Influence of complex terrain and anthropogenic emissions on atmospheric CO\textsubscript{2} patterns—a high-resolution numerical analysis

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The accuracy of regional or mesoscale carbon budgeting by means of inverse modelling depends strongly on the ability of the atmospheric model to capture relevant atmospheric transport processes. In order to analyze the influence of terrain-induced flow dynamics and intense local anthropogenic emissions, we present high-resolution (forward) simulations of spatio-temporal CO\textsubscript{2} variations using a recent biosphere–atmosphere model. The selected region is characterized by complex terrain and both rural and densely populated areas. The results indicate that, in situations with weak synoptic forcing, the nocturnal near-surface CO\textsubscript{2} distribution is strongly affected by terrain-induced turbulent kinetic energy (TKE) above mountain ridges and by local convergent downslope winds. By increasing the grid spacing from \(\approx 1\) to \(\approx 3\) km, we show that, due to the smoothed model topography, the atmospheric flow causing the CO\textsubscript{2} heterogeneity cannot be resolved any more. Finally, we quantify the influence of intense anthropogenic CO\textsubscript{2} sources on atmospheric CO\textsubscript{2} concentrations. A significant anthropogenic signal can be identified around and downstream of industrial and urban areas, especially in the morning but also within a well-mixed planetary boundary layer in the daytime. The results provide valuable information for including non-background CO\textsubscript{2} observations in mesoscale inverse modelling studies using coarser resolutions than in this study.

Key Words: CO\textsubscript{2} transport; complex orography; slope winds; turbulent kinetic energy; fossil fuel emissions; mesoscale modelling; inverse modelling

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1. Introduction

Accurate knowledge of the terrestrial carbon budget is crucial for predicting the future CO\textsubscript{2} increase, which is needed to understand current and future climate changes. The only existing in situ observations of surface CO\textsubscript{2} fluxes are eddy-covariance measurements having a footprint of a few 100 m to some kilometres (Baldocchi et al., 2001). An upsampling to larger scales (‘bottom-up’ approach) is challenging, especially in heterogeneous regions (Xiao et al., 2011). A common method to quantify the CO\textsubscript{2} budget at larger scales is inverse modelling of observed CO\textsubscript{2} mixing ratios (‘top-down’ approach). This method determines terrestrial CO\textsubscript{2} fluxes from the variability of observed CO\textsubscript{2} mixing ratios by means of an atmospheric transport model. The accuracy of such flux estimates depends strongly on the quality of simulated atmospheric transport processes (e.g. advection, turbulence) and on the location of CO\textsubscript{2} point measurements. Ideally, the latter should not be influenced by mesoscale dynamics, which are not resolved by models using rather coarse grids.

Increasing interest is being directed to quantifying regional and mesoscale biogenic and anthropogenic CO\textsubscript{2} sources/sinks, which are needed for understanding terrestrial CO\textsubscript{2} budgets, important for regional climate modelling. Thus, current inverse modelling studies focus on the quantification of CO\textsubscript{2} fluxes at these scales (Gerbig et al., 2009; Göckede et al., 2010; Schuh et al., 2010; Broquet et al., 2011; Meesters et al., 2012, and others). For these studies, a much denser observation network is necessary (Lauvaux et al., 2012) than for global inversions to reduce the large number of unknowns that have to be constrained. The inclusion of non-background CO\textsubscript{2} observations is then unavoidable. However, high-resolution modelling studies and observations showed that mesoscale circulations and local fluxes generate high mesoscale atmospheric CO\textsubscript{2} gradients (e.g. Dolman et al., 2006; Sarrat et al., 2007; Ahmadov et al., 2007). Therefore, an important research question is which horizontal grid spacing resolves these processes sufficiently to yield reliable flux estimates by inverse modelling of terrestrial CO\textsubscript{2} observations.

A major uncertainty of these inverse modelling studies is the representativeness of a precise CO\textsubscript{2} point measurement for the grid-cell average of a transport model. The mismatch, referred to as the representation error, can be large for terrestrial stations. Many coastal and continental CO\textsubscript{2} towers are regularly influenced by mesoscale circulations (van der Molen and Dolman, 2007;
important role of terrain-induced advective \( \text{CO}_2 \) transport in the
et al., 2012) estimated that coarse-resolution models tend to
Pérez-Landa (2012): Pérez-Landa et al. (2014) stated that coarse-resolution models tend to miss substantial parts of the \( \text{CO}_2 \) exchange caused by unresolved mesoscale circulation in complex terrain regions, explaining this mismatch. In the last decade, several studies identified remarkable terrain-induced mesoscale circulations having a significant impact on ecosystem carbon exchange (Sun and De Wekker, 2012): Pérez-Landa et al. (2007) indicated pronounced mesoscale \( \text{CO}_2 \) patterns in a coastal complex terrain region caused by mountain-valley circulations at night combined with a sea breeze during the daytime. Sun et al. (2007) identified the importance of terrain-induced advective \( \text{CO}_2 \) transport in the front range of the Rocky Mountains, especially at night. Even relatively modest topography height differences can generate horizontal \( \text{CO}_2 \) gradients of the order of 30 ppmv (van den Molen and Dolman, 2007). Oney et al. (2015) found that the suitability of observations for regional carbon budgeting in complex terrain depends on the ability of the model to capture the atmospheric dynamics. They showed limits in steep topography, even using a high-resolution model. Thus, in inverse modelling studies, observations from mountain sites have been usually excluded, although they provide a larger scale representativeness than tower measurements over flat terrain (Pillai et al., 2011). High-resolution numerical simulations (\( \sim 1 \) km) are essential to investigate the influence of terrain-induced turbulent patterns and dynamics on the near-surface \( \text{CO}_2 \) distribution and may help to include mountain stations in future inverse studies. This holds in particular for retrieving respiration from night-time observations, which have often been excluded in inversions, due to strong transport and representation errors (e.g., Meesters et al., 2012).

Moreover, the quantification of the influence of anthropogenic \( \text{CO}_2 \) emissions on atmospheric \( \text{CO}_2 \) mixing ratios is an important task when using non-background \( \text{CO}_2 \) observations in an atmospheric inverse system. Especially in urbanized regions (e.g., Central Europe), where anthropogenic and biogenic fluxes overlap, a separation of both signals on \( \text{CO}_2 \) mixing ratios is important for regional \( \text{CO}_2 \) flux estimates (Lac et al., 2013). The relevance of fossil fuel emissions in populated areas was pointed out in past studies (e.g., Pérez-Landa et al., 2007; Ter Maat et al., 2010) using a rather coarse dataset (\( 1^{\times}1^{\circ} \)), disaggregated to urban grid cells only (i.e., disregarding rural \( \text{CO}_2 \) emissions). Ter Maat et al. (2010) stated that their simulations would be improved if more realistic high-resolution data and temporal downscaling were available. Other studies used emission data with 10 km resolution (Ahmadov et al., 2007; Tolk et al., 2009; Sarrat et al., 2009), but Lac et al. (2013) found that this is still too coarse for urban regions with highly heterogeneous fossil fuel emissions. The present study utilizes high-resolution disaggregated anthropogenic \( \text{CO}_2 \) emissions with a detailed temporal downscaling. \( \text{CO}_2 \) emissions from road networks and settlements in rural regions are considered explicitly. Uebel et al. (2017) indicated heterogeneous mesoscale \( \text{CO}_2 \) patterns in a region characterized by complex terrain and urban areas. The objectives of this subsequent study are, at first, a detailed analysis of the thermodynamic processes responsible for strong near-surface \( \text{CO}_2 \) gradients in mountainous regions. We focus on night-time effects in situations with weak synoptic forcing, when inverse modelling of \( \text{CO}_2 \) is still challenging, possibly due to high transport errors (e.g., Tolk et al., 2011). Second, the influence of sensitivity model topography on simulated \( \text{CO}_2 \) patterns is investigated, to assess which horizontal grid spacing is necessary to resolve terrain-induced circulations at night as well as \( \text{CO}_2 \) mixing ratios within a convective planetary boundary layer (PBL) during daytime appropriately. Finally, by means of sensitivity studies, we quantify the anthropogenic signal originating from \( \text{CO}_2 \) emissions of urban areas and big power plants at different times of the day. We provide valuable information for present and future mesoscale \( \text{CO}_2 \) budgeting by inverse modelling. The results can be used to integrate non-background \( \text{CO}_2 \) towers (e.g., mountain stations) into transport models using coarser grids than in this study, as well as to estimate the representation error of observed \( \text{CO}_2 \) mixing ratios.

In section 2, the modelling system and domain, anthropogenic emissions and case studies are described. Section 3 analyzes the simulated \( \text{CO}_2 \) variability for both case studies. In section 3, terrain-induced dynamic flow patterns influencing nocturnal \( \text{CO}_2 \) variability are investigated, as well as the appropriate horizontal grid spacing to resolve these patterns. Section 5 quantifies the anthropogenic effects, followed by a discussion of the results and final conclusions in sections 6 and 7, respectively.

2. Modelling configuration and methods

2.1. Model system: TerrSysMP-\( \text{CO}_2 \)

For the numerical simulations we apply the model system TerrSysMP-\( \text{CO}_2 \), which is based on the Terrestrial Systems Modelling Platform (TerrSysMP: Shrestha et al., 2014), extended by a prognostic treatment of \( \text{CO}_2 \) dynamics in the atmosphere (Uebel et al., 2017). TerrSysMP-\( \text{CO}_2 \) consists of the non-hydrostatic limited-area numerical weather prediction (NWP) model developed by the Consortium for Small-scale Modelling (COSMO, version 4.21: Baldauf et al., 2011) coupled to the Community Land Model (CLM, version 3.5: Dai et al., 2003; Oleson et al., 2010) via the external coupler OASIS3 (Valcke, 2013). This coupler allows for use of different grid spacings (section 2.2) and model time steps appropriate for COSMO and CLM.

Photosynthetic \( \text{CO}_2 \) uptake is calculated by CLM, which couples a biochemical photosynthesis model with a plant physiological stomatal conductance model similar to Collatz et al. (1991). Due to the two-way coupling of \( \text{CO}_2 \) between COSMO and CLM, in TerrSysMP-\( \text{CO}_2 \) the stomatal control of leaves is influenced by the prognostic near-surface distributions of air pressure, humidity and \( \text{CO}_2 \), i.e., in addition to the atmospheric humidity, the spatio-temporal varying \( \text{CO}_2 \) content influences photosynthesis and transpiration. The upscaling from leaf to canopy level is performed by the canopy integration scheme of Thornton and Zimmermann (2007), which is based on a "two-big-leaf" approach distinguishing between shaded and sunlit leaves.

In the parametrization of soil respiration, heterotrophic and autotrophic respiration are treated separately. Heterotrophic respiration is simulated by the carbon turnover model Roth-C26.3 (Coleman and Jenkinson, 2005) calculating the decomposition of organic plant material in the mineral soil using a carbon-pool concept. This approach explicitly considers the influence of the actual soil temperature and soil moisture distribution on microbial activity. The initialization of the carbon pools depends on land use and is based on organic carbon profiles measured at several locations within the model domain (see Uebel et al., 2017). By using this method, a long spin-up run for the carbon-pool initialization is not necessary. In forests, additionally the decomposition within the forest floor and of above-ground litter is calculated by exponential decay equations with characteristic

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decomposition rates, again depending on temperature and moisture availability. Finally, autotrophic respiration is derived from mean photosynthesis and leaf respiration rates, considering the actual moisture content in the root zone. The sum of respiration and photosynthesis forms the net ecosystem exchange (NEE), i.e. the biogenic CO₂ source/sink that influences the atmospheric CO₂ content (see below). For a more detailed description of the process-based parametrizations of TerrSysMP-CO₂, the reader is referred to Uebel (2016) and Uebel et al. (2017).

In order to simulate the atmospheric transport of CO₂ mixing ratios and the CO₂ exchange between the atmosphere and the biosphere, CO₂ was included as a new prognostic variable in TerrSysMP-CO₂ (Uebel, 2016). For that, we implemented CO₂ mixing ratios into COSMO by means of passive fluid tracers. In this context, ‘passive’ means that CO₂ cannot be changed within the atmosphere by chemical reactions or transitions of the state of aggregation and that feedbacks on radiative transfer are neglected. The prognostic equation of the mass-specific CO₂ content \( (q_{\text{CO₂}}) \) may be written formally as

\[
\frac{\partial q_{\text{CO₂}}}{\partial t} = -\mathbf{v} \cdot \nabla q_{\text{CO₂}} + \left( \frac{\partial q_{\text{CO₂}}}{\partial t} \right)_{\text{turb}} + \left( \frac{\partial q_{\text{CO₂}}}{\partial t} \right)_{\text{conv}} + \left( \frac{\partial q_{\text{CO₂}}}{\partial t} \right)_{\text{biogenic}} + \left( \frac{\partial q_{\text{CO₂}}}{\partial t} \right)_{\text{anthro}}.
\]

The first term on the right-hand side describes the advection of CO₂ with the (grid-scale) atmospheric wind field. The second and third terms denote local changes of CO₂ by subgrid-scale vertical turbulent mixing and convective transport. In the high-resolution simulations, we assume that deep cumulus convection is resolved by the model, whereas shallow convection is still parametrized (see Baldauf et al., 2011). The fourth term represents the local change by biogenic CO₂ sources and sinks (i.e. NEE) as calculated by CLM (see above) and the last term describes the influence of anthropogenic emissions (section 2.3). Due to the link between atmospheric CO₂ content, photosynthesis and transpiration (via the stomatal resistance), atmospheric CO₂ becomes active because the water and heat transfer between the biosphere and the atmosphere is directly influenced by CO₂. Nevertheless, the passive behaviour within the atmosphere enables us to include several prognostic CO₂ tracers in the COSMO model. This is a convenient method to analyze the effects of different CO₂ sources/sinks on the spatio-temporal CO₂ distribution separately (see sections 2.4 and 5.2) and was successfully applied, e.g. in Smallman et al. (2013) and Tolk et al. (2009).

2.2. Model domain characteristics and nesting

We performed high-resolution characterizations of spatio-temporal CO₂ variations with TerrSysMP-CO₂. As in Uebel et al. (2017), the atmospheric model (COSMO) uses a horizontal grid spacing of \( \Delta x = 0.01° \) (\( \approx 1.1 \text{ km} \), rotated spherical grid). A stretched vertical grid (50 levels) allows for a rather high vertical resolution within the PBL, with the lowermost model layer having a thickness of 20 m. With a horizontal grid size of \( 0.005° \times 0.00775° \) (\( \approx 500 \text{ m}, \text{regular geographical coordinates, lat/lon} \)), the spatial resolution of the vegetation and soil model (CLM) is even finer, to represent the heterogeneous land surface with the necessary detail. Hydrological processes in the mineral soil are calculated by CLM with a stretched vertical grid (10 levels).

Figure 1 shows the model domain, consisting of the western part of Germany (DE, Deutschland) as well as parts of the Netherlands (NL), Belgium (BE) and Luxemburg (LUX). This region was selected because it is characterized by complex and flat terrain including densely populated regions (red shaded areas). Thus, it is an appropriate region to investigate both the effects of natural heterogeneities and the influence of anthropogenic emissions on atmospheric CO₂ patterns. The Eifel region (\( \approx 300–700 \text{ m above sea level (a.s.l.)} \)) is located in the central and southern part of the domain, characterized by several mountain ridges separated by narrow valleys (e.g. Ahr, Rur, Urft). Further hilly terrain, Bergisches Land and Hunsrück, is found east of the rivers Rhine ( Rhein) and Moselle (Mosel), respectively. In contrast, the northern and northwestern part of the domain is rather flat.

The land cover is described by plant functional types (PFTs) based on Moderate Resolution Imaging Spectroradiometer (MODIS) land cover data (Shrestha et al., 2014). The flat terrain is dominated by agriculture and, especially along the rivers Rhine and Maas, urban areas. The low mountain ranges and the Moselle valley are mainly covered by broad-leaf forest and, in particular above 500 m a.s.l., needle-leaf forest. Deviating from the default plant physiological parameters of CLM, for agriculture the cereal parameters of Sulis et al. (2015) are used, which are calibrated to fertilized winter wheat in Central Europe. For the soil classification, the FAO–UNESCO (1975) Soil Map of the World is used. Most of the domain is dominated by loamy soils.

Initial conditions (ICs) and lateral boundary conditions (LBCs) for atmospheric state variables and for CO₂ mixing ratios are provided by TerrSysMP-CO₂ forecasts for a domain of about 1100 x 1400 km² surrounding the inner domain. This domain size was selected to ensure that during the forecast the PBL of the inner domain is not influenced by LBC effects of the outer domain. For the nesting simulations, a grid spacing of \( \Delta x = 0.025° \) (\( \approx 2.8 \text{ km} \)) was used for COSMO and CLM, to be driven by hourly COSMO-EU model analysis data (\( \Delta x = 0.0625°, \approx 7 \text{ km} \)). The nesting runs start with a lead time of 24 h to the high-resolution runs, using a homogeneous background CO₂ mixing ratio as IC and LBCs. This allows us to initialize the high-resolution simulations (inner domain) with a heterogeneous three-dimensional CO₂ distribution according to the predominant dynamical situation. Additional information on initialization and nesting of TerrSysMP-CO₂ can be found in Uebel et al. (2017).

2.3. Anthropogenic emissions

Especially along the river Rhine, the population density is very high, including the metropolises Cologne (Köl n), Dusseldorf (D), Aachen (AC) and Maastricht (MA). The areas ‘A1’ and ‘A2’ are analyzed in detail in section 4.
anthropogenic emissions are an important CO₂ source. In the simulations, these emissions are represented by a preliminary version of the TNO-CAMS CO₂ inventory, provided by the Netherlands Organisation for Applied Scientific Research, TNO (H. Denier van der Gon, October 2013; personal communication). It is consistent with an updated version of the TNO-MACC-II emission inventory (based on Kuenen et al., 2014), a gridded dataset on the basis of yearly official national air pollution and greenhouse gas reports (2000–2011). Considering the different origins of these emissions, Source Nomenclature for Air Pollution (SNAP) sectors are introduced. Relevant CO₂ sources stem from power generation (SNAP 1), non-industrial combustion (SNAP 2), industrial combustion (SNAP 3), road transport (SNAP 7), other mobile sources (SNAP 8) and waste treatment (SNAP 9).

To obtain an appropriate horizontal resolution for Germany and large parts of its bordering countries, the data were disaggregated to a resolution of 1.0 km by the Rheinisches Institut für Umweltforschung an der Universität zu Köln (P. Franke, E. Bem and J. Klimpt, October 2013; personal communication). High-resolution proxy data were applied, containing detailed information on local CO₂ emitters (e.g., road maps, geographical coordinates of industrial plants). Figure 2 depicts the annual sum of emissions, interpolated to the COSMO grid of the inner domain. The high fossil fuel emissions in urbanized areas are apparent, stemming mainly from urban traffic, industrial and residential combustion. In contrast, in parts of the sparsely populated Eifel, anthropogenic emissions are about two orders of magnitude lower than in the cities. The major contribution to fossil fuel emissions results from power generation, e.g. \( \approx 1/3 \) of all anthropogenic CO₂ emissions in North Rhine–Westphalia (NRW) are released by the three biggest lignite-fired power plants (‘+’ in Figure 2).

Anthropogenic emissions show an intrinsic temporal variability on different time-scales. Hence, for each SNAP sector we apply the emission time factors used in the LOTOS-EUROS chemistry-transport model (Schaap et al., 2005) describing the dependence on different months, day of the week and time of day. Figure 3 depicts seasonal and diurnal variations of selected SNAP sectors. Whereas road transport (SNAP 7) is rather constant throughout the year, power generation (SNAP 1) shows slightly lower and non-industrial combustion (SNAP 2) considerably lower emission in summer than in winter, due to domestic heating in the winter season (Figure 3(a)). Emissions of road transport have a pronounced diurnal variation (Figure 3(b)), clearly indicating rush hour effects around 0800 and 1800 CEST (0600 and 1600 UTC). Similarly, non-industrial combustion shows a distinct diurnal variation, whereas power generation is only slightly lower at night. Applying the emission time factors for each SNAP sector, hourly anthropogenic emissions can be calculated for the required simulation period, being added as CO₂ source term (\( \partial q_{\text{CO}_2}/\partial t \)anthro (Eq. (1)) to the simulated CO₂ mixing ratio. The elevated release and thermal plume rise of industrial and power plants is taken into account by spreading these emissions vertically between \( \approx 100 \) and 500 m above ground level (a.g.l.).

The high spatial resolution of these data, combined with the varying emission time factors for different SNAP sectors, provides detailed spatio-temporal information on fossil fuel emissions in Central Europe. This allows us to compare the influence of anthropogenic emissions on the CO₂ distribution in urbanized regions with rural regions (section 5).

### 2.4. Case studies and numerical simulations

For the numerical analyses, we selected two case studies. The first case, 24 July 2012, was an almost cloudless day. The temperatures ranged from 9–15 °C in the morning to 21–28 °C in the afternoon. At night, except for the mountain ridges of Eifel and Hunsrück, the easterly wind was only light to gentle (see wind arrows in Figure 4(a)). In narrow valleys, almost calm conditions are simulated, leading to a shallow nocturnal PBL with low near-surface temperatures and a strong temperature inversion. With the evolution of a convective PBL in the daytime, the wind intensified slightly and turned to more northern directions (Figure 4(c)).

For this clear-sky day (CS), we performed several numerical simulations. The main model run (CS2407) simulates ‘realistic’ conditions including biogenic CO₂ fluxes and anthropogenic emissions. A second run (CS2407-bio) only considers biogenic CO₂ fluxes. In both the nesting domain and the inner domain, anthropogenic emissions are omitted. By comparing

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**Figure 2.** Annual sum of anthropogenic emissions (kg m⁻²). The crosses mark the three biggest power plants in NRW (adopted from Uebel et al., 2017, figure 2).

**Figure 3.** LOTOS-EUROS emission time factors for selected SNAP sectors: (a) monthly and (b) hourly factors (Central European Summer Time, CEST).

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CS2407 and CS2407-bio, the influence of anthropogenic emissions—originating in the inner domain (Figure 2) and in the nesting domain—on atmospheric CO₂ mixing ratios can be studied (section 5.1). Moreover, in order to separate the influence of orography and heterogeneous vegetation on atmospheric CO₂ patterns, we performed an idealized simulation with a homogeneous forest vegetation (CS2407-forest, section 3). Finally, a simulation with a grid spacing of Δx ≈ 2.8 km (CS2407-low) demonstrates the importance of fine grid resolutions to represent effects of complex terrain. All simulations (inner domain) start at 1800 UTC (23 July). The background CO₂ mixing ratio used for the IC and LBCs of the nesting run is 388 ppmv (atmospheric mean, observed in 100 m height at a meteorological tower within the domain).

In the second case, 3 September 2012, at night a band of low- and mid-level clouds reached the northwestern part of the domain and moved southeastwards. In the morning, it ranged from Luxemburg and eastern Belgium to Bergisches Land. The southeastern part of the domain was cloudless. In the afternoon, the simulated clouds were mostly concentrated in mountainous regions, whereas satellite images show clouds shifted to the northern part of the domain. Only a light breeze occurred at night, which weakened further, leading to an almost calm situation in the morning (Figures 5(a) and (b)). During daytime, a light to gentle northern wind was observed and the temperatures were moderate (16–22 °C).

Similar to the first case, we performed a ‘realistic’ numerical simulation for this cloudy (CL) day (CL0309). In addition to the active CO₂ tracer being coupled to CLM (‘real’), we used the inert (i.e. passive) property of tracers in COSMO to include a second CO₂ tracer. This tracer (‘biogenic’) receives only local tendencies from biogenic CO₂ fluxes, i.e. \((∂q_{CO₂}/∂t)_{biogenic}\) in Eq. (1), whereas anthropogenic emissions are excluded, i.e. \((∂q_{CO₂}/∂t)_{anthro} = 0\). The ICs and LBCs for the ‘real’ and ‘biogenic’ CO₂ tracers are identical, considering anthropogenic effects in the nesting simulation. In contrast to the comparison of CS2407 and CS2407-bio, the differences of the tracers ‘real’ and ‘biogenic’ indicate the influence of emitted CO₂ within the inner domain and during the simulation period only (section 5.2). For reference, analogously to the first case study, a second simulation, omitting anthropogenic emissions completely (CL0309-bio), was performed, but in this article the results of this simulation are not presented in detail. The background CO₂ mixing ratio used for the IC and LBCs of the nesting run is 390 ppmv.

### 3. Diurnal variability of near-surface CO₂ mixing ratios

The mesoscale application of TerrSysMP-CO₂ in Uebel et al. (2017) pointed out that, additionally to the synoptic flow, in regions with hilly terrain the CO₂ distribution within the PBL is strongly influenced by terrain-induced local circulations. In order to analyze the thermo-dynamical processes explaining this behaviour in detail, at first the spatio-temporal variability of near-surface CO₂ patterns is described for the two selected case studies.
night. Especially in valleys east of the Rhine (e.g. Sieg, Agger), where negligible winds persist for several hours until 0300 UTC (BLH < 50 m), CO₂ contents increase to 430–455 ppmv. Along Moselle, Ahr and in the northwestern part of the domain, CO₂ concentrations of 420–445 ppmv occur. In agreement with CS2407, above mountain ridges the CO₂ contents remain low (395–410 ppmv).

At 0600 UTC, the CO₂ mixing ratios differ strongly from CS2407. Significantly higher CO₂ mixing ratios are simulated for CL0309, reaching widespread 425–450 ppmv and 440–480 ppmv along and east of the Rhine (Figure 5(b)). Due to negligible winds (0–2 m s⁻¹ in most regions), anthropogenic emissions originating from urban areas can produce a very prominent increase of near-surface CO₂ mixing ratios in these regions. In contrast to CS2407, even in rural regions very high CO₂ concentrations are simulated. This can be explained by the occurrence of optically thick clouds, strongly reducing global radiation, in combination with a weak atmospheric flow. On the one hand, the onset of CO₂ uptake by photosynthesis is thus inhibited and on the other hand the formation of a convective PBL in the early morning is suppressed or delayed by clouds. Additionally, the later sunrise on 3 September compared with midsummer contributes to this delay. Only in the cloudless southeastern part of the domain, above the Hohe Eifel and Hunsrück, have the atmospheric CO₂ concentrations already decreased, leading to distinctly lower CO₂ contents in the following hours compared with the cloudy regions (not shown).

In the following hours, the influence of clouds on CO₂ content in the PBL disappears. In the afternoon, increased CO₂ mixing ratios are simulated downstream of fossil fuel emissions, whereas in rural areas CO₂ mixing ratios of 385–395 ppmv occur (Figure 5(c)). Thus, for this day a significant atmospheric CO₂ sink is not recognizable (see section 5.2 and Table 1). As in CS2407, the CO₂ mixing ratios within the well-mixed PBL are rather homogeneous.

4. Influence of complex terrain on nocturnal CO₂ patterns

In former studies, lower CO₂ contents above mountain ridges than in valleys and in flat terrain at night were often explained by thermally driven mountain–valley circulations (‘drainage flow’) and buoyancy-driven (katabatic) downslope winds in weak synoptic forcing conditions (e.g. Pérez-Landa et al., 2007; Pillai et al., 2011). However, Eq. (1) shows that a locally convergent wind field (e.g. nocturnal downslope winds) alone cannot create strong near-surface CO₂ gradients if the original CO₂ field in the afternoon is rather homogeneous within the PBL, i.e. negligible CO₂ gradients (Vg CO₂ ≈ 0). Spatially different turbulent mixing (qCO₂/∂t turb or heterogeneities in surface CO₂ sources (qCO₂/∂t) source with source = [biogenic, anthropogenic] are necessary to initiate near-surface (i.e. terrain-following) CO₂ gradients. We apply the methodology of van der Molen and Dolman (2007), analyzing the correlation of CO₂ mixing ratios and turbulent kinetic energy (TKE) in complex terrain, for two selected areas (‘A1’ and ‘A2’, see Figure 1), using a considerably finer grid spacing (Δx ≈ 1 km) than in their study (Δx = 27 km).

Figure 6 depicts the nocturnal conditions in the Ahr and Wied region (‘A1’), which stand out with high near-surface CO₂ gradients in CS2407 (cf. Figure 4(a)). After sunset, in the valleys (blue marked regions), thermal cooling induces the formation of a shallow and stable nocturnal PBL, causing a buoyancy-driven consumption of TKE at the surface. In contrast, between 2000 and 2300 UTC above mountain ridges (red marked regions), TKE is produced by wind shear. At 2230 UTC, along the mountain ridges Hohe Eifel (region 1) and Siebengebirge (region 3), as well as at a hilltop north of the Ahr (region 2), significant TKE is simulated in the lower atmosphere (Figure 6(b)). Areas with high TKE values are well correlated with low near-surface CO₂ contents (Figure 6(c)). Hence, compared with the conditions in valleys and in flat terrain, above mountain tops intensified vertical mixing inhibits the accumulation of CO₂ being respired at the surface.

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In contrast, in the valleys of Ahr and Wied (TKE minimum in regions 4 and 5), high CO$_2$ mixing ratios are simulated. Due to almost calm conditions, locally respired CO$_2$ can accumulate near the surface within a very stable and shallow nocturnal boundary layer.

A vertical cross-section along the line in Figure 6 shows that the TKE maxima at the top of the mountain ridges are restricted to the lowest few decametres (Figure 7(a)). Here, the wind speed at the surface level is significant (4–6 m s$^{-1}$) and further increases by 4–6 m s$^{-1}$ in the lowermost 70 m a.g.l. Indicating wind shear as a source for TKE production in the lower atmosphere. In the flat terrain and in valleys, the wind speed and its variations with height are low ($\approx 1$ m s$^{-1}$). Enhanced wind speeds above hilltops and decreasing wind speeds on the lee-side slopes are common in moderately stable conditions (see e.g. van der Molen and Dolman, 2007). The mountain tops can be considered to be in the transition zone from the nocturnal boundary layer to the residual layer above. The CO$_2$ maximum at the bottom of the Ahr valley and on the lower part of the northern mountain slope (Figure 7(b): 50.44–50.51°N) coincides well with a lack of TKE. After 0000 UTC, TKE reduces continuously due to decreasing wind shear and is then even more concentrated on the mountain tops. However, in the early morning TKE still exists. Convergences of downslope flows intensify CO$_2$ gradients further in complex terrain, which is shown in more detail for area ‘A2’. A similar correlation between wind shear, TKE and near-surface CO$_2$ contents occurs in parts of the Moselle valley (not shown).

Figure 8 depicts the situation in the northern Eifel (Rur–Urf–Olef region, area ‘A2’). Similarly to ‘A1’, after sunset significant TKE is produced (Figure 8(b)) above the Hohes Venn (region 1), but also above other elevations (regions 2 and 3). Again, in these regions rather low CO$_2$ mixing ratios are simulated (Figure 8(c)). In the valleys of Urf and Olef (region 4) and in the northern part of the Rur valley (region 5), TKE was consumed and regions with low TKE correlate with high near-surface CO$_2$ contents. A second TKE minimum (region 6) is simulated in the southern Rur valley, but in its northern part the low TKE is shifted slightly towards the northern mountain slope. In its southern part, CO$_2$ accumulates at the bottom of the Rur valley, due to negligible winds. However, further northwards intensified southeasterly winds occur along the slope and at the top of the mountain ridge. Thus, a stronger advective removal of locally respired CO$_2$ causes significantly reduced CO$_2$ mixing ratios, whereas CO$_2$ accumulation is restricted to the Rur valley floor only. This wind pattern persists in the second half of the night (Figure 8(d), region A) and thus in this region the CO$_2$ concentrations remain low (Figure 8(f)).

After 2300 UTC, TKE decreases and at 0200 UTC high TKE is only simulated above the Hohes Venn and some hilltops, whereas in most regions TKE is rather low (Figure 8(e)). Hence, the increasing terrain-following CO$_2$ gradients between 2230 and 0200 UTC can only be partly explained by the TKE distribution. In addition, the wind field 10 m a.g.l. contains strong terrain-induced convergences east of the Olef valley and southeast of the Rur valley (regions B in Figure 8(d)). A light to gentle southeasterly downslope wind (3–4.5 m s$^{-1}$) blows at the edges and along the lee sides slopes of these mountain ridges, whereas calm conditions (< 1 m s$^{-1}$) occur in the lower parts of the valleys. Thus, air with low CO$_2$ mixing ratios is advected from the mountain tops to the lee side, causing low CO$_2$ contents in the upper part of the mountain slopes (Figure 8(f)). Further down and at the floors of the Rur and Olef valleys, locally respired CO$_2$ is not removed by advective transport and vertical mixing (TKE) is suppressed in the stable PBL, i.e. CO$_2$ can accumulate near the surface. In other words, the local tendency of the CO$_2$ content ($\partial q_{CO_2}/\partial t$) at valley floors is stronger than at hilltops and in the upper part of the lee side slopes. Here, advective ($\mathbf{v} \cdot \nabla q_{CO_2}$) and turbulent transport (($\partial q_{CO_2}/\partial t$)$_{turb}$) inhibit a strong local CO$_2$ tendency, whereas both terms of Eq. (1) can be neglected at the valley floors. Thus, during the night the near-surface CO$_2$ gradient increases continuously. In summary, stronger vertical mixing at hilltops, in particular in the first half of the night, initially generates terrain-following CO$_2$ gradients, which are intensified further by terrain-induced convergences of the wind field.

In order to confirm that the near-surface CO$_2$ gradients result from the complex topography and not from land use heterogeneity, we performed a simulation assuming that the entire domain (except urban areas) is covered by broad-leaf forest (CS2407-forest). This is the dominant vegetation type in the hilly terrain of ‘A1’ and ‘A2’. Apart from some higher absolute values of CO$_2$ mixing ratios due to slightly higher area-averaged respiration rates, the general horizontal CO$_2$ patterns look rather similar to the near-surface CO$_2$ distribution using the true land use (Figure 9). Hence, heterogeneities in the surface CO$_2$ flux cannot explain the strong CO$_2$ gradients, which in CS2407-forest are similar to or (in some regions) even slightly stronger than those in CS2407. In other words, terrain-induced local dynamics (turbulence, slope winds) generate the simulated near-surface CO$_2$ heterogeneity. Higher CO$_2$ mixing ratios in the flat terrain (northwestern part of ‘A1’, northeastern part of ‘A2’) are caused by higher respiration rates of broad-leaf forests than of crops normally dominating these regions.

An important research question (e.g. for inverse modelling studies) is the minimum horizontal grid spacing necessary to resolve horizontal CO$_2$ gradients reasonably. Hence, in Figure 10 we compare the near-surface CO$_2$ distributions of CS2407 with a respective simulation with a horizontal grid spacing of $\Delta x \approx 2.8$ km (CS2407-low), as used for the nesting simulation.
At night (0200 UTC), the CO$_2$ patterns differ strongly in both simulations (Figure 10(a)). Whereas in valleys the CO$_2$ contents of CS2407 are much higher than in CS2407-low (e.g. Moselle basin, Wied valley, region 'A2'), lower CO$_2$ contents occur at mountain ridges using the fine grid spacing. Thus, over complex terrain the near-surface CO$_2$ gradients in CS2407-low are distinctly lower than in CS2407. In area 'A2', the strong CO$_2$ accumulation in the valleys of the Rur–Urft–Olef region (cf. Figure 8(f)) is not resolved by the coarser grid (Figure 10(c)). Slightly higher CO$_2$ mixing ratios along the valley floors are only adumbrated. This can be explained by a stronger smoothing of the model topography with increasing grid spacings. These results are in close accordance to a study around the Swiss Plateau, being characterized by similar complex terrain, using the COSMO model with 2 km grid spacing (Oney et al., 2015). They found that local atmospheric conditions and wind directions are reproduced poorly by the model for mountaintop observations. Both model elevations deviating from the truth and the smoothing of steep terrain slopes inhibit the formation of orographically induced local dynamics (e.g. slope winds) in the simulations of Oney et al.
Figure 9. Simulated CO2 mixing ratio (ppmv) at COSMO full-level 50 (≈ 10 m a.g.l.) with homogeneous land use (deciduous broad-leaf forest, CS2407-forest): (a) ‘A1’ on 23 July 2012 at 2230 UTC (as Figure 6(c)), (b) ‘A2’ on 23 July 2012 at 2230 UTC (as Figure 8(c)) and (c) ‘A2’ on 24 July 2012 at 0200 UTC (as Figure 8(f)).

Figure 10. Difference ‘CS2407 minus CS2407-low’ simulation of CO2 mixing ratios (ppmv) (lowermost COSMO level) at (a) 0200 UTC and (b) 1200 UTC. (c) Simulated CO2 mixing ratio (ppmv, shaded) and horizontal wind (m s⁻¹, arrows) of ‘A2’ (lowermost COSMO level) at 0200 UTC using Δx ≈ 2.8 km (CS2407-low).

and in our own simulations. In contrast to the high-resolution simulation, the synoptic wind field is only slightly modified by the topography (cf. Figures 8(f) and 10(c)). This points out that, for a grid spacing of several kilometres, the model topography is already too smooth to reproduce terrain-induced turbulent patterns and mesoscale circulations reasonably in a region with steep mountain slopes and narrow valleys, at least when synoptic forcing is weak and a stable nocturnal PBL evolves. In the flat terrain, the differences are weaker but significant as well (> 5 ppmv in some regions). At about 100 m a.g.l., the deviations are smaller but still apparent (not shown).

In the well-mixed PBL in the daytime (1200 UTC), the differences between CS2407 and CS2407-low are considerably smaller (Figure 10(b)). Significant deviations occur at the foot of Bergisches Land east of the Rhine, downstream of strong anthropogenic emissions (e.g. power plants) and in some small and randomly distributed areas. In rural regions, apart from the edge of mountain ranges influencing the regional-scale flow, the CO2 contents of both simulations are comparable for this case study. The better model orography of CS2407 in the Eifel has almost no effect on the CO2 distribution within the well-mixed PBL.

5. Influence of anthropogenic emissions

Fossil fuel emissions are an important contribution to the local and global CO2 budget. Thus, the influence of anthropogenic emissions on the spatio-temporal CO2 distribution was investigated. Table 1 lists the CO2 sources and sinks of CS2407 and CL0309, averaged over the entire model domain and over 24 h.

In clear-sky conditions in summer (CS2407), efficient CO2 uptake by photosynthesis (−11.1 μmol(CO2) m⁻² s⁻¹) cannot be compensated by biogenic (6.6 μmol(CO2) m⁻² s⁻¹) and anthropogenic (3.0 μmol(CO2) m⁻² s⁻¹) CO2 sources, causing a net loss of −1.5 μmol(CO2) m⁻² s⁻¹. For the cloudy case in early autumn (CL0309), the biosphere is still a weak sink (−1.5 μmol(CO2) m⁻² s⁻¹), but the strong anthropogenic emissions cause a total CO2 gain of 1.7 μmol(CO2) m⁻² s⁻¹. In this particular region, with 30.8% (CS2407) and 38.8% (CL0309) the amount of anthropogenic emissions to the total (biogenic + anthropogenic) CO2 source is exceptionally high. On the one hand, the high anthropogenic contribution can be explained by urbanised regions along the rivers Rhine and Maas (e.g. industry, road traffic). On the other hand, more than half of all fossil fuel emissions stem from power generation (SNAP 1). In 2012, three of the five lignite-fired power plants (Neurath, Niederaußem, Weisweiler), which have the highest CO2 emissions of all power plants in Europe, were located in this rather small area.

5.1. Case study: 24 July 2012

At first, the influence of anthropogenic CO2 sources, originating from emissions within the inner model domain and from advection into the considered area (e.g. emitted from the nearby Ruhr area), is investigated. For that, the near-surface CO2 mixing ratios of CS2407 and CS2407-bio (see section 2.4) are compared. Considering anthropogenic emissions in the nesting simulation of CS2407, in and around Cologne and Dusseldorf the initial CO2 content (2012/07/23 1800 UTC) is about 10–25 ppmv higher than...
in CS2407-bio (not shown). In the surrounding flat terrain the difference is 3–10 ppmv and in the Eifel the initial concentrations of CS2407 and CS2407-bio are comparable. During night-time, the differences remain at this level but the urban effects diminish (Figure 11(a)). Hence, the nocturnal CO₂ increase is caused mainly by leaf and soil respiration.

With rapidly increasing road traffic in the morning, the difference between CS2407 and CS2407-bio is much more pronounced (Figure 11(b)). Especially in the flat terrain and in the valleys east of the river Rhine, the CO₂ mixing ratios are 5–15 ppmv higher in CS2407 than in CS2407-bio. In addition to rush-hour effects, the stable stratification supports a strong anthropogenic influence. Moreover, the metropolitan areas Bonn–Cologne–Düsseldorf and Liège stand out with 15–30 ppmv (locally 50 ppmv) higher near-surface CO₂ contents than in CS2407-bio. However, without using an urban scheme in the current version of TerrSysMP-CO₂, urban effects on the atmospheric conditions (e.g. urban heat islands) are neglected, which may have an influence on the simulated BLH and thus on CO₂ mixing ratios in urban areas (Lac et al., 2013).

In the following hours, downstream of the big cities the near-surface CO₂ concentration remains elevated (3–10 ppmv) and a northeasterly wind advects high CO₂ concentrations to the northern Eifel (Figure 11(c)). Within the convective PBL, the tremendous emissions of the above-mentioned power plants are mixed to the surface and advected in a southwestern direction. In the rural Eifel region (southwestern part of the domain, see Figure 2), the difference between CS2407 and CS2407-bio is negligible. Here, the CO₂ mixing ratios within the well-mixed PBL are controlled by natural sources and sinks. The sharp gradient east of the Rhine results from a strong wind convergence (NW along the Rhine, E in Bergisches Land, cf. Figure 4(c)) at this time.

Figure 12 depicts selected vertical CO₂ profiles of CS2407 (solid) and CS2407-bio (dashed) at different times of the day. The grid cell ‘Cologne-East’ (Figure 12(a), △ in Figure 11) and its neighbouring grid cells are influenced by very intense anthropogenic CO₂ sources (e.g. freight depot, industry, much frequented motorway junctions). The other profiles are located apart from strong local emissions: ‘Jüllich’ (×), Figure 12(b) (flat terrain) and ‘Wied valley’ (♦), Figure 12(c) (valley floor).

All profiles show that the strong near-surface increase during night-time (e.g. ≈ 50 ppmv in the Wied valley) is caused mainly by biogenic CO₂ sources (0400 UTC). In contrast, at 0600 UTC, especially at Cologne-East, the simulated near-surface CO₂ concentration of 458 ppmv is caused by humans (cf. 409 ppmv in CS2407-bio). However, the anthropogenic signal is restricted to the nocturnal PBL (BLH ≈ 100 m). The rural profiles indicate only slightly increased CO₂ contents compared with CS2407-bio (Figures 12(b) and (c)). Before 0900 UTC, a shallow convective PBL has begun to evolve and the CO₂ mixing ratios at Cologne-East are still noticeably increased. Whereas here and at Jüllich a surplus of CO₂ occurs within the PBL, in the Wied valley the high photosynthesis rates of broad-leaf forest lead to a net CO₂ sink, i.e. lower concentrations than in the free troposphere. In the afternoon (1400 UTC), the Jüllich profile shows considerably higher CO₂ contents in CS2407 (390–397 ppmv) than in CS2407-bio (< 380 ppmv). This profile is influenced by advection of high CO₂ concentrations from the metropolitan region Cologne–Düsseldorf and is partly affected by the trails of the power plants Neurath and Niederaußem, as indicated by the bulge between 300 and 600 m a.g.l. (see also
1800 UTC). Without anthropogenic emissions, a CO\textsubscript{2} loss is simulated for all profiles (i.e. 5–10 ppmv lower CO\textsubscript{2} contents within the PBL than in the free troposphere). A reason for the relatively strong anthropogenic effect in this case study is a rather shallow PBL with a BLH of about 800 m at Jülich, 1000 m in the Rhine valley and 1100 m in the Wied valley. The BLH is a key variable for the influence of CO\textsubscript{2} fluxes on the atmospheric CO\textsubscript{2} content (e.g. Sarrat et al., 2007; Lac et al., 2013). In the evening, the beginning near-surface stabilization and anthropogenic sources can be seen at Cologne-East. In the Wied valley, the effect of anthropogenic CO\textsubscript{2} emissions is rather weak in the afternoon/evening, i.e. the CO\textsubscript{2} content within the PBL is controlled by biogenic CO\textsubscript{2} fluxes.

### 5.2. Case study: 3 September 2012

In a second sensitivity experiment, for 3 September 2012 (CL0309) only the anthropogenic signal originating from emitters within the model domain is investigated by comparing the mixing ratios of the ‘real’ and ‘biogenic’ CO\textsubscript{2} tracers (see section 2.4). Thus, in contrast to the sensitivity study described above, the differences in the CO\textsubscript{2} distributions are only affected by local anthropogenic sources over the period of the simulation, whereas the ICs and LBCs of both tracers are the same. Figure 13(a) indicates that, at 2000 UTC (i.e. only 2 h after initialization), human produced CO\textsubscript{2} causes a widespread increase of 1–5 ppmv in the flat terrain and, locally, an increase of 10–30 ppmv near urban and industrial regions. The anthropogenic signal at this time of day is distinctly stronger than in CS2407, because of the earlier sunset (i.e. earlier formation of a stable nocturnal PBL). In the sparsely populated Eifel, the difference between ‘real’ and ‘biogenic’ is negligible. At night, the general patterns are similar but the differences in urban areas decrease, because, compared with biogenic fluxes, in most regions anthropogenic CO\textsubscript{2} fluxes are small (< 0.5 \textmu mol(CO\textsubscript{2}) m\textsuperscript{-2} s\textsuperscript{-1}); see Figure 14(a). The major sources result from industrial combustion and power plants, which mainly emit above the nocturnal PBL (BLH ≤ 100 m in most regions).

During the morning rush hour (Figure 13(b)), the difference between ‘real’ and ‘biogenic’ increases to widespread differences of 5–20 ppmv in the flat terrain and in valleys. In and around big cities, the near-surface CO\textsubscript{2} mixing ratios of the ‘real’ CO\textsubscript{2} tracer are 20–50 ppmv higher than those of the ‘biogenic’ tracer, due mainly to urban traffic (Figure 3(b)). This clearly indicates a strong anthropogenic signal in the selected domain, supported by the meteorological situation, as described in section 3.2. Figure 14(b) shows that at 0600 UTC, except for the southwestern part of the domain, anthropogenic CO\textsubscript{2} fluxes exceed 1 \textmu mol(CO\textsubscript{2}) m\textsuperscript{-2} s\textsuperscript{-1} in rural areas, 3–10 \textmu mol(CO\textsubscript{2}) m\textsuperscript{-2} s\textsuperscript{-1} in grid cells including high frequent roads and 10–20 \textmu mol(CO\textsubscript{2}) m\textsuperscript{-2} s\textsuperscript{-1} in urban areas (> 100 \textmu mol(CO\textsubscript{2}) m\textsuperscript{-2} s\textsuperscript{-1} in industrial zones). Thus, fossil fuel emissions are the major CO\textsubscript{2} source in urban areas, but even in rural areas these emissions cannot be neglected. As expected, in the afternoon the differences between the ‘real’ and ‘biogenic’ tracer are weaker, due to a well-mixed and fully developed PBL. Advection of 1–5 ppmv higher CO\textsubscript{2} contents towards the Eifel can be seen, as well as trails of the power plants (Figure 13(c)).

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**Figure 13.** Difference of the CO\textsubscript{2} tracers ‘real’ minus ‘biogenic’ (ppmv) of CL0309 (lowest COSMO level) on (a) 2 September 2012 at 2000 UTC and 3 September at (b) 0600 UTC and (c) 1400 UTC. The markers depict the locations of time series shown in Figure 15: Cologne-East (\(\triangle\)), Agger valley (\(\circ\)) and Hohes Venn (\(\ast\)).

**Figure 14.** CO\textsubscript{2} fluxes (\textmu mol(CO\textsubscript{2}) m\textsuperscript{-2} s\textsuperscript{-1}) originating from anthropogenic emissions on 3 September 2012 (used for CL0309): (a) 0000 UTC, (b) 0600 UTC.
The negligible differences in the northern part result from the inflow of identical LBCs for both CO2 tracers (cf. Figure 5(c)).

For consistency, an additional simulation following the method of CS2407−bio was performed, confirming the findings of section 5.1. Neglecting anthropogenic emissions in the nesting simulation, the initial CO2 concentration is 3−10 and 15−50 ppmv lower in rural and urban regions (along the Rhine), respectively. No additional conclusions to the results of section 5.1 and the experiment described above can be drawn with this simulation. Thus, a detailed analysis is omitted in this article.

Finally, Figure 15 depicts selected time series of near-surface CO2 mixing ratios. In the moderately settled Agger valley (●), in the first half of the night the comparable behaviour of the ‘real’ and ‘biogenic’ tracers indicates that the pronounced CO2 increase is mainly of biogenic origin. In the morning, a slightly stronger influence of local anthropogenic CO2 (≈ 15 ppmv) can be seen. The almost identical CO2 content in the afternoon results from advection of inflowing LBCs. At the mountain ridge of the Hohes Venn (★), the diurnal amplitude is distinctly lower, in accordance to the findings of Uebel et al. (2017) and section 3. The slightly higher CO2 contents (≈ 5 ppmv) of the ‘real’ tracer during the daytime are caused by advective transport (see Figure 13(c)). The time series at the anthropogenically influenced Cologne-East (△) location shows a rather different behaviour, with a very prominent peak of the ‘real’ tracer at 1900 UTC followed by 20−40 ppmv higher concentrations than for the ‘biogenic’ tracer at night and in the morning. Whereas the ‘biogenic’ tracer decreases after 0530 UTC, due to photosynthesis, the ‘real’ CO2 tracer remains increased until 0900 UTC, when the convective PBL arises. Even in the well-mixed PBL in the afternoon, the CO2 mixing ratio of the ‘real’ tracer is significantly higher than the corresponding values of the ‘biogenic’ tracer and at the other locations. This shows the intense anthropogenic source at Cologne-East.

6. Discussion

6.1. Dependence of simulated CO2 patterns on atmospheric conditions and complex terrain

The mesoscale application of TerrSysMP-CO2 for a contrasting terrestrial region (mountainous ↔ flat, urban ↔ rural) showed that respiration causes a very heterogeneous CO2 increase in weak synoptic situations at night. In narrow low mountain valleys, the near-surface CO2 accumulation is much stronger than above mountain ridges. As expected, within well-mixed PBLs in the daytime, CO2 is distributed more homogeneously and reduced by photosynthetic uptake. The CO2 content depends strongly on atmospheric conditions. Although respiration is lower, in CL0309 the nocturnal CO2 mixing ratios are significantly higher than in CS2407, due to a very low BLH in this case. Moreover, in the morning of CL0309 clouds delay the onset of photosynthesis as well as the formation of a convective PBL, both inhibiting a CO2 reduction in rural areas, as observed in CS2407.

The mountain–valley contrast of near-surface CO2 mixing ratios results mainly from a buoyancy-driven consumption of TKE in valleys, whereas above mountain ridges TKE is produced by wind shear. Hence, CO2, respired by soil and vegetation, can easily accumulate near valley floors, in contrast to mountain tops, where vertical mixing inhibits a strong accumulation. Convergences of downslope winds in the upper parts of mountain slopes can intensify terrain-following CO2 gradients further. In mathematical terms, the local CO2 tendency at the bottom of a valley is stronger than on the slope and at the top of a mountain ridge, where turbulent and advective transport counteracts the positive CO2 tendency by respiration (and anthropogenic emissions).

These results are in agreement with van der Molen and Dolman (2007), who simulated local TKE maxima at the edge of the east Siberian plateau at night, explaining mesoscale CO2 patterns in a region with modest topographic height changes (500 m over 200 km). By means of a high-resolution simulation (ΔX ≈ 1 km), we pointed out that TKE production above hilltops is a typical feature in complex topography during nights with weak synoptic forcing, also occurring at smaller spatial scales (mountain ↔ valley, i.e. > 200 m over 10 km). Terrain-induced local circulations and turbulent mixing are the main drivers for the complex CO2 patterns and not heterogeneities in the surface CO2 fluxes. Due to this fact, the direct integration of CO2 concentrations from mountain-top stations into inverse transport models, which do not resolve these dynamics would result in pseudo-distributions of terrestrial CO2 fluxes deviating strongly from reality. This is problematic because, with increasing spatial resolution of CO2 fluxes derived from mesoscale inversions, more and more dense observation networks are required. However, many such terrestrial CO2 towers (e.g. on mountain tops, near coast lines) are influenced by mesoscale circulations (van der Molen and Dolman, 2007). This indicates a pressing need to adapt measured CO2 mixing ratios by means of high-resolution simulations before the integration into coarser transport models used for inverse modelling (see also next section). Moreover, we conclude that extensive measurement strategies over complex terrain would be a good opportunity to investigate the influence of mesoscale circulations on CO2 distributions further, to underpin the conclusions drawn from this numerical study. For example, CO2 concentration measurements at the valley floor (capturing e.g. nocturnal CO2 accumulations) and at mountain ridges (indicating the mountain–valley contrasts) could be combined with CO2 flux towers (measuring CO2, moisture and heat exchange at different locations) and flight measurements (capturing PBL dynamics and vertical CO2 distributions).

6.2. Necessity of high-resolution simulations

An important task for the simulation of CO2 patterns within the PBL is to find the maximum horizontal grid spacing to resolve relevant atmospheric flow characteristics influencing terrestrial CO2 mixing ratios, for both forward simulations and mesoscale/regional inversions. The results clearly indicate that, during nights with weak synoptic forcing, even a grid spacing of about 3 km is not sufficient to simulate the strong near-surface gradients in a low mountain range characterized by narrow valleys and mountain ridges (Figure 10). A grid spacing of at least ΔX ≈ 1 km seems to be necessary to resolve terrain-induced circulations and local turbulent patterns controlling the CO2 distribution in stable nocturnal PBLs. The reason for this is the smoothed model topography leading to deviations of elevations and terrain slopes from the real topography. Thus, terrain-induced mesoscale circulations in mountainous regions (e.g. slope winds, mountain–valley circulations, TKE patterns) cannot evolve in simulations with grid spacings of several kilometres. Considering the effective model resolution of ≈ 5ΔX (Bierdel et al., 2012), it is not surprising that, in regions with strong topographic changes on scales below the
effective resolution, model results are poor for some specific atmospheric conditions (e.g. nocturnal PBL). Current regional or even mesoscale model inversions use significantly coarser grids. Hence, it is essential to apply strategies to reduce the representation error of the model for complex terrain stations, which may be very large, as seen in this study. Possible solutions are the use of ‘zoomed’ atmospheric models (e.g. Peylin et al., 2005) or a two-step inversion scheme based on independent models (e.g. Rodenbeck et al., 2009). The latter scheme could also be applied for mesoscale inversions using TerrSysMP-CO₂, as long as a suitable and dense observation network (Lauvaux et al., 2012) or satellite-based information on atmospheric CO₂ contents (e.g. Pillai et al., 2016) is available.

Even apart from complex terrain, significant differences in the near-surface CO₂ distribution of CS2407 and CS2407-low show that a precise simulation of terrestrial CO₂ mixing ratios during nights with weak synoptic forcing is challenging. This should be considered for the use of measured CO₂ mixing ratios in mesoscale inversions to estimate nocturnal respiration rates. In the well-mixed PBL during daytime, the simulated CO₂ mixing ratio is less sensitive to the horizontal grid spacing, due to stronger vertical mixing and higher wind speeds, i.e. terrain-induced circulations have only negligible effects on simulated CO₂ mixing ratios in CS2407. This is a first indication that a grid spacing of few kilometres may be appropriate to estimate high-resolution terrestrial CO₂ fluxes by means of mesoscale inversions on clear-sky days with high BLHs. However, the effect of mesoscale circulations during daytime and thus of different grid spacings depends strongly on atmospheric conditions (e.g. cloud cover, PBL cloudiness, wind, precipitation, BLH...). A comprehensive sensitivity study for different weather situations would be interesting to finally answer the question of which grid resolution is necessary for precise simulations of CO₂ mixing ratios. However, this is beyond the scope of this study.

### 6.3. Anthropogenic signal in densely populated regions

The domain is characterized by an exceptionally high amount of fossil fuel emissions of the total CO₂ source (Table 1), enabling a quantification of the anthropogenic effect on atmospheric CO₂ contents. The emissions result from urban areas along the Rhine and in flat terrain, as well as from three power plants that have some of the highest CO₂ emissions in Europe. The use of very high-resolution data (1 km) based on a recent emission inventory allows for a comparison of the influence of human-produced CO₂ in industrial zones and rural areas with small villages and road networks. Thus, errors arising from coarser datasets, which do not represent the very heterogeneous urban emissions (see e.g. Lac et al., 2013), are reduced. Moreover, the separation into several classes (SNAPs), which have specific annual to diurnal variations (i.e. emission time factors, Figure 3), realizes realistic simulations of the diurnal signal of anthropogenic emissions.

In the summer period, nocturnal near-surface CO₂ increases are caused primarily by respiration because, compared with biogenic fluxes, anthropogenic sources are weak, except for industrial and power plants, which emit mainly above the nocturnal PBL. During the morning rush hour, in valleys and in flat terrain the near-surface anthropogenic signal is very pronounced, especially around cities (up to 50 ppmv). At this time of day, in urban areas fossil fuel emissions are by far the highest CO₂ sources, but also in rural regions the effect of road networks cannot be neglected (Figure 14). With the evolution of a convective PBL, the anthropogenic signal is mixed up to the PBL top (Figure 12). At noon and in the afternoon, even several tens of kilometres downstream of urban areas significantly higher CO₂ mixing ratios occur compared with simulations without fossil fuel emissions (Figures 11, 13). Thus, for inverse modelling a detailed knowledge of fossil fuel emissions is essential to distinguish between anthropogenic and biogenic sources, strongly overlapping in Europe. Whereas without anthropogenic emissions all simulated profiles indicate a net surface CO₂ sink, two of the simulated profiles show a net CO₂ source by including fossil fuel emissions in the simulations (Figure 12). Thus, the influence of urban regions or strong emitters in the near and far vicinity of CO₂ towers has to be considered. However, the anthropogenic signal depends strongly on the BLH as well as on turbulent and advective transport.

The current version of TerrSysMP-CO₂ does not include an urban parametrization. Lac et al. (2013) found modifications in atmospheric conditions (temperature, wind, humidity) by including an urban scheme in their model. Urban heat-island effects, local circulations caused by urban–rural contrasts or stronger vertical mixing over urban areas are not considered in our simulations, but may have an influence on simulated CO₂ mixing ratios. Moreover, the assumed distributions and intensities of CO₂ emissions may differ significantly between different emission inventories (Pillai et al., 2016), which is an unavoidable uncertainty in the quantification of the anthropogenic signal.

### 7. Conclusions

In this study, we presented numerical simulations of spatio-temporal CO₂ variations using the recent biosphere–atmosphere model TerrSysMP-CO₂. We selected a heterogeneous region consisting on the one hand of rural areas including a low mountain range and on the other hand of urbanized flat terrain with exceptionally high anthropogenic emissions.

Strong near-surface CO₂ gradients are simulated at night, resulting from terrain-induced atmospheric dynamics in mountainous regions, as well as in the morning, caused by high fossil fuel emissions in urban areas. During nights with weak synoptic forcing, CO₂ accumulates in narrow valleys due to a stably stratified nocturnal PBL, i.e. vertical turbulent mixing is suppressed by buoyancy-driven consumption of TKE. In contrast, TKE production by vertical wind shear forces an efficient vertical mixing of respired CO₂ above mountain ridges. This contrasting TKE behaviour explains the evolution of near-surface CO₂ gradients in complex terrain. Additionally, convergences due to thermally driven downslope winds enhance these CO₂ gradients further. Simulations using two different horizontal grid resolutions showed that a grid spacing of at least 1 km is necessary to resolve the terrain-induced local dynamics. However, a slightly coarser grid spacing seems to be appropriate to represent the smoother CO₂ distribution in a well-mixed PBL during daytime. These outcomes motivate researchers to perform extensive measurements (e.g. flight campaigns, ground-based CO₂ concentration and flux measurements) over complex terrain to confirm the results of the numerical simulations and to understand the atmospheric circulations better in these regions.

Moreover, very high-resolution data of fossil fuel emissions allowed for a quantification of human-produced effects by comparing CO₂ mixing ratios from realistic simulations with simulations neglecting anthropogenic emissions. Especially during the morning rush hour, below a still existing stable nocturnal PBL a pronounced increase of CO₂ concentrations occurs in urban regions. During daytime, downstream of fossil fuel emissions, a significant anthropogenic signal is simulated up to the PBL top. Thus, a precise knowledge of anthropogenic CO₂ sources is essential for an interpretation of terrestrial CO₂ observations.

Hence, the study provides valuable information for improving high-resolution carbon budgeting by means of regional or mesoscale inverse modelling. Conclusions can be drawn regarding the limitations of using coarser grid spacings as proposed in this study, as well as which effects have to be considered by integrating non-background CO₂ tower observations over complex terrain or in urbanized regions.

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