Simulating the atmospheric CO$_2$ concentration across the heterogeneous landscape of Denmark using a coupled atmosphere-biosphere mesoscale model system

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Abstract. Surface heterogeneity can be challenging to fully encompass by modelling studies of CO$_2$ surface exchanges, especially when it comes to land-sea boarders. The relative importance of the marine and the terrestrial surface fluxes on the atmospheric CO$_2$ concentration were examined by developing a mesoscale modelling framework capable of simulating surface exchanges at a high spatiotemporal resolution. This study exploits the complexity of the Danish landscape and the many land-sea boarders found along the nation’s 7,300 km of coastline.

An atmospheric transport model, DEHM, with a horizontal spatial resolution of 5.6 km $\times$ 5.6 km constituted the basis of the modelling framework. A mechanistic biosphere model, SPA, was coupled to DEHM in order to simulate terrestrial surface exchanges applying a tiling approach with the seven most dominant land-use classes in Denmark to account for sub-grid heterogeneity. Detailed surface fields of pCO$_2$ were used to simulate the air-sea CO$_2$ exchange for the study region. Monthly mean diurnal cycles of surface water pCO$_2$ were imposed onto these, in order to include short-term variability in surface water pCO$_2$.

The Danish biospheric fluxes simulated by the SPA-DEHM model system experienced an east-west gradient corresponding to the distribution of the land-use classes and their biological activity. The relative importance of the seven land-use classes varied throughout the year according to their individual growth patterns. A major contribution to the monthly net ecosystem exchange (NEE) through all seasons came from grasslands, while the influence from croplands increased from March to July. Grasslands had, on an annual basis, the largest impact on the biospheric net uptake with -1,423 GgC yr$^{-1}$.

The total Danish biospheric uptake for 2011 was -6,302 GgC yr$^{-1}$. Relating the annual natural biospheric surface fluxes to the CO$_2$ emitted by fossil fuel combustions and industrial processes by Denmark, the Danish terrestrial uptake corresponded to 52 % of these, while the Danish annual marine uptake was negligible in comparison, although hiding larger seasonal variations.
During 2013-2014, the simulated atmospheric CO\textsubscript{2} concentrations compared well with measurements made at the Risø tall tower located on the shore of Roskilde Fjord (R = 0.88 and RMSE = 4.87 ppm). The origin of the simulated CO\textsubscript{2} concentrations at Risø varied between seasons with biospheric fluxes and fossil fuel emissions having the largest impact on the variations. Impact from Roskilde fjord was difficult to detect in the simulated CO\textsubscript{2} concentrations. These difficulties in simulating the local impact from the Roskilde Fjord might arise from (i) the fjord not being adequately resolved in the constructed model system, (ii) the lack of a realistic representation of the surface water pCO\textsubscript{2} dynamics, or (iii) that the fjord is not in the simulated footprint and only had a modest impact on the simulated atmospheric CO\textsubscript{2} at the Risø tall tower.

1 Introduction

To have the best chance of accurately predicting the future evolution of the carbon cycle, and its implications for our climate, it is important to eliminate, or at least minimise, the uncertainty that exists in the present estimates. Enhanced knowledge, improved temporal resolution of surface exchange processes and improvements in the spatial surface representation are factors that could minimise these uncertainties.

On an annual basis the biosphere is estimated to absorb 3.0 ± 0.8 PgC yr\textsuperscript{-1} of the 9.4 ± 0.5 PgC yr\textsuperscript{-1} anthropogenic carbon emitted to the atmosphere (Le Quéré et al., 2018). The heterogeneity and the dynamics of the land surface complicates such estimates. Biosphere models of various complexity have been developed to spatially simulate surface fluxes of CO\textsubscript{2}, but global bottom-up estimates are poorly constrained by surface observation. Although a huge observational effort has been and is still being put into observing surface exchanges, the observations are far from a global surface representation (Ciais et al., 2014; Zscheischler et al., 2017).

In order to omit the lack of surface observation, atmospheric models have been used to construct inverse modelling systems, where atmospheric CO\textsubscript{2} measurements have been used to constrain surface fluxes (e.g. Baker et al. (2006); Gurney et al. (2002, 2004); Peylin et al. (2013)). These atmospheric inversions are capable of capturing the year to year changes in natural surface fluxes, the magnitude and distribution of regional fluxes, and distinguish between land and ocean fluxes (Le Quéré et al., 2015). However, atmospheric inversions are limited by the availability of atmospheric measurements, and therefore, only regional inversions have been conducted in areas with very dense data network, e.g. Europe (Broquet et al., 2011; Rödenbeck et al., 2009), the corn belt of the United States (Lauvaux et al., 2012), and national scale inversions for the Netherlands (Meesters et al., 2012; Tolk et al., 2011).

Studying surface exchanges of CO\textsubscript{2} on regional to local scale can be accomplished with mesoscale atmospheric transport models. Their resolution is in the range from 2 km to 20 km and their advantage is their capability to get a better processes understanding of both atmospheric and surface exchange mechanisms in order to improve the link between observations and models at all scales, i.e. for both mesoscale, regional and global models (Ahmadov et al., 2007). The higher spatial resolution of mesoscale models allows for a better representation of atmospheric flows and for a more detailed surface description, which in particular for heterogeneous areas is advantageous. In previous mesoscale model studies, biosphere models have been coupled to the mesoscale atmospheric models ranging in their complexity from simple diagnostic (Sarrat et al., 2007b; Ahmadov et al.,...
to mechanistic process based biosphere models (Tolk et al., 2009; Ter Maat et al., 2010; Smallman et al., 2014; Uebel et al., 2017). The modelled CO₂ concentrations and surface fluxes from mesoscale model systems compare better with observations than global model systems (Ahmadov et al., 2009). The atmospheric impact on surface processes related to the ecosystem’s sensitivity and CO₂ exchange can be examined in greater details (Tolk et al., 2009) and tall towers footprints can be studied more concisely (Smallman et al., 2014).

The heterogeneity of coastal ocean contributes with a large uncertainty to assessment of the air-sea CO₂ exchange (Regnier et al., 2013). Coastal seas, rich in nutrient and organic material, might contribute with an in-proportional amount to the global air-sea CO₂ flux, with regards to its limited spatial area when compared to the open oceans (Gattuso et al., 1998). The observed high spatial and temporal variability (Kuss et al., 2006; Leinweber et al., 2009; Vandemark et al., 2011; Norman et al., 2013; Mørk et al., 2016) are not always included in marine models (Omstedt et al., 2009; Gypens et al., 2011; Kuznetsov and Neumann, 2013; Gustafsson et al., 2015; Valsala and Murtugudde, 2015), let alone taken into account in atmospheric mesoscale systems simulating CO₂ (Sarrat et al., 2007a; Geels et al., 2007; Law et al., 2008; Tolk et al., 2009; Broquet et al., 2011; Kretschmer et al., 2014). Moreover, a recent study has found that short-term variability in the partial pressure of surface water CO₂ (pCO₂) can be very influential of the annual flux for some coastal areas (Lansø et al., 2017).

In this study we aim to simulate surface exchanges of CO₂ at a high spatial-temporal resolution together with mesoscale atmospheric transport. Interactions between the atmosphere - ocean, and atmosphere - biosphere are contained in a mesoscale modelling framework together with high resolution anthropogenic emissions of CO₂. The study area comprises of Denmark, a country that is characterised by a varied land mosaic containing urban, forest and agricultural areas. With more than 7,300 km of coastline encircling approximately 43,000 km² of land, many land-sea borders are found throughout the country adding to the complexity. The developed mesoscale modelling system is used to assess and understand the dynamics and relative importance of the marine and terrestrial CO₂ fluxes across this particular region with a special focus on the impact from the Roskilde Fjord system. The Danish Eulerian Hemispheric Model, DEHM, forms the basis of the framework, while the mechanistic biospheric Soil-Plant-Atmosphere model, SPA, is dynamically coupled to the atmospheric model. The air-sea CO₂ exchange is simulated at a high temporal resolution with the best applicable surface fields of pCO₂ for the Danish marine areas. Tall tower observations are used to evaluate the simulated atmospheric concentrations of CO₂.

2 Model setup

The model framework used in the present study consists of two models; DEHM and SPA. A coupling between the two was made for the inner most nest of DEHM in order to simulate the exchange of CO₂ between the atmosphere and terrestrial biosphere at a high temporal (1 hour) and spatial resolution (5.6 km × 5.6 km) for the area of Denmark.

2.1 DEHM

DEHM is an atmospheric chemical transport model covering the Northern Hemisphere with a polar stereographic projection true at 60°N. Originally, developed to study sulphur and sulphate (Christensen, 1997), the DEHM model now contains 58
chemical species and nine groups of particular matter (Brandt et al., 2012). This adaptable model has been used to study such different things as atmospheric mercury (Christensen et al., 2004), persistent organic pollutants (Hansen et al., 2004), biogenic volatile organic compounds influence on air quality (Zare et al., 2014), emission and transport of pollen (Skjøth et al., 2007), ammonia and nitrogen deposition (Geels et al., 2012a, b) and atmospheric CO$_2$ (Geels et al., 2002, 2004, 2007; Lansø et al., 2015). The CO$_2$ version of DEHM was used in the present study. DEHM has 29 vertical levels distributed from the surface to the 100 hPa surface, and horizontally 96 × 96 grid points, which through it’s nesting capabilities increases in resolution from 150 km × 150 km in the main domain to 50 km × 50 km, 16.7 km × 16.7 km and 5.6 km × 5.6 km in the three nests.

2.1.1 Inputs to DEHM

Anthropogenic emissions of CO$_2$, wild fire emissions and optimized biospheric fluxes from NOAA’s ESRL Carbon Tracker system (Peters et al., 2007) version CT2015 were used as inputs to DEHM. Their resolution is 1° × 1° with updated values every third hour. Similarly, CT2015 three-hourly molefractions of CO$_2$ were used as boundaries towards the Southern Hemisphere.

Hourly anthropogenic emissions on a 10 km × 10 km grid from the Institute of Energy Economics and the rational Use of Energy (IER, Pregger et al. (2007)) were applied for Europe instead of emissions from CT2015. Furthermore, these are for the area of Denmark substituted by hourly anthropogenic emissions with an even higher spatial resolution of 1 km × 1 km (Plejdrup and Gyldenkærne, 2011). As the European and Danish emission inventories were from 2005 and 2011, respectively, the emissions were scaled to annual national total CO$_2$ emission of fossil fuel and cement production conducted by EDGAR (Olivier et al., 2014), in order to include the yearly variability in national anthropogenic CO$_2$ emissions.

The necessary meteorological parameters for DEHM were simulated by the Weather Research and Forecast Model (WRF) (Skamarock et al., 2008), nudged by six hourly ERA-Interim meteorology, which was also used as initial and boundary conditions (Dee et al., 2011).

2.1.2 Air-sea CO$_2$

The exchange of CO$_2$ between the atmosphere and the ocean, $F_{CO_2}$, was calculated by $F_{CO_2} = Kk_{660}\Delta p_{CO_2}$, where $K$ is solubility of CO$_2$ calculated as in Weiss (1974), $k_{660}$ is the transfer velocity of CO$_2$ normalised to a Schmidt number of 660 at 20°C, and $\Delta p_{CO_2}$ is the difference in partial pressure of CO$_2$ between the surface water and the overlying atmosphere. The transfer velocity parameterisation $k = 0.266u^2_{10}$, where $u^2_{10}$ is the wind speed at 10 m, determined by Ho et al. (2006) has been found to match Danish fjord systems (Mørk et al., 2016), and was applied in the current study. Surface values of marine pCO$_2$ were described by a combination of the open ocean surface water climatology of pCO$_2$ by Takahashi et al. (2014), and the climatology developed by Lansø et al. (2015, 2017) for the Baltic Sea and Danish waters. Furthermore, short-term temporal variability was accounted for in the surface water pCO$_2$ by imposing monthly mean diurnal cycles onto the monthly climatologies following the method described in Lansø et al. (2017).
2.2 SPA

The SPA model is a mechanistic terrestrial biosphere model (Williams et al., 1996, 2001). SPA has a high vertical resolution with up to 10 canopy layers (Williams et al., 1996) allowing for variation in the vertical profile of photosynthetic parameters and multi-layer turbulence (Smallman et al., 2013). Within the soil up to 20 soil layers can be included (Williams et al., 2001).

The radiative transfer scheme estimates the distribution of direct and diffuse radiation, and sunlit and shaded areas (Williams et al., 1998). SPA uses the mechanistic Farquhar model (Farquhar and von Caemmerer, 1982) of leaf level photosynthesis, and the Penman-Monteith model to represent leaf level transpiration (Jones, 1992). Photosynthesis and transpiration are coupled via a mechanistic model of stomatal conductance, where stomatal opening is adjusted to maximise carbon uptake per unit nitrogen within hydraulic limitations, determined by a minimum leaf water potential tolerance, to prevent cavitation.

Ecosystem carbon cycling and phenology is determined by a simple carbon cycle model (DALEC, Williams et al. (2005), which is directly coupled into SPA. DALEC simulates carbon stocks in foliage, fine roots, wood (branches, stem and coarse roots), litter (foliage and fine root) and soil organic matter (including coarse woody debris). Photosynthate is allocated to autotrophic respiration and living biomass via fixed fractions, while turnover of carbon pools is governed by first order kinetics. In addition, when simulating crops, a storage organ (i.e. the crop yield) and dead, but still standing, foliage pools are added (Sus et al., 2010).

SPA has been extensively validated against site observations from temperate forests (Williams et al., 1996, 2001), temperate arable agriculture (Sus et al., 2010) and Arctic tundra (Williams et al., 2000). SPA has more recently been coupled into the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008), the resulting WRF-SPA model was used in multi-annual simulations over the United Kingdom and assessed against surface fluxes of CO$_2$, H$_2$O and heat, and atmospheric observations of CO$_2$ from aircraft and a tall tower (Smallman et al., 2013, 2014).

SPA needs vegetation and soil input parameters. Initial soil carbon stock estimates were obtained from the Regridded Harmonized World Soil Database (Wieder, 2014). The vegetation inputs and plant traits for SPA were partly taken from previous parameter sets used in SPA, but also from the Plant Trait Database (TRY, Kattge et al. (2011)), and from literature (Penning de Vries et al., 1989; Wullschleger, 1993). As these parameters and plant traits were determined at various sites that not necessarily corresponds to Danish conditions, a calibration of the vegetation inputs to SPA was conducted for Danish Eddy Covariance (EC) flux sites (Table 1). Only data from five sites were available and these were divided in two sets – one for calibration (all available observations before 2013), the other validation (all available observations from 2013 and 2014).

2.2.1 SPA calibration

The calibration was conducted by selecting a set of inputs parameters (plant traits, carbon stocks etc.) and for each parameter five values within a realistic range were chosen. Next, 200 SPA simulations with randomly chosen parameter values were conducted. These results were statistically evaluated against observations of net ecosystem exchange (NEE) from the different flux sites, and the simulations with the lowest root mean square error (RMSE) in combination with highest correlation were selected. Based on this random parameter testing, it was possible to choose the best set of realistic vegetation input parameters.
that could improve the model performance at the Danish sites. The best found vegetation parameters values corresponded in some cases to the values already applied in SPA for the given land cover.

### 2.2.2 Validation

Comparing to observations of NEE, SPA was, in general, able to capture the phenology and seasonal cycle throughout the entire simulation period (Fig. 1). Correlations and RMSEs between the model and the independent data from the validation period (Fig. 1) likewise indicate a good model performance. At Sorø both variability, as inferred from the standard deviations, the amplitude and the onset of the growing season were well reproduced by the SPA model. However, difficulties with simulating the evergreen forest at Gludsted is evident with more variation modelled than given by the observations, and a lag of the start of the growing season when compared to the observations. The evergreen plant functional type in SPA experiences phenological problems with rapid leaf growth in response to environmental drivers, which causes a delay in spring photosynthesis. Voulund alternates between winter and spring barley for the calibration period starting with winter barley in 2009. Note that the whole observed time series of NEE at Voulund is shown together with model NEE of both winter and spring barley (Fig. 1c and d). While the phenology and amplitude are well captured by spring barley at Voulund, SPA is not able to capture the seasonal amplitude of the winter barley that seems to be more sensitive to the meteorological drives, and seasons with harder winters had lower NEE peaks in summer. At the grassland site Skjern Enge, NEE is for winter, spring, and the first part of the summer reasonably modelled. The difficulties for late summer and autumn arise from the management practices at the site, where both grazing and grass cutting are conducted, limiting NEE (Herbst et al., 2013). Although grazing is included in the SPA model, it does not simulate the same reduction in NEE.

In the inner most nest of DEHM for the area of Denmark, a coupling was made between DEHM and SPA. Thus, the coarser optimized biospheric fluxes from CT2015 were for Denmark replaced by hourly SPA simulated CO$_2$ fluxes. With this change, the spatial resolution for the biosphere fluxes was increased from $1° \times 1°$ to 5.6 km $\times$ 5.6 km allowing for a better representation of the Danish surface, and hence also the biospheric fluxes.

A tiling approach with seven most common biospheric land-use classification were selected for this study including deciduous forest, evergreen forest, winter wheat and other winter crops, winter barley, spring barley and other spring crops, grassland and agricultural other, but excluding urbanised areas. This classification corresponds to the actual crop distribution of 2011 (Jepsen and Levin, 2013). Denmark is dominated by agriculture, and more than 60 % of the used classification is agricultural land. DEHM provides on an hourly basis meteorological drivers and atmospheric CO$_2$ concentrations to SPA, while SPA each hour returns NEE to DEHM.

### 2.3 Observations of atmospheric CO$_2$

Tall tower continuous measurements of atmospheric CO$_2$ concentrations made by a Picarro 118 m above the surface have been conducted at the Risø site since the middle of 2013. The Risø site is located on the eastern inner shore of Roskilde Fjord (55°41’N, 12°05’E), Zealand, which is a narrow microtidal estuary 40 km long with a surface area of 123 km$^2$ and a mean depth of 3 m (Mørk et al., 2016).
3 Results

The model system was run from 2008 to 2014, however, the first three years were regarded as a spin-up period. Given that the land-use classification corresponds to the actual distribution of 2011 an emphasis will be put on the terrestrial fluxes for this particular year during the analysis together with an estimation of the annual Danish carbon budget. Measurements of atmospheric CO$_2$ will be used to assess the performance of the DEHM-SPA model system, and evaluate local impacts from fjord systems on atmospheric CO$_2$ concentrations.

3.1 Surface fluxes

3.1.1 Biospheric fluxes

Across Denmark, there is an east-west gradient in the simulated biospheric fluxes of carbon for both January and July in 2011. Larger fluxes of gross primary productivity (GPP), ecosystem respiration and NEE are evident in the western part of Denmark, while the islands and Eastern Jutland have lower biosphere fluxes (Fig. 2). This gradient follows the distribution of the individual land-use classifications (Appendix A Fig. A1) and their grow patterns, but also reflects the population density which is highest in the eastern part of the country.

Table 2 and Table 3 show the contribution of each individual land-use class to the total GPP and respiration on a national scale (see Appendix B Table B1 - B3 for monthly GPP, respiration and NEE per land-use class). The monthly contributions to the country-wide total inherently reflect the total area for each land-use class. In winter, GPP is highest for evergreen, grassland and agricultural other. These land-use classes are well represented in Western Jutland explaining why the largest biological production is found here during January (Fig. 2a). As the crops develop, their contribution to the total monthly GPP increases, and in July winter crops are together with grasslands the most productive land-use classes. Also, deciduous forest increases its share of GPP from the onset of the growing season, but as its spatial extent only amount to 10%, its monthly contribution is never dominant. Respiration is less concentrated for individual land-use classes and the individual monthly contributions vary much less for respiration than GPP throughout the year, though with a notable increase in contribution from both winter and spring crop through spring and summer until harvest (Table 3). The highest contributions of respiration are throughout the year found for grassland and agriculture other.

Integrating over all land-use classes for 2011, the Danish terrestrial land surfaces is net source of CO$_2$ to the atmosphere in the months from January to April, and October to December, with the highest monthly release of 1,129 GgC month yr$^{-1}$ in December. From May to September, the biosphere is a net sink with a maximum uptake in June of -4,687 GgC month yr$^{-1}$. The total annual surface exchange of CO$_2$ between the atmosphere and Danish biosphere is -6,302 GgC yr$^{-1}$ for 2011, where grassland has the largest contribution with -1,423 GgC yr$^{-1}$. 
3.1.2 Marine fluxes

The air-sea CO$_2$ exchange in the Danish inner waters experiences large seasonal variations, while the variations in the North Sea are less pronounced as illustrated by Fig. 3. Bordering the Baltic Sea, the Danish inner waters are rich in nutrients and organic material (Kuliński and Pempkowiak, 2011). This fosters high biological activity in spring and summer lowering surface water pCO$_2$ allowing for uptake of atmospheric CO$_2$. In winter, mineralisation increases pCO$_2$ (Wesslander et al., 2010), and outgassing of CO$_2$ to the atmosphere takes place. The North Sea is a continuously sink of atmospheric CO$_2$, where a continental shelf-sea pump removes pCO$_2$ from the surface water and transport it to the North Atlantic Ocean (Thomas et al., 2004).

To estimate the annual exchange of CO$_2$ between the atmosphere and the ocean for the Danish marine areas, the exclusive economic zone (EEZ) is used. In the EEZ the coastal state (in this case Denmark) has the right to explore, exploit and manage all resources found within it (United Nations Chapter XXI: Law of the Sea, 1984). Thus, assuming the air-sea CO$_2$ exchange counts as a natural resource for Denmark, the air-sea flux from EEZ was used for the annual budget estimation of CO$_2$. The 2011 simulated annual air-sea CO$_2$ exchange in the 105,000 km$^2$ covered by the Danish EEZ amounts to -422 GgC yr$^{-1}$. However, this number hides large spatial differences and monthly numerical larger fluxes. While the North Sea area contained within the EEZ continuously had monthly uptakes in the range -73 GgC mon$^{-1}$ to -191 GgC mon$^{-1}$, the monthly fluxes from the near coastal (marine areas extending up to 10 km of shore) Danish inner waters varied in the range -46 GgC mon$^{-1}$ to 540 GgC mon$^{-1}$.

3.1.3 Danish CO$_2$ budget

The annual CO$_2$ budget for year 2011 is assessed, because the crop distribution used for the land-use classification was based on data from this specific year as was the spatial distribution of the Danish fossil fuel emissions. In 2011, Denmark emitted 12,205 GgC yr$^{-1}$ of CO$_2$ to the atmosphere due to fossil fuel combustion and industrial processes (Nielsen et al., 2015). The Danish terrestrial biosphere took up -6,302 GgC yr$^{-1}$ of CO$_2$ during 2011, which equals 52 % of the CO$_2$ emitted by anthropogenic activity in Denmark. The marine uptake of -422 GgC yr$^{-1}$ is annually less influential and corresponds to only 3.5 % of the anthropogenic emitted CO$_2$.

3.2 Atmospheric CO$_2$ concentrations

The time series of measured and simulated CO$_2$ show good agreements (Fig. 4) with R = 0.88 and RMSE = 4.87 ppm for daily averaged time series demonstrating that the model is capable of capturing the synoptic scale variability. Also, good statistical measures are obtained for the hourly time series with R = 0.84 and RMSE = 5.95 ppm, but the short-term variability is not always fully captured by the model. Overall the model simulates the atmospheric CO$_2$ quite well, indicating that the simulated surface exchange of CO$_2$ is acceptable.

To investigate the origin of the CO$_2$ simulated at the Risø site, concentration rose plots of simulated atmospheric CO$_2$ have been made (Fig. 5). The concentration rose shows the wind direction and associated CO$_2$ concentrations. Roskilde Fjord lies...
in the approximate sector of 200°–360° relative to the Risø tower, the city of Roskilde (with 50,000 inhabitants) is positioned approximately 5 km south to south-west of the tower, while Copenhagen lies 20 km east. Division has been made between seasons, and day and night time values both showing distinct seasonal and diurnal patterns. The highest values of CO₂ are obtained during winter, where very little diurnal variation is seen. During summer the lowest values are obtained in particular during daylight, when photosynthesis occurs.

The individual contribution from fossil fuel emissions, marine and biospheric exchanges to the atmospheric CO₂ (see Appendix C Fig. C1 - C3) indicate that the biosphere contributes most to the variations simulated at Risø (Fig. C2) – both seasonally and daily. Emissions of fossil fuel experience little diurnal variability, but seasonally with the greatest contribution during autumn and winter (Fig. C1). Highest values are seen originating from the sectors encapsulating the city of Roskilde and the capital region. In all seasons, the simulated oceanic contribution is negative, i.e. indicating uptake of atmospheric CO₂, but the marine contribution is small with little variation (Fig. C3). The less negative values in autumn and winter may be a result of the simulated outgassing of CO₂ from the Baltic Sea and Danish inner waters during the winter season (Lansø et al., 2015), which however is still dominated by the uptake by global open oceans.

The local impact from Roskilde Fjord is difficult to detect in the marine concentration plots. Flux measurements at Roskilde Fjord have shown uptake of CO₂ during spring, while release in the remaining seasons (Mørk et al., 2016), which is accurately captured by the modelling system (Lansø et al., 2017). A footprint analysis of the Risø tower has shown that the fluxes from Roskilde Fjord have a contribution to the total CO₂ flux measured at the top of the 118 m high tower, but only minor, since fluxes over water typically is an order of magnitude smaller than fluxes over land (Sogachev and Dellwik, 2017). Therefore, we investigated a period with observed large outgassing from Roskilde Fjord - a storm event in October 2013 that was observed to increase the monthly release of CO₂ in the fjord by 66 % (Mørk et al., 2016). The storm event passed Denmark on 28 October 2013, and at 06 UTC southerly winds transport air masses with higher CO₂ towards the Risø site (Fig. 6a), while at the same time a detectable increase in the oceanic contribution to the CO₂ concentration at the Roskilde Fjord system is seen (Fig. 6b). The model system simulates the small peak in the observed atmospheric CO₂ concentrations for 28 October (Fig. 7a) at the Risø site, but distinguishing between contributions from fossil fuel emissions, the biosphere and the ocean to the atmospheric CO₂ concentration at Risø (Fig. 7b) reveals no oceanic impact, and hence no apparent influence from Roskilde Fjord during the storm event.

4 Discussion

4.1 Biospheric fluxes

Spatially, the Danish biospheric fluxes of CO₂ follows the land-use classification that mirrors the population density being highest in eastern and lowest in western Denmark. While GPP follows phenology and productivity resulting in changing percentage contribution from the land-use classes to the monthly total GPP, the mutual proportions of respirations are much less varied. Autotrophic respiration depends on plant productivity, but the heterotrophic respiration is temperature dependent and will change proportionally for each land-use class maintaining a more constant percentage-wise distribution.
The simulated annual uptake by deciduous forest of -300 gC m\(^{-2}\) yr\(^{-1}\) for 2011, fits the observed range of annual estimated NEE at Sorø from 1996 to 2009 spanning 32 gC m\(^{-2}\) yr\(^{-1}\) to -331 gC m\(^{-2}\) yr\(^{-1}\) (Pilegaard et al., 2011). Even though SPA experiences a lag in the seasonal onset for the evergreen forest, the annual estimated uptake of -386 gC m\(^{-2}\) yr\(^{-1}\) compares well with previous estimates of temperate evergreen forests with -402 gC m\(^{-2}\) yr\(^{-1}\) (Luyssaert et al., 2007) and Danish evergreen plantations of -503 gC m\(^{-2}\) yr\(^{-1}\) (Herbst et al., 2011). However, improvements to the evergreen plant functional type in SPA are needed, and an addition of a labile pool to the evergreen carbon assimilation would omit the seasonal lag (Williams et al., 2005). Such adjustments have already been made to the DALEC carbon assimilation system utilised by SPA (Smallman et al., 2017), but not yet incorporated into SPA.

Previous annual estimates at Danish agricultural field sites found carbon uptake with -31 gC m\(^{-2}\) yr\(^{-1}\) estimated from a mixed agricultural landscape (Soegaard et al., 2003) and -245 gC m\(^{-2}\) yr\(^{-1}\) from winter barley (Herbst et al., 2011). The SPA-DEHM model system simulated annual uptakes for winter wheat and spring crops of -137 gC m\(^{-2}\) yr\(^{-1}\) and -207 gC m\(^{-2}\) yr\(^{-1}\), respectively, and thus fits the previous estimates well, while winter barley had a small release of 32 gC m\(^{-2}\) yr\(^{-1}\).

The calibration and validation (1c) shows difficulties in simulating the observed NEE during growing seasons for winter barley particularly after cold and snow covered winters (winter 2010-11, 2012-13). For 2011 winter barley had the smallest total area of the agricultural land-use classifications, and was dominated by the respiration. As pointed out in previous studies, the crop modelling component in SPA could likewise be improved e.g. inclusion of intra-seasonal crops (Smallman et al., 2014). The current study annually estimated the Danish grasslands to be a sink of CO\(_2\) with -205 gC m\(^{-2}\) yr\(^{-1}\), which are similar to the -267 gC m\(^{-2}\) yr\(^{-1}\) observed at the Skjern Enge grassland site(Herbst et al., 2011) and the -312 gC m\(^{-2}\) yr\(^{-1}\) observed at the Lille Valby grassland site, Denmark (Gilmanov et al., 2007). The European grassland study by Gilmanov et al. (2007) found large variation in annual fluxes from grassland driven by environmental conditions and management practises at the sites varying from 171 gC m\(^{-2}\) yr\(^{-1}\) to -707 gC m\(^{-2}\) yr\(^{-1}\), but with most site having an annual uptake of carbon. As seen in Fig. 1e more work on grassland calibration could have been done, but the conditions and management regimes at Skjern Enge does not necessarily fit the rest of the Danish grassland. With the chosen parameters, very comparable results were obtained indicating that such an additional calibration might not be advantageous.

A tilling approach has been used for the land-use classification in the SPA-DEHM modelling framework, including sub-grid heterogeneity in the model system. However, the seven land-use classes do not fully encompass the ecosystem variability in Denmark. Both grassland and agricultural other cover a broad range of sub-categories with both heather and meadow included in the grassland class, while agricultural other among other things contains vegetables fields, hedgerows, woodland patches and uncultivated land. Moreover, large urbanised areas are not accounted for in the current classes either. Adding more land-use classifications could give a better and more realistic surface description, if data for both calibration of validation for the lacking land-use classes, preferably from similar climatic region as Denmark, were available.

### 4.2 National CO\(_2\) budget

The current estimate of the global carbon budget appraises the global biosphere to take up 32 % of the CO\(_2\) emitted by fossil fuel and industry, while the global oceans are estimated to take up 26 % (Le Quéré et al., 2018). National scale biospheric
uptakes can vary greatly between countries depending on land coverage, land-use and management practices (Janssens et al., 2005). Meesters et al. (2012) estimated an annual biospheric uptake for the Netherlands of -17,400 GgC yr\(^{-1}\) (approximately -497 gC m\(^{-2}\) yr\(^{-1}\)), which corresponded to 33 % of the annual fossil fuel emitted by the country. An annual uptake of -99 gC m\(^{-2}\) yr\(^{-1}\) has been estimated for Scotland by Smallman et al. (2014). The current study estimates a total annual biospheric uptake of -6,302 Gg C yr\(^{-1}\), which equals 52 % of the CO\(_2\) emitted by anthropogenic activities in Denmark. Integrating over all land-use classes the uptake per area is -195 gC m\(^{-2}\) yr\(^{-1}\), which places this Danish estimate within the bounds of the previous national estimates at similar latitudes. Caution should be taken when assessing the Danish CO\(_2\) budget. We present a one year snap-shot of the state of the Danish surface exchanges of CO\(_2\), but important processes such as product use, biomass burning and river runoff linking land and ocean are lacking in our estimate in order to fully close the budget.

4.3 Atmospheric CO\(_2\) and land-sea signals

The SPA-DEHM modelling system resembles the synoptic and diurnal variability in the atmospheric CO\(_2\) concentrations measured at Risø site. The variability at the Risø site is dominated by the biospheric impact and fossil fuel emissions of CO\(_2\). The signal from Roskilde Fjord is difficult to detect in the simulated CO\(_2\) concentrations. Even when the marine contribution to the atmospheric concentration alone is examined, the Roskilde Fjord signal is hard to distinguish at the Risø tower. Rather the global and regional signals are obtained, e.g. the winter release of CO\(_2\) from the Baltic Sea resulting in less negative values for the concentration rose plot. Even though the oceanic contribution to the atmospheric short-term variability is low, oceanic impact is still important on monthly and annual scale time scales. As the Roskilde Fjord by a footprint analysis previously was found to have an impact on the atmospheric CO\(_2\) concentration at the top of the tower (Sogachev and Dellwik, 2017), a period with observations of large outgassing from Roskilde Fjord was examined to more clearly envisions its impact in the constructed modelling system. Both the simulated and observed atmospheric CO\(_2\) increased during the storm event on October 28 (Fig. 6a), but no concurrent increase was seen for the oceanic contribution to atmospheric CO\(_2\) at the Risø site (Fig. 6b). This might be explained by the southerly winds that transports the CO\(_2\) released from the fjord northward and away from the Risø Tower, which is positioned in the southern part of the fjord. Moreover, in this study the increased flux from Roskilde Fjord was only caused by increased wind speed together with the impose diurnal cycle of marine pCO\(_2\) (the diurnal amplitude for October was approximately 10 \(\mu\)atm), while measurements suggested that also an increase in surface water pCO\(_2\) of approximately 300 \(\mu\)atm sustained the observed CO\(_2\) flux (Mørk et al., 2016). The lack of such increase in surface water pCO\(_2\) in the current modelling study, could explain why no impact on the simulated atmospheric CO\(_2\) is seen from the marine component during the storm event.

Thus the results could indicate that (i) the narrow Roskilde Fjord was not sufficiently resolved in the current model framework, where the horizontal grid resolution is 5.6 km \(\times\) 5.6 km, (ii) the surface water pCO\(_2\) was not described in enough details in the model system, or (iii) Roskilde fjord is not in the footprint of the tower and thus only has a minor impact on the atmospheric CO\(_2\) concentrations at Risø.
4.4 Uncertainties in relation to surface exchanges of CO$_2$

Some of the largest uncertainties lie in the parameters underlying the terrestrial carbon cycle, in particular those governing allocation to plant tissues and their subsequent turnover. Most often these are based on maps of land cover or plant functional type, but parameter estimation via data assimilation analysis has shown substantial spatial variation of terrestrial ecosystem parameters within plant functional type groupings with consequences for carbon cycling predictions (Bloom et al., 2016). Increasing the amount of observational data used in data assimilation system have been found to reduce uncertainty in retrieved parameters and thus simulated carbon stocks and fluxes of CO$_2$ (Smallman et al., 2017); including all observations counting both in situ and satellite, Smallman et al. (2017) halved the uncertainty of the net biome productivity. While SPA also uses DALEC to simulate carbon allocation and turnover, it is currently impractical to conduct a similar data assimilation analysis or to repeatedly simulate atmospheric transport to robustly quantify the impact of flux uncertainties on atmospheric CO$_2$ concentrations due to their computational requirements. While conducting such an analysis remains a future ambition we consider it to be out of scope for the current study, since the terrestrial surface fluxes in this study is constrained by one data stream consisting of EC measurements. This study has focused on surface fluxes over a relative short time period, and the model framework was capable of producing such fluxes a satisfactorily level, including their aggregated impact on atmospheric CO$_2$ concentrations (Fig. 4) with $R = 0.88$ and RMSE = 4.87 ppm for daily values. The usage of satellite retrievals by data assimilations systems and their accompanying improvements moreover highlights the future enhancement to the current modelling framework, where satellite products could be utilized for upscaling reducing the related error.

Uncertainties of the marine fluxes can be associated with both the choice of transfer velocity parameterisation, choice of wind speed product and the used surface water pCO$_2$ maps. Sensitivity analysis of global transfer velocity parameterisation based on $^{14}$C bomb inventories shows uncertainties of 20 %, while varying the applied wind speed products for these formulation increase the difference in the global annual flux by 40 % (Roobaert et al., 2018). Including empirical formulations of transfer velocity parameterisation in the analysis increase the sensitivity to wind speed product to nearly 70 %, while the uncertainty to the parameterisation itself is more the 200 %. More than a doubling of the annual uptake by the usage of different transfer velocity formulations has likewise been shown for the study region (Lansø et al., 2015), while the choice of surface map could change the study region from an annual sink to source of atmospheric CO$_2$ (Lansø et al., 2017) As shown by Roobaert et al. (2018) the ERA-Interim and the transfer velocity formulation by Ho et al. (2006) used in the present study have a combined uncertainty estimate around 20 %. The improved data-driven near coastal Danish pCO$_2$ climatology better reflects the observed spatial dynamics and seasonality in the Danish inner waters (Mørk, 2015), albeit not diminishing the uncertainty related to surface maps of pCO$_2$ but reducing it.

5 Conclusions

By usage of the designed mesoscale modelling framework, it was possible to get a detailed insight into the spatio-temporal variability of the Danish surface exchanges of CO$_2$. The simulated biospheric fluxes experienced an east-west gradient corresponding to the distribution of the land-use classes and their biological activity. The relative importance of the seven land-use
classes varied throughout the course of the year. In general, grasslands had a high contribution to the monthly NEE through all seasons, while crop-lands influence grew from March to July. On an annual basis, grasslands had the largest impact on the biospheric uptake with -1,423 GgC yr$^{-1}$.

The 12,205 GgC yr$^{-1}$ of CO$_2$ emitted by Danish fossil fuel during 2011 is comparable to almost double the uptake by the Danish land and marine areas with a biospheric uptake of 52% and marine uptake corresponding to a few percent. The Danish total annual biospheric uptake was not unreasonable, when compared to other national scale annual estimates. However, the biospheric uptake could benefit both from model improvement and divisions into more land-use classes.

Good accordance between simulated and observed concentrations was found between modelled and observed atmospheric CO$_2$ concentrations for 2013 and 2014 at the Risø site. The origin of the modelled CO$_2$ concentrations at Risø varied with biospheric fluxes having the largest impact on diurnal variability, while on a seasonal scale fossil fuel emissions also had a dominant role. The local impact from Roskilde Fjord was difficult to detect, while regional impact from the Baltic Sea and Danish inner Straits are apparent in winter. The results may indicate that Roskilde Fjord and its localised impact (i.e. at the Risø site) on atmospheric CO$_2$ is not adequately resolved in the current model set-up or only have modest effect.

In order to further examine the air-sea signal at the complex Risø site surrounded by a mosaic of fjord systems, land masses and the Danish inner water, more model experiments could be made, where a bigger focus was put on other marine areas than Roskilde Fjord as e.g. the Danish Inner Straits, Kattegat and the Baltic Sea. Although the total annual marine flux was small, it disguises large monthly variations, and further investigations could help to understand the carbon dynamics in coastal regions. A runoff component in the modelling system would moreover be beneficial for such studies.

Code availability. Scientist with an interest in the atmospheric chemical transport model, DEHM, can contact Jesper H. Christensen (jc@envs.au.dk) with enquiries. Scientist with an interest in the Soil-Plant-Atmosphere model, SPA, can visit its webpage (https://www.geos.ed.ac.uk/homes/mwilliam/spa.html) or contact Mathew Williams (mat.williams@ed.ac.uk).

Appendix A: Land-use classification

Appendix B: Biospheric fluxes

Appendix C: Concentration Roses

Competing interests. The authors declare that there is no competing interests.
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Figure 1. Monthly averaged values of observed (black dashed, calibration period; black solid, validation period) and simulated (red) net ecosystem exchange (NEE) plotted together with standard deviations (shaded area) for the Danish flux sites with measurements in the simulation period. The model mean (Mean\textsubscript{model}), observational mean (Mean\textsubscript{obs}), correlation (R) and root mean square error (RMSE) for the validation period (2013 and 2014) are shown for each site.
Figure 2. (a, b) 2011 January and July gross primary productivity (GPP), (c, d) 2011 January and July respiration including autotrophic and heterotrophic respiration, (e, f) 2011 January and July NEE.
Figure 3. Simulated air-sea CO$_2$ exchange for January (a) and July (b) 2011.
Figure 4. One-hour averages and daily averages of modelled and continuously measured atmospheric CO$_2$ at the Risø site for 2013-2014. The annual mean values have been removed from the time series.
Figure 5. Concentration roses of modelled atmospheric CO$_2$ [ppm] at the Risø site for 2011-2014. The wind direction is split into 10° intervals and the frequency indicated by the concentric circles. The colours indicate the CO$_2$ concentrations with mean removed that have been transported to the site from the given wind directions.
Figure 6. (a) Atmospheric CO$_2$ concentrations including the annual background across Denmark 28 October 2013 06 UTC during the October storm. (b) The contribution to the atmospheric CO$_2$ concentration alone from marine exchange 28 October 2013 06 UTC. The less negative values at the Roskilde Fjord system indicates release of CO$_2$ to the atmosphere.
Figure 7. (a) One-hour averages of modelled and continuously measured atmospheric CO$_2$ at the Risø site for 19-29 October 2013 with annual means removed. (b) Contributions from fossil fuel emission, oceanic surface exchange and biospheric surface exchange to the atmospheric CO$_2$ concentration, shown as one-hour averages of modelled concentrations at the Risø site for 19-29 October 2013.
Figure A1. Square kilometers of the seven land-use classification contained in each model grid.
**Figure C1.** Concentration roses of the fossil fuel emission contribution to the modelled atmospheric CO$_2$ [ppm] at the Risø site for 2011-2014. The wind direction is split into 10° intervals and the frequency indicated by the concentric circles. The colours indicate the CO$_2$ contribution that have been transported to the site from the given wind directions.
Figure C2. Concentration roses of the biospheric contribution to the modelled atmospheric CO₂ [ppm] at the Risø site for 2011-2014. The wind direction is split into 10° intervals and the frequency indicated by the concentric circles. The colours indicate the CO₂ contribution that have been transported to the site from the given wind directions.
Figure C3. Concentration roses of the oceanic contribution to the modelled atmospheric CO$_2$ [ppm] at the Risø site for 2011-2014. The wind direction is split into 10° intervals and the frequency indicated by the concentric circles. The colours indicate the CO$_2$ contribution that have been transported to the site from the given wind directions.
**Table 1.** Location, species, land-use classification in the model system for the five Danish Eddy Covariance (EC) sites used for calibration and validation of the SPA model.

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<th>Location</th>
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<th>Validation</th>
<th>Species</th>
<th>LU in SPA-DEHM</th>
<th>Reference</th>
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<td>2013-2014</td>
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<td>Spring/winter barley</td>
<td>HOBE</td>
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Table 2. Percentage of monthly GPP from each land-use class to the total monthly GPP for Denmark during 2011.

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Table 3. Percentage of monthly respiration from each land-use class to the total monthly respiration for Denmark during 2011.

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Table B1. Monthly gross primary production (GPP) for Denmark [GgC/month] as simulated by each of the seven land-use classes.

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Table B2. Monthly respiration for Denmark [GgC/month] as simulated by each of the seven land-use classes

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Table B3. Monthly net ecosystem exchange (NEE) for Denmark [GgC/month] as simulated by each of the seven land-use classes

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