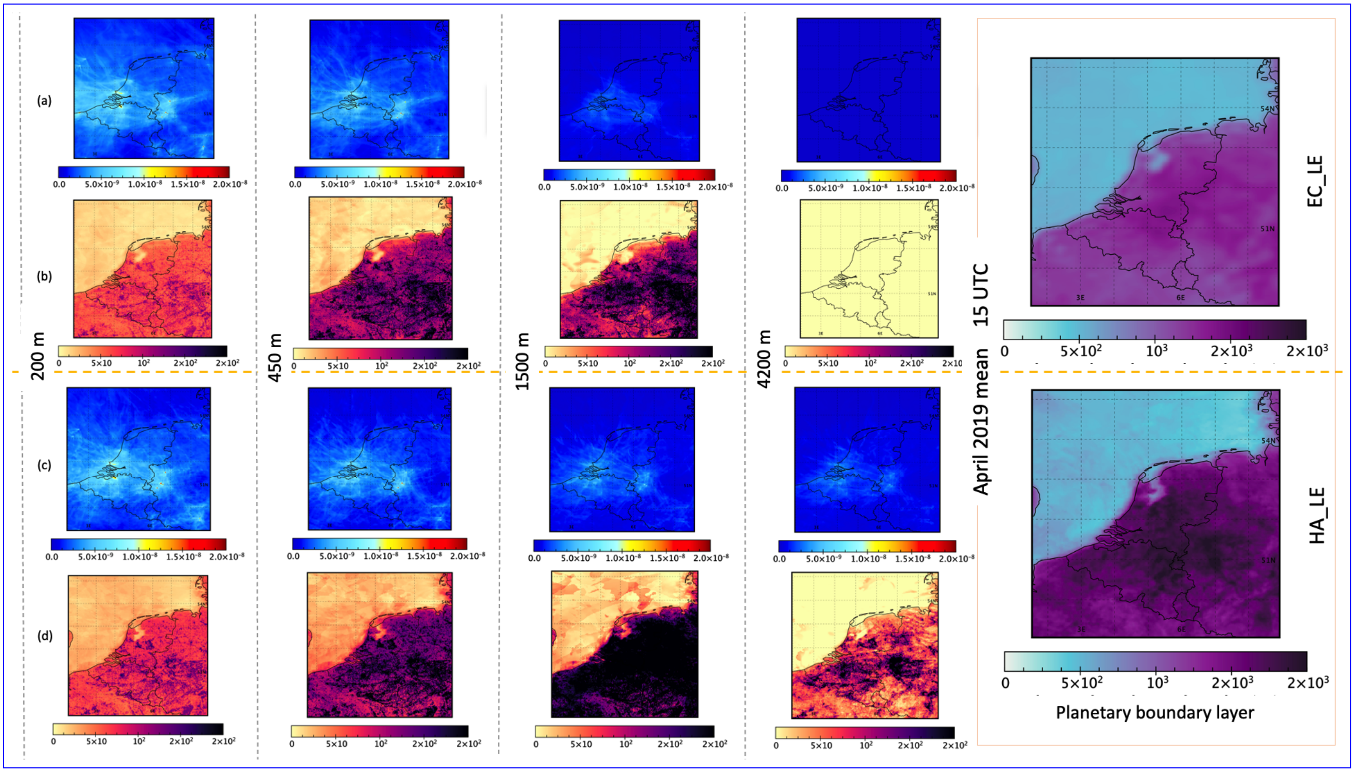

Figure 10. April mean (15 UTC) NO_2 concentration fields [mol mol^{-1}] and Kz [m~~[2]~~^{2} s^{-1}] at 200, 450, 1500, and 4200 m altitude (a,c) for EC_LE and (b,d) for HA_LE. Base maps from (http://www.gadm.org/).

The HARMONIE atmospheric model stands out with its enhanced structure and distinct field shape compared to the ECMWF. However, it exhibits a discrepancy when simulating the boundary layer height, overestimating it compared to real-world observations. This disparity significantly affects air pollutant concentrations, particularly ~~in the upper~~ at higher lvels in the atmosphere. The higher simulated boundary layer height in HARMONIE allows pollutants to be transported to higher altitudes, leading ~~to complex~~ tochanges in chemical reactions and the formation of secondary pollutants. ~~This phenomenon~~ The amount of upward mixing affects regional air quality, climate, and the understanding of long-range pollutant transport. Accurately representing the boundary layer height is therefore crucial for reliable air quality forecasts and ~~assessing~~ the assessment of (surface) pollutant impacts. Resolving this issue requires further research and refinement of the model's parameterizations and processes related to boundary layer dynamics, enabling improved simulations of pollutant ~~dispersion in~~ vertical dispersion into different atmospheric layers.

The comparison between the LOTOS-EUROS simulated retrieval of the tropospheric column of NO_2 and the TROPOMI average tropospheric vertical column which corresponds to the input needed for the data assimilation stage are shown in Figure

(11). Panel (a) of 11shows the TROPOMI average tropospheric vertical column Yr product, panel (c) the LOTOS-EUROS simulated retrieval of the tropospheric column of NO_2 Ys, while panel in the middle show the difference.

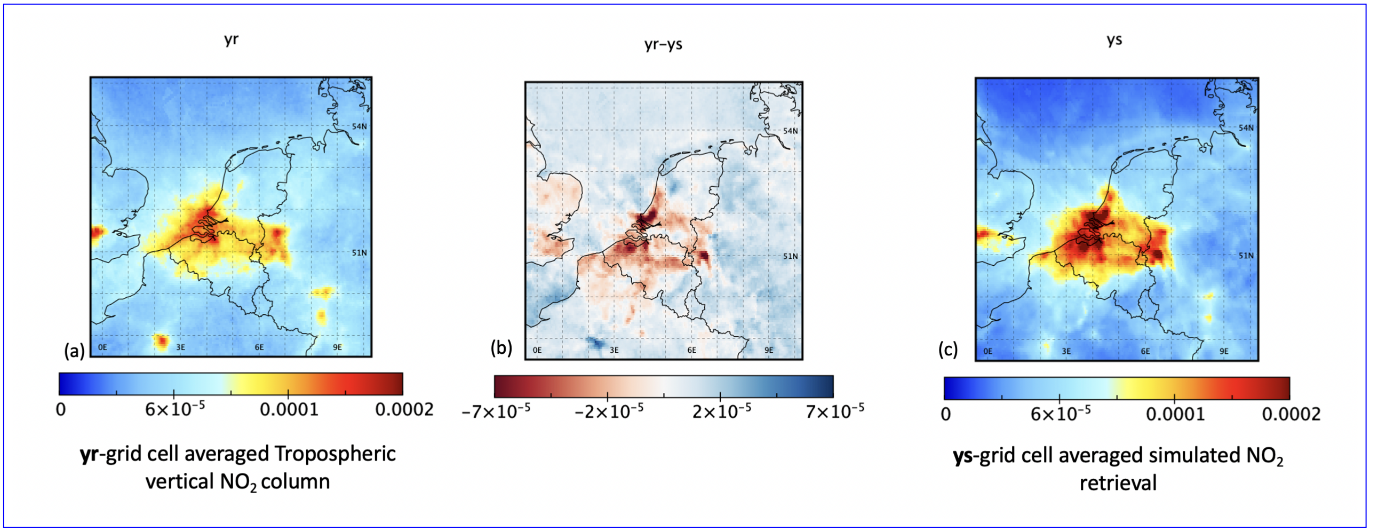

Figure 11. Comparison between the LOTOS-EUROS simulated retrieval of the tropospheric column of NO_2 Ys and the TROPOMI average tropospheric column Yr. Base maps from (http://www.gadm.org/)

4 Discussion

In this study two meteorological models which have different features served as input for the LOTOS-EUROS chemical transport model. The hydrostatic nature of a meteorological model as ECMWF refers to the assumption that the atmosphere is in a state of hydrostatic equilibrium, meaning that the vertical pressure gradient balances the gravitational force. In this configuration, the atmospheric equations used by the model do not include the effects of non-hydrostatic processes, such as wind, turbulence, and gravity waves. In contrast, a non-hydrostatic meteorological model allows for including non-hydrostatic processes in the atmospheric equations. This can provide a more accurate representation of the mixing dynamics of the atmosphere, especially in regions where these processes are significant, such as near the coast, over forests, and in urban areas.

The choice of a hydrostatic or non-hydrostatic meteorological configuration can significantly impact the performance of a chemical transport model. A hydrostatic configuration may be sufficient in some cases, but a non-hydrostatic configuration may be necessary to better represent the transport of pollutants in the atmosphere accurately. Overall, it is essential to carefully consider the meteorological model's capabilities and the study region's specific characteristics when choosing a hydrostatic or non-hydrostatic configuration for a chemical transport model. This can ensure that the model can accurately represent the transport ~~and impact of pollutants on~~ of pollutants and quantify air quality.

~~The~~ In this study, the vertical velocity fields in ~~the~~ LOTOS-EUROS ~~model~~ are calculated using the convergence and divergence of the horizontal winds from the meteorological model. This allows the model to simulate the effects of vertical motion in the atmosphere on pollutants' transport and chemical reactions. The availability of vertical meteorological fields can impact the accuracy and reliability of the LOTOS-EUROS model's predictions.If vertical wind data is unavailable or is of low quality, the model the transport of pollutants.Uncertainties in the vertical transport and mixing cause that a CTM may not accurately represent the vertical motion of pollutants in the atmosphere. This can lead to significant errors in the model's predictions of the distribution and impact of pollutants on air quality. Other models, such as CHIMERE, recently evaluated the vertical a new vertical advection mechanism to improve also the vertical transport (Menut et al., 2021). To improve the performance of the LOTOS-EUROS model, it is crucial to ensure that high-quality vertical wind data is available from the meteorological model. This can provide more accurate and realistic representations of the vertical motion of pollutants in the atmosphere and improve the accuracy of the model's predictions. and a new vertical advection scheme that strongly reduces excess vertical diffusion (Menut et al., 2021).

Using high-resolution high-spatial resolution meteorology in a chemical transport model CTM like LOTOS EUROS can improve the accuracy and reliability of the model 's predictionssimulations. High-resolution meteorological data provides more detailed information about the atmosphere's wind, temperature, pressure, and humidity conditions, which can be used to simulate the movement of pollutants and trace gases more accurately. In particular, high-resolution meteorology can provide more accurate representations of the effects of small-scale atmospheric processes, such as turbulence and convection, on pollutant transport and chemical reactions. This can improve the model's ability to simulate the distribution and impact of pollutants on air quality and can provide more detailed and helpful information for air quality forecastingand environmental management.

The input meteorological information is part of the CTM model error, and should be included in the uncertainty description when assimilating observations. The following step is the preparation for assimilation from the side of the data and the model perspectivethe assimilation experiments using satellite column measurements. Figure (11) shows the two products needed to assimilate. These results highlight the importance of carefully considering model configurations perform the assimilation, the difference between both provide the input to correct in any of the data assimilation techniques. It is important to carefully consider the model configuration and meteorological factors in atmospheric chemistry modeling and such as vertical mixing in a CTM for the potential benefits of satellite remote sensing data in improving the accuracy of the modeled modelled NO_2 concentrations. The comparison between the LOTOS-EUROS simulated retrieval of the tropospheric column of NO_2 Ys and the TROPOMI average tropospheric vertical column Yr product from the CSO preprocessing tool that is the input needed for the data assimilation stage is shown in Figure (11).

Panel (a) of 11 shows the LOTOS-EUROS simulated retrieval of the tropospheric column of NO_2 Ys, while panel (b) shows the TROPOMI average tropospheric vertical column Yr product. The comparison indicates that there are There might be significant differences between the two products, particularly in regions where there are simulated and observed products, in particular in regions with high NO_2 concentrations. These differences Underlying model uncertainties due to e.g. vertical mixing are important to consider in the data assimilation stage, as they can impact the accuracy of the assimilated data and, ultimately, the accuracy of the analysis modeled modelled NO_2 concentrations.

Comparison between the LOTOS-EUROS simulated retrieval of the tropospheric column of NO_2 Ys and the TROPOMI average tropospheric vertical column Yr product from the CSO preprocessing tool. Base maps from ()

Using high-resolution meteorology in chemical transport models like LOTOS EUROS can provide valuable insights into the transport and impact of pollutants on air quality and support decision-making and policy development to improve air quality and protect public health.

For data assimilation, it is essential to get estimations of the accuracy of the observations to construct the observation error covariance matrix; the error from the observations is used to build a diagonal matrix R because the error values at this stage are correlated only with the observed state in the already remapped grid. The inaccuracies in the TROPOMI observations result from the retrieval method's three stages, which are a previous step in pre-processing the satellite information from manipulating the crude light spectroscopy data to have the NO_2 vertical column density. The stages that add errors in this process are the quantification of slant columns, the separation of the stratospheric and tropospheric components of slant columns, and the tropospheric air mass factors multiplication (Van Geffen et al., 2020). The overall error is provided per pixel in the TROPOMI data product.

5 Conclusions

The HARMONIE (

This study explores the coupling of meteorological data from the HARMONIE (cy43) coupling with) model with the LOTOS-EUROS mimicking the ECMWF with LOTOS-EUROS technically works, showing comparable results in meteorology Chemical Transport Model (CTM) to simulate NO_2 concentrations, comparing these results with simulations that utilize ECMWF meteorological data. The research seeks to evaluate the performance and accuracy of these different meteorological couplings in predicting NO_2 levels. A general comparison between the two setups reveals that both meteorological variables and NO_2 concentrations. Differences in the details can be perceived mostly simulations are comparable, indicating a level of technical consistency between the HARMONIE and ECMWF configurations.

Despite the overall comparability, notable differences emerge in the vertical column concentration, for which in concentrations of NO_2. Specifically, the HARMONIE configuration , highly values appear exhibits higher values in the upper layer of the atmosphere than in the ECMWF configuration, which was caused for the differences in the vertical diffusion coefficient. The HARMONIE atmospheric model stands out with its enhanced structure and distinct field shape compared to the ECMWF. However, it exhibits a discrepancy when simulating the compared to the ECMWF setup. This difference is at least partly attributed to the vertical diffusion coefficients and planetary boundary layer height, highlighting the sensitivity of NO_2 dispersion to model-specific meteorological parameters. Our analysis reveals that HARMONIE provides a more detailed structure for meteorological drivers than the coarser ECMWF fields. This granularity is particularly evident in the simulation of the boundary layer height, overestimating it compared to real-world observations. This disparity significantly affects which, along with the diffusion coefficient discrepancies, significantly impacts air pollutant concentrations , particularly in the upper atmosphere near the surface and their transport to the higher layers of the atmosphere.

The study underscores the importance of accurately representing the boundary layer height, as it plays a crucial role in the distribution and chemical transformation of pollutants. The higher simulated boundary layer height in HARMONIE allows

~~pollutants to be transported to~~ simulated by HARMONIE facilitates the transport of pollutants to higher altitudes, ~~leading to complex chemical reactions and~~ where they can undergo chemical reactions leading to the formation of secondary pollutants. This phenomenon ~~affects~~ has implications for regional air quality, climate, and the understanding of long-range pollutant transport. ~~Accurately representing the~~ Addressing the discrepancies in boundary layer height ~~is crucial for reliable air quality forecasts and assessing pollutant impacts. Resolving this issue~~ simulation requires further research~~and refinement of~~ , focusing on refining the model's parametrizations and processes related to boundary layer dynamics ~~, enabling improved simulations~~ to enhance the simulation of pollutant dispersion ~~in~~ across different atmospheric layers~~; so far, inconclusive concerning performance in the surface concentrations compared with ground stations~~. ~~The fields evaluated (meteorology and NO2 concentrations) are comparable, with no significant improvement in~~

The analysis also points to a slight improvement in surface ~~NO2 compared to observations at surface stations . There is potential to further develop LOTOS-EUROS at high spatial~~ NO_2 concentrations when compared with observations from ground stations in the HARMONIE configuration, though it emphasizes that these findings do not significantly enhance our understanding of surface NO_2 levels. In terms of the statistics, an slight improvement for the performance in the surface NO_2 concentrations compared with ground stations was observed with the high-spatial resolution meteorology. The study calls for further examination of vertical transport processes and additional validation efforts, particularly with NO_2 profile measurements from MAX-DOAS. Highlighting the computational advantages and the need for high-spatial resolution in the HARMONIE configuration~~because of the less work in nesting domains to simulate at least the resolution objective in this work properly (0.025 °). The next step in this work is to use both configurations,~~ , the research suggests further development of LOTOS-EUROS to leverage these benefits fully.

Looking ahead, the study proposes using both the ECMWF and HARMONIE ~~, in the~~ configurations in a data assimilation experiment ~~of~~ with TROPOMI NO2 ~~using LOTOS-EUROS to understand~~ data. This approach aims to understand better the impact of ~~this non-hydrostatic meteorology in the~~ uncertainties in the meteorology on the horizontal and vertical transport of contaminants, marking an essential step towards refining air quality models and improving our ability to predict and mitigate the effects of air pollution on the environment and public health.

Author contributions. Conceptualization, AYB and MvW; methodology, AYB; software; AYB and AS validation; analysis AYB and MvW and AS and HE formal resources MvW and HE and PS ; data curation AYB ; writing original draft preparation AYB ; writing review and editing MvW, AS ; visualization AYB; project administration MvW . All authors have read and agreed to the published version of the manuscript.

6 ~~Appendix~~

Transport plumes of NO_2 TROPOMI Tropospheric column observations compared with the CABAUW observations for wind direction and magnitude for 7 levels from 2 m to 200m. © OpenStreetMap contributors 2021. Distributed under the Open Data Commons Open Database License (ODbL) v1.0.

Transport plumes of NO_2 TROPOMI Tropospheric column observations compared with the CABAUW observations for wind direction and magnitude for 7 levels from 2 m to 200m from 2019-04-22 to 2019-04-27 in which a scenario of changing air mass direction drive the transport of contaminants. © OpenStreetMap contributors 2021. Distributed under the Open Data Commons Open Database License (ODbL) v1.0.

Competing interests. Authors declare that no competing interests are present

Code availability

The codes are available at the GitLab repository dataset used in this study was obtained from a public repository hosted on Zenodo, a widely-recognized open-access repository that facilitates research data sharing and collaboration. This particular dataset, accessible at https://doi.org/10.5281/zenodo.8431342, provides comprehensive data essential for our analysis. It was published in 2023 and support the findings of this research. To download the Cabauw data: https://dataplatform.knmi.nl/dataset/cesar-tower-

Map data copyrighted by OpenStreetMap contributors and available from https://www.openstreetmap.org

The NO_2 data was downloaded for the ground stations at different places in the Netherlands from www.luchtmeetnet.nl

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Appendix: Appendix

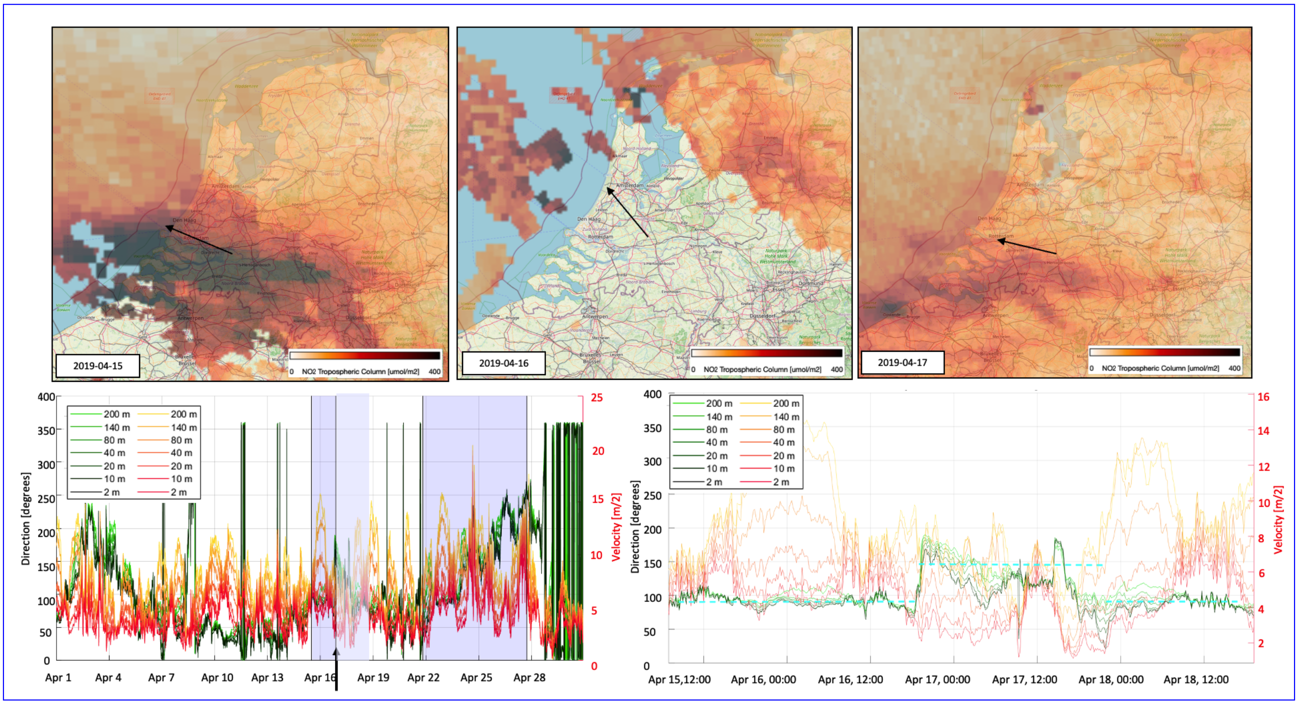

Figure 1. Transport plumes of NO_2 TROPOMI Tropospheric column observations compared with the CABAUW observations for wind direction and magnitude for 7 levels from 2 m to 200m 2019-04-15 to 2019-04-17. © OpenStreetMap contributors 2021. Distributed under the Open Data Commons Open Database License (ODbL) v1.0.

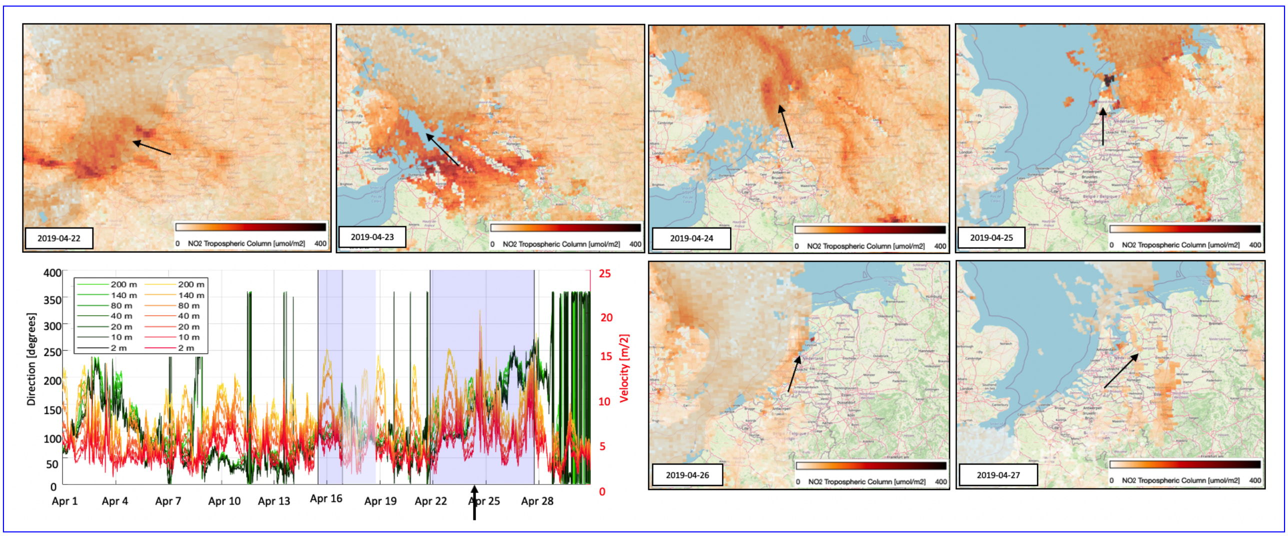

Figure 2. Transport plumes of NO_2 TROPOMI Tropospheric column observations compared with the CABAUW observations for wind direction and magnitude for 7 levels from 2 m to 200m from 2019-04-22 to 2019-04-27 in which a scenario of changing air mass direction drive the transport of contaminants. © OpenStreetMap contributors 2021. Distributed under the Open Data Commons Open Database License (ODbL) v1.0.

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