



# The potential of using remote sensing data to estimate air–sea CO<sub>2</sub> exchange in the Baltic Sea

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**Abstract.** In this article, we present the first climatological map of air–sea CO<sub>2</sub> flux over the Baltic Sea based on remote sensing data: estimates of  $p\text{CO}_2$  derived from satellite imaging using self-organizing map classifications along with class-specific linear regressions (SOMLO methodology) and remotely sensed wind estimates. The estimates have a spatial resolution of 4 km both in latitude and longitude and a monthly temporal resolution from 1998 to 2011. The CO<sub>2</sub> fluxes are estimated using two types of wind products, i.e. reanalysis winds and satellite wind products, the higher-resolution wind product generally leading to higher-amplitude flux estimations.

Furthermore, the CO<sub>2</sub> fluxes were also estimated using two methods: the method of Wanninkhof et al. (2013) and the method of Rutgersson and Smedman (2009). The seasonal variation in fluxes reflects the seasonal variation in  $p\text{CO}_2$  unvaryingly over the whole Baltic Sea, with high winter CO<sub>2</sub> emissions and high  $p\text{CO}_2$  uptakes. All basins act as a source for the atmosphere, with a higher degree of emission in the southern regions (mean source of 1.6 mmol m<sup>-2</sup> d<sup>-1</sup> for the South Basin and 0.9 for the Central Basin) than in the northern regions (mean source of 0.1 mmol m<sup>-2</sup> d<sup>-1</sup>) and the coastal areas act as a larger sink (annual uptake of -4.2 mmol m<sup>-2</sup> d<sup>-1</sup>) than does the open sea (-4 mmol m<sup>-2</sup> d<sup>-1</sup>). In its entirety, the Baltic Sea acts as a small source of 1.2 mmol m<sup>-2</sup> d<sup>-1</sup> on average and this annual uptake has increased from 1998 to 2012.

## 1 Introduction

From the early 2000 and onwards, there has been a more active attempt to investigate, understand, and quantify the global carbon cycle by the scientific community since the greenhouse gas carbon dioxide (CO<sub>2</sub>) plays a key role in controlling Earth's climate. The oceanic uptake of anthropogenic CO<sub>2</sub> helps regulate atmospheric CO<sub>2</sub> through air–sea exchange. Coastal and marginal seas represent nutrient-rich areas with strong biological activity and are influenced by various anthropogenic factors. As the oceans take up a major part of the anthropogenic emissions of CO<sub>2</sub>, many oceanic regions are experiencing ongoing acidification. There are still major uncertainties in assessing the oceanic uptake of anthropogenic CO<sub>2</sub>: during 2005–2014 it was estimated at 2.6 GtC yr<sup>-1</sup>, an estimated 26 % of the total anthropogenic CO<sub>2</sub> emissions (Le Quéré et al., 2015). For a long time, the

lack of information on the coastal seas has barely been considered in the oceanic and global carbon budgets. The coastal ocean's role in terms of carbon export and relative productivity is disproportionately large in respect to its total surface area (7 %), when compared with the open ocean (Bourgeois et al., 2016). As the annual amplitude of air–sea  $p\text{CO}_2$  difference is significantly larger in coastal regions than open ocean (Rödenbeck et al., 2013), the variability in the exchange is high.

Various methods, both direct and indirect, are used to determine the air–sea flux of CO<sub>2</sub> (FCO<sub>2</sub>) (e.g. Smith et al., 1996; McGillis et al., 2001; Krasakopoulou et al., 2009). Both direct and indirect measures of FCO<sub>2</sub> were used in this study (McGillis et al., 2001; Rutgersson and Smedman, 2009; Gutiérrez-Loza and Ocampo-Torres, 2016).

Other studies have calculated FCO<sub>2</sub> across ocean basins using climate databases (Takahashi et al., 2002) or biogeochemical numerical models (Lenton et al., 2013; Arruda et al., 2015). These calculations, however, have failed to provide outputs covering the global coastlines. This is primarily due to the sparseness of the temporal and spatial data sets (such as *p*CO<sub>2</sub> of the surface ocean or wind fields). The wide range of values of in situ coastal FCO<sub>2</sub> entail even wider uncertainties in global estimates of FCO<sub>2</sub>, as there is the potential to under- or overestimate FCO<sub>2</sub> when performing a spatio-temporal integration (Wollast, 1991; Takahashi et al., 2009; Ribas-Ribas et al., 2011). A better comprehension of the local processes controlling FCO<sub>2</sub> along each coastal setting of continental margins will therefore lead to a better constrained set of global FCO<sub>2</sub> estimates. Since the year 2000, many different FCO<sub>2</sub> estimates and measurements have been reported for various near-shore, coastal, and inner-shelf environments. The question of which coastal seas can be a source or a sink remained open until recently: in the study of Chen et al. (2013) the coastal seas act as a sink with a mean value of air to sea flux of  $-1.09 \pm 2.9 \text{ mol C m}^{-2} \text{ yr}^{-1}$ . The study shows that most of the shelves absorb CO<sub>2</sub> from the atmosphere, except at the low latitudes where they act as a source ( $0.11 \text{ Pg C yr}^{-1}$ ) compared to high and temperate latitudes ( $-0.33 \text{ Pg C yr}^{-1}$ ). The study shows that the shelves in the Atlantic Ocean, which represent 33 % of the total absorption, have the highest total absorption. This corresponds to a mean air–sea CO<sub>2</sub> flux of  $-1.2 \text{ mol C m}^{-2} \text{ d}^{-1}$ . The spread of these values is a result of the heterogeneous and coupled biogeochemical processes in near-shore and coastal systems (Laruelle et al., 2010). It is necessary to increase our comprehension of the ocean carbon cycle and the air–sea exchange of CO<sub>2</sub> along the continental margins (Alin et al., 2012) because of their high social and ecological impact (Vargas et al., 2012).

High biological activity is the major cause of high CO<sub>2</sub> fluxes between the coastal and marginal seas. With this information, coastal seas may contribute disproportionately to the open ocean storage of CO<sub>2</sub> (Thomas et al., 2004) via a mechanism called the continental shelf pump (Tsunogai et al., 1999). In recent years, detailed field studies of CO<sub>2</sub> fluxes have taken place in a few areas, such as the East China Sea, northwest European Shelf, Baltic Sea, and North Sea (Chen and Wang, 1999; Thomas et al., 1999; Thomas and Schneider, 1999; Frankignoulle and Borges, 2001; Borges and Frankignoulle, 2002; Borges et al., 2003). However, only limited information about these CO<sub>2</sub> fluxes is available on a global scale (Liu et al., 2000b, a; Cai et al., 2003; Chen et al., 2003; Omstedt et al., 2009; Norman et al., 2013b).

The Baltic Sea is a semi-enclosed sea in northern Europe (Meier et al., 2014) that has been relatively well studied (e.g. Omstedt et al., 2004; Hjalmarsen et al., 2008; Backer and Leppänen, 2008; Wesslander, 2011) and monitored. It is characterized by an important upwelling variability (Norman et al., 2013a; Myrberg and Andrejev, 2003; Lehmann and

Myrberg, 2008; Sproson and Sahlée, 2014) and by important river runoffs (Bergstrom, 1994), which were estimated at  $17241.9 \text{ m}^3 \text{ s}^{-1}$  in 2015 (Johansson, 2017). Siegel and Gerth (2012) show that in the Baltic Sea decomposition of organic matter and biological production control the biogeochemical processes. The nutrient and carbon distribution in the water column, as well as light availability, is the limiting factor of these processes. In the Baltic Sea, the former factors are affected by physical constraints such as the stratification of the water, the salinity and temperature profiles, and the sea currents.

In recent years, the Baltic Sea has also been paid more attention to as a coastal system affecting both the uptake–release of anthropogenic CO<sub>2</sub> and the natural CO<sub>2</sub> cycle (Thomas and Schneider, 1999; Lansøet et al., 2015). Between 1994 and 2008 direct CO<sub>2</sub> measurements from cargo ships have been recorded, with a monthly resolution.

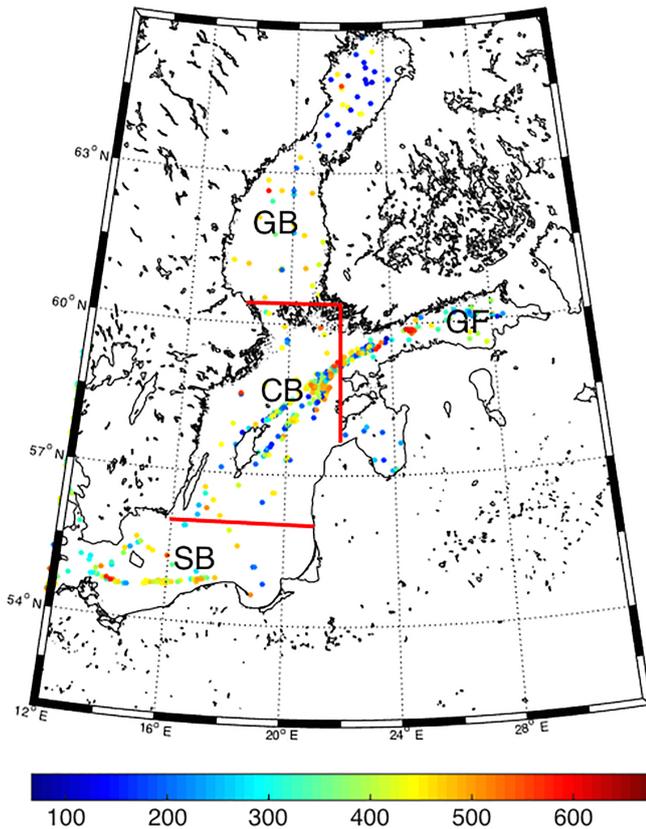
The net annual air–sea exchange of CO<sub>2</sub> in the central Baltic Sea and the Kattegat varied both regionally and inter-annually. Wesslander et al. (2010) show that the Kattegat was a sink of CO<sub>2</sub>, while the eastern Gotland and Bornholm seas were sources. They show that the interannual variability in the annual net flux is mainly controlled by the winter conditions. This is due to the CO<sub>2</sub>-enriched water mixes in winter in the central Baltic Sea. A second point is that the central Baltic Sea receives large amounts of organic material from river water inflow; this may give rise to a heterotrophic system, making the central Baltic a net CO<sub>2</sub> source. Conversely, the Kattegat is highly influenced by oceanic conditions.

The balance between mineralization and production and the depth of the mixed layer in the different oceanic zones examined were shown to be the main drivers of their respective sink–source distributions (Wesslander et al., 2010).

The goal of the present study is to develop an air–sea CO<sub>2</sub> flux estimation based on remote sensing products with a monthly time resolution and 4° spatial resolution and to estimate the error of this method of flux estimation in the Baltic Sea. In addition, we will further describe the processes and air–sea fluxes of CO<sub>2</sub> from 1998 to 2011 in the entire Baltic Sea and discuss the advantages and the limits of the method.

In this study, the air–sea CO<sub>2</sub> flux is estimated with the ocean surface *p*CO<sub>2</sub> in the Baltic Sea estimated from products derived from satellite data in Parard et al. (2015, 2016). The outputs of the method have a horizontal resolution of 4 km and cover the period from 1998 to 2011. Previous studies of the net uptake or release of CO<sub>2</sub> in the Baltic Sea have produced a wide range of results, with net exchange varying between  $-3.6$  and  $+2.9 \text{ mol CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$  in different time periods between 1994 and 2009 (Norman et al., 2013b).

The study is structured in four sections. Section 2 presents the data and method used in this work. Section 3 presents the wind products used to estimate the exchange (based on satellite data and reanalysis data). In Sect. 4, we analyse the wind products' quality, as well as various aspects of the estimated fluxes, and in Sect. 5 we present our conclusions.



**Figure 1.** Data available for the Baltic Sea for 1998–2011. The dashed red lines indicate the division into the Central Basin (CB), Gulf of Finland (GF), Gulf of Bothnia (GB), and South Basin (SB). The colour bar shows the  $p\text{CO}_2$  values ( $\mu\text{atm}$ ).

## 2 Data and method

### 2.1 $p\text{CO}_2$ map

We used self-organizing map classification along with class-specific linear regressions (SOMLO methodology) (Sasse et al., 2013) to reconstruct the sea surface  $p\text{CO}_2$  concentrations. The SOMLO methodology combines two statistical approaches: self-organizing maps (SOMs) (Kohonen, 1990) and linear regression.

SOMs are a subfamily of neural network algorithms used to perform multidimensional classification. During its training phase, the SOMLO methodology first uses SOMs to discretize a data set of explanatory parameters into classes and then locally learns a set of linear regression coefficients to infer the  $p\text{CO}_2$  for each class. When presented with a new vector of explanatory parameters, it first classifies it on the SOM map, then uses the calculated regression coefficients to estimate the  $p\text{CO}_2$ .

We divided the Baltic Sea into the four regions from Parard et al. (2016): the Gulf of Bothnia (GB), Gulf of Finland (GF), Central Basin (CB), and South Basin (SB) (Fig. 1).

We then trained the SOMLO methodology on the data belonging to each of these basins, reconstructing each point by combining the results obtained through each training, weighted by the distance from each point to the centre of each region.

The covariance of the explicative variables with the  $p\text{CO}_2$  was taken into account when attributing a data vector to a class, by means of a modified distance function. This allows for certain extreme parameter values to be more easily associated with the areas of the SOM in which the  $p\text{CO}_2$  is more correlated with these values.

In addition, we chose to perform a principal component analysis (Jolliffe, 2002) of the training data belonging to each class of each SOM. We kept the first four axes of the principal component analysis and taught the regression coefficients using the data projections on these four axes instead of performing a regression on all the parameters.

### 2.2 Wind products

In this study we used wind products to calculate the transfer velocity, based on a mesoscale reanalysis product. A reanalysis is a combination of measurements and a model in which the available data are assimilated into a high-quality numeric modelling system. The reanalysis used in this paper was provided by the Swedish Meteorological and Hydrological Institute (SMHI) with the High Resolution Limited Area Model (HIRLAM) geometry (22 km horizontal grid spacing and 60 levels in the vertical; the model top is at 10 hPa) (Soci et al., 2011). HIRLAM is downscaled and dynamically adapted to a higher resolution (5 km grid) with a simplified HIRLAM called the Dynamic Adaptation Model (DYNAM). The observations of 10 m winds assimilated into the system are from four databases: the Integrated Surface Database (ISD) Station History maintained by NOAA's National Centers for Environmental Information (NCEI), the Meteorological Archival and Retrieval System (MARS) at ECMWF, the European Climate Assessment & Dataset (ECA&D) used as input for E-OBS version 6.0, and the national climate databases of SMHI and Météo France (MF). The temporal resolution is 6 h. In the following, this product will be referred to as SMHIp. The method requires for the explicative data to stay coherent in terms of resolution, and as such we chose a temporal and spatial resolution of monthly  $4 \times 4$  km  $p\text{CO}_2$  pixels.

In order to estimate the impact of the wind product on the air–sea CO<sub>2</sub> flux, we computed the flux with a remote sensing product on a daily scale. The wind data are reprocessed QuikSCAT (QSCAT) and ASCAT data (Bentamy and Croizé-Fillon, 2013) with a spatial resolution of  $25 \times 25$  km. The data are available from 2000 to 2011.

### 2.3 Calculation of CO<sub>2</sub> flux

The flux of CO<sub>2</sub> (FCO<sub>2</sub>) from sea to air (positive value) or air to sea (negative value) is often calculated using the difference in the partial pressure of CO<sub>2</sub> between the surface water and the atmosphere ( $\Delta p\text{CO}_2$ ).

Here, the atmospheric  $p\text{CO}_2$  was estimated using the method from Rutgersson et al. (2009) and the sea surface  $p\text{CO}_2$  concentrations are reconstructed with the SOMLO methodology (Sasse et al., 2013), as performed by Parard et al. (2015, 2016).

In addition, the exchange efficiency was required, which was expressed in terms of a transfer velocity,  $k$ . The flux was then calculated according to

$$\text{FCO}_2 = k K_0 \Delta p\text{CO}_2, \quad (1)$$

where  $K_0$  is the salinity- and temperature-dependent solubility constant (Weiss et al., 1982). The gas transfer velocity was computed using the parameterization from Wanninkhof et al. (2009):

$$k = \sqrt{\frac{660}{Sc}} (3 + 0.1U + 0.064U^2 + 0.011U^3), \quad (2)$$

where  $U$  is the wind velocity at a reference height of 10 m and  $Sc$  is the solubility-dependent Schmidt number. Daily values of  $k$  were computed with a 6 h frequency for SMHIp; Eq. (2) is valid for all wind speed ranges. This method will be defined as Method 1.

We compare the results with another method to compute the transfer velocity  $k$  from Rutgersson and Smedman (2009):

$$k = 0.24 \cdot U^2 + (3022 \cdot w - 20), \quad (3)$$

where  $w$  is the water-side convection, which is estimated from the model used in Norman et al. (2013b). This method will be defined as Method 2.

## 3 Results

### 3.1 Analysis of the wind products

#### 3.1.1 Validation of the wind product

To validate our wind product, we compare the SMHI product with a product based on remote sensing data on a daily scale. Here called SATp, 10 m wind data are reprocessed QuikSCAT (QSCAT) and ASCAT data (Bentamy and Croizé-Fillon, 2013) with a spatial resolution of  $25 \times 25$  km. The two products are quite coherent when compared to all the station data used here, though SMHIp seems better, having a higher average correlation coefficient, i.e.  $R = 0.84$  versus 0.67 for the remote sensing data wind (we chose not to show here). This is to be expected, as SATp has a much coarser spatial resolution (25 km) than SMHIp does (5 km). In the

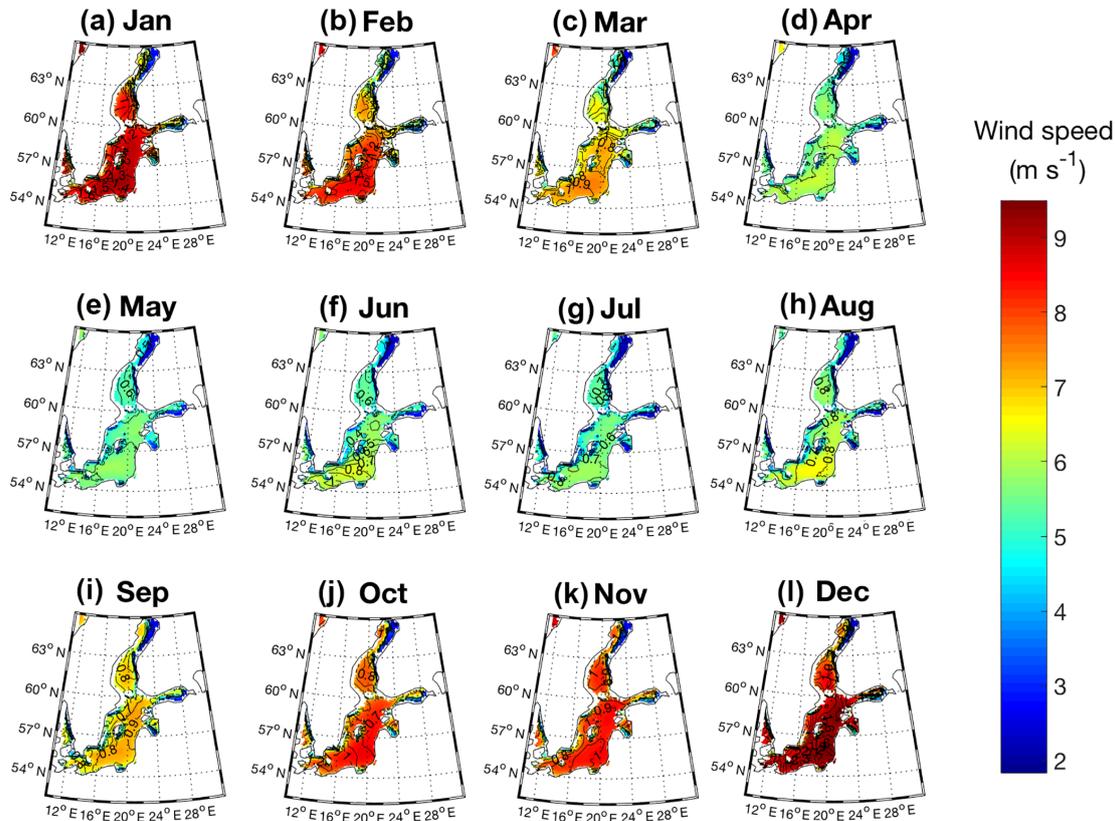
**Table 1.** RMS ( $\text{ms}^{-1}$ ), bias ( $\text{ms}^{-1}$ ), and correlation coefficients for in situ data from SMHI, the Östergarnsholm wind tower, and satellite products.

Tower	SMHIp		
	Bias	RMS	$R$
Total	0.67	2.49	0.84
Östergarnsholm	2.42	3.15	0.74
Falsterbo	1.70	2.27	0.86
Helsingborg	−0.88	1.65	0.85
Hanö	3.64	4.07	0.88
Ölands södra udde	0.62	1.70	0.86
Hoburg	−1.05	1.91	0.88
Nidingen A	3.68	4.17	0.85
Vinga	3.33	3.84	0.88
Ölands norra udde	−0.29	1.52	0.87
Visby	−1.88	2.56	0.87
Måseskär	3.82	4.29	0.91
Nordkoster	2.87	3.30	0.88
Harstena	−0.33	1.45	0.86
Landsort	1.73	2.41	0.83
Gotska Sandön	−1.60	2.20	0.91
Svenska Högarna	1.57	2.31	0.8
Örskär	1.07	2.02	0.86
Kuggören	−0.52	1.90	0.79
Brämön	0.29	1.86	0.78
Skagsudde	−0.37	1.78	0.79
Holmogadd	−0.60	1.85	0.82
Holmön	−0.75	2.13	0.78
Bjuröklubb	0.13	2.16	0.75
Luleå Airport	−2.32	3.17	0.68

following we decided to use the SMHI product to compute the transfer velocity.

The wind product SMHIp used here to compute the air–sea CO<sub>2</sub> flux was compared with wind tower data available from 24 stations in the Baltic Sea, including data from the Östergarnsholm measurement site Högström (2008); Rutgersson et al. (2008). Here, a micrometeorological tower, situated at 57.42° N, 18.99° E, has been running since 1995, making high-quality wind speed measurements at five heights. To validate the satellite data, we used measurements made 12 m above mean sea level in the 1995–2002 and 2005–2009 periods. In addition, we validated the winds using synoptic station data from SMHI for 21 sites along the coast of Sweden.

The wind product SMHIp values agree quite well with the station data (Table 1). Most of the synoptic stations are very close to the coast; thus, there might be a bias due to land influence. The correlation coefficient ( $R$ ) is quite high (0.66–0.91) and the high average correlation coefficient is  $R = 0.84$ . This is to be expected given that the spatial resolution is quite high for SMHIp (5 km).



**Figure 2.** Monthly mean wind speed (indicated by the colour bar) and annual variability (indicated by contours).

The root-mean-square differences (RMSDs) are given in Table 1. We increase the resolution of the wind products by means of linear interpolation to compute the air–sea CO<sub>2</sub> flux. This was done to provide coherency between our data sets.

### 3.1.2 Wind variability over the Baltic Sea

We examine the annual and monthly mean wind speeds and wind variability for the entire Baltic Sea (Fig. 2) for all 12 months during 13 years from 1998 to 2011. Figure 2 shows the wind speed in colours and the annual wind variability in contours on the seasonal timescale. The mean winds are higher in the CB than the GB, i.e. about  $7\text{--}7.4\text{ m s}^{-1}$  versus  $5\text{--}6\text{ m s}^{-1}$ . In terms of variability, the wind can vary by as much as  $1.5\text{--}2.1\text{ m s}^{-1}$  in the CB and  $1.4\text{--}1.9\text{ m s}^{-1}$  in the GB. On a monthly scale, high mean winds ( $8\text{--}9\text{ m s}^{-1}$ ) are seen in the Baltic Sea from November to February (Fig. 2). Of the four regions, the CB experiences the highest winds in the winter months. March and September are transition months with winds generally between 7 and  $8\text{ m s}^{-1}$ . May and June are the months when the winds are generally low,  $4\text{--}5\text{ m s}^{-1}$ . The largest variability in the winds, as represented by the contours (Fig. 2), is observable from September to December. The variability remains strong

from December to February ( $1.2\text{--}2.4\text{ m s}^{-1}$ ) in all the basins, while the lowest variability is observed in July ( $< 0.8\text{ m s}^{-1}$ ).

## 3.2 Air–sea CO<sub>2</sub> flux

### 3.2.1 Air–sea CO<sub>2</sub> flux estimation and variability

The air–sea CO<sub>2</sub> flux estimations are shown in Fig. 3, fluxes are computed using the SMHIp wind data, and figures represent the time period from 1998 to 2011. Figures 3 and 4 show the seasonal cycle. We observed the same patterns reflecting the surface  $p\text{CO}_2$  partial pressure (the air–sea difference in partial pressure) as previously seen in Parard et al. (2016). April to August show an uptake and October to February an outgassing. The interannual variability is slightly larger during the spring; this can indicate a large interannual variability in the onset of biological activities. Spatial differences are larger during the biologically active period. For example, in April the northern basins act as a source area, while the southern basins represent an uptake of the atmospheric CO<sub>2</sub>. Transfer velocity is largest in the South Basin and during winter following the wind speed pattern. In Fig. 4, the annual mean concentrations are shown. The flux displays high seasonal and spatial variability, ranging from  $-11$  to  $27\text{ mmol m}^{-2}\text{ d}^{-1}$ . On average, between 1998 and 2011, the entire Baltic Sea acts as a sink of  $-1.2\text{ mmol m}^{-2}\text{ d}^{-1}$

(Fig. 3). The values estimated from the remote sensing products are in agreement with those from other studies, indicating that the Baltic Sea can be a small source on average or a small sink of CO<sub>2</sub>. Most previous research results concerning the carbon budget cover shorter periods, indicating a range between  $-1.16$  and  $2.9 \text{ mol m}^{-2} \text{ yr}^{-1}$  (e.g. Wesslander, 2011; Kulinski and Pempkowiak, 2012), though the maximum values reported in these studies are all found in the same one or two years (Algesten et al., 2006). Half of the studies demonstrate that the Baltic Sea or certain basins of it act as sources, while the other studies demonstrate that it acts as a sink for the atmosphere (Norman et al., 2013a). In Chen et al. (2013), the Baltic Sea shows an air–sea CO<sub>2</sub> flux of  $-1.95 \text{ mol m}^{-2} \text{ yr}^{-1}$ , which is also in agreement with the results of our method.

The annual mean values for transfer velocity,  $p\text{CO}_2$ , and fluxes for these four regions are presented in Fig. 4.

During the entire study period, the four basins act, in general, as a source. The CB acts as a source, except for in four years: 2003, 2004, 2009, and 2010, with a lower value in 2009 ( $-0.8 \text{ mmol m}^{-2} \text{ d}^{-1}$ ). The GF acts as a source of the same order of magnitude as the CB with four years as a sink: 2005, 2007, 2008, and 2009, with a lower value in 2009 ( $-0.8 \text{ mmol m}^{-2} \text{ d}^{-1}$ ). The SB and the GB act as a source in all the years except 2010 with a low sink ( $-0.01 \text{ mmol m}^{-2} \text{ d}^{-1}$ ) and 2009 ( $-0.4 \text{ mmol m}^{-2} \text{ d}^{-1}$ ). The interannual variability is of the same order of magnitude for all the basins; however, the largest variability is seen in the GB, acting as a source until 2008 ( $> 1.7 \text{ mmol m}^{-2} \text{ d}^{-1}$ ) and a smaller source afterwards ( $< 0.8 \text{ mmol m}^{-2} \text{ d}^{-1}$ ). The seasonal cycle does not show different patterns for the different basins. The seasonal cycle is smaller for the northernmost basin (GB) (Fig. 3).

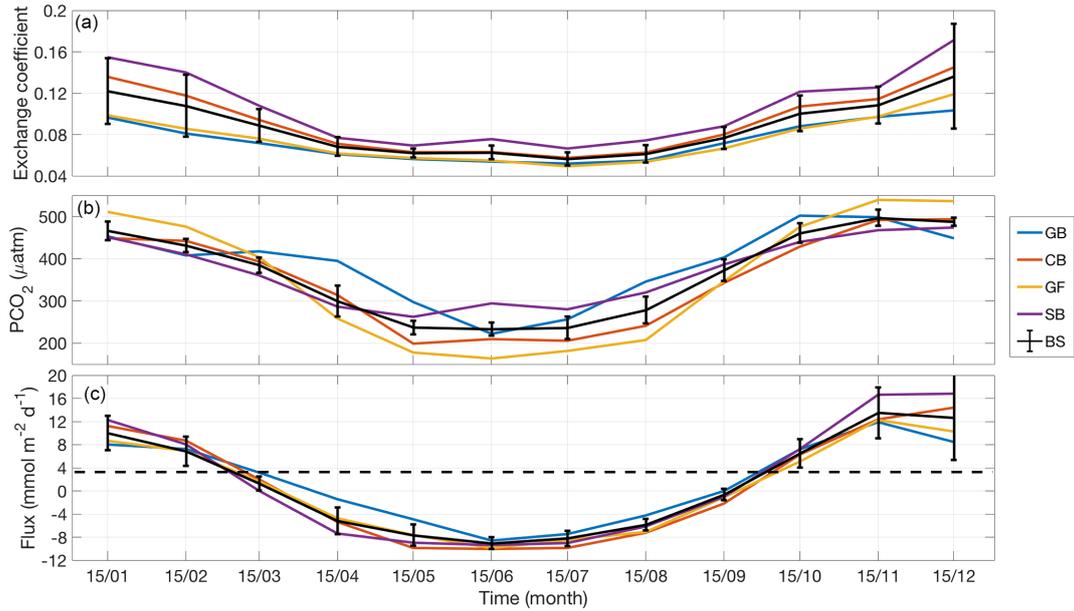
Between 1998 and 2011, the annual air–sea CO<sub>2</sub> flux in the Baltic Sea is always positive (Fig. 4), but we observed a higher flux before 2003 and after 2007. The four basins display a decrease in the flux from 1998 to 2011 (Fig. 4). The decrease is larger in the GB; after 2008 the value is half of the value before. A smaller decrease is observed in the GF. A decreasing trend can be explained by transfer velocity or  $p\text{CO}_2$ , but the decreasing pattern in the flux is not really reflected in the annual values of these parameters. The trend can also be explained by changes in the seasonal distribution of parameters. The seasonal cycle shows a shift in time when the first 5 years (1998 to 2002) are compared to the last 5 years (2007 to 2011) in Fig. 5. In all the basins the uptake is larger in April and May. For the later period, the differences are particularly large in the basins most influenced by ice cover (GB and GF). There is also an indication in the GB and GF for a reduced outgassing in early winter. As the data are not entirely homogeneous as is described in Parard et al. (2015) one should not draw conclusions too far from the suggested trend. The differences in the basins could, however, be related to the higher  $p\text{CO}_2$  concentrations in the atmosphere due to anthropogenic emissions; the corresponding increase

in CO<sub>2</sub> concentration in the atmosphere during this period is  $23.7 \mu\text{atm}$ . As the trend, to a large extent, is explained by an earlier onset of springtime, uptake differences in temperature and ice cover might be a more likely explanation.

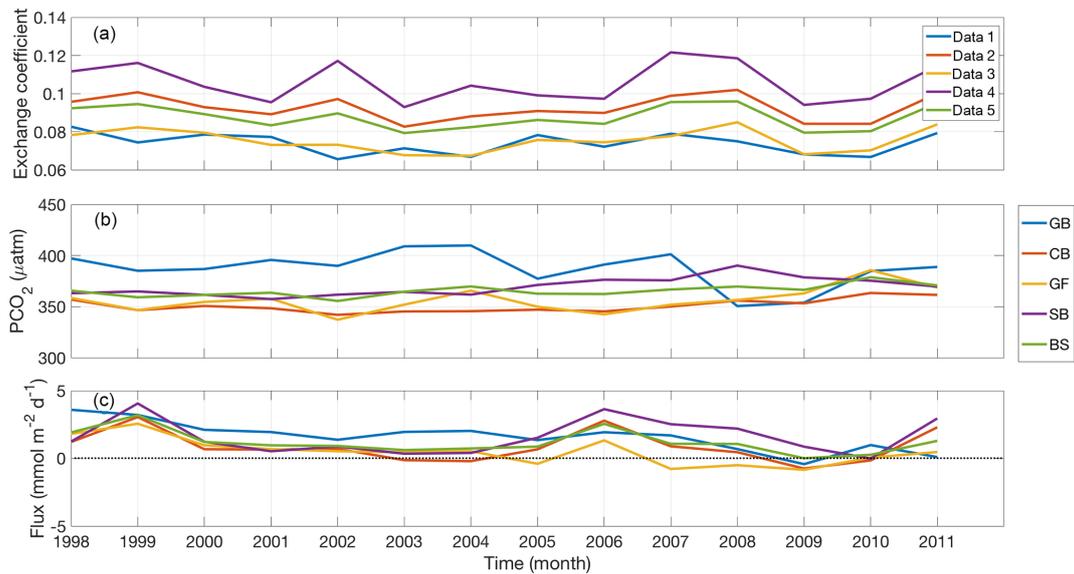
The coastal region is defined by a distance of  $0.5^\circ$  in latitude and longitude from the coast. Farther than  $0.5^\circ$  in latitude and longitude from the closest coast is defined as the open sea. The CO<sub>2</sub> flux computed in the coastal region is lower in winter and higher in summer than it is in the open sea (Fig. 6). The average difference in CO<sub>2</sub> flux is  $-0.5 \text{ mmol m}^{-2} \text{ d}^{-1}$ , with a variability of between  $-5.5$  and  $2.5 \text{ mmol m}^{-2} \text{ d}^{-1}$ . The higher difference ( $-1.6 \text{ mmol m}^{-2} \text{ d}^{-1}$ ) is observed in 2007, with a lower value for the coastal region. The air–sea CO<sub>2</sub> fluxes are lower for the entire year in the coastal region. Annually, there are three periods when we observe a greater difference, i.e. February–March, June–July, and October (Fig. 6). Biological activity is one explanation for the lower air–sea CO<sub>2</sub> in the coastal region in March–April and October compared to the open ocean region. The biological activity is higher along the coast at these times (Schneider, 2011) due to upwelling near the coast (Omstedt et al., 2009; Norman et al., 2013a); this has the effect of reducing the CO<sub>2</sub> emitted to the atmosphere. In the coastal region we observed a change in the sink between the first 5 years between 1998 and 2002 and the last 5 years between 2007 and 2011 (Fig. 7). The lower air–sea CO<sub>2</sub> flux is observed during the last years and the minimum of the air–sea CO<sub>2</sub> flux is in April and May. It correlates with the observations in Fig. 5. The sink increases in April from  $-2.9 \text{ mmol m}^{-2} \text{ d}^{-1}$  and in May from  $-1.8 \text{ mmol m}^{-2} \text{ d}^{-1}$ . The monthly difference is small compared with that observed on the seasonal scale, though we may be underestimating the effect of the upwelling on the monthly scale. A review of Baltic Sea upwelling (Lehmann and Myrberg, 2008) demonstrates that the typical upwelling lasts from several days to 1 month on a horizontal scale of 10–20 km offshore. It is therefore possible that the effect of the upwelling may be underestimated.

### 3.2.2 Uncertainty analysis

The method used to compute the  $p\text{CO}_2$  has the advantage of computing a monthly map of  $p\text{CO}_2$  for the entire Baltic Sea on a monthly scale from 1998 to 2011 from the data set of in situ data present in Fig. 1. As explained in Parard et al. (2016) for the reconstructed  $p\text{CO}_2$  values, the correlation coefficient ( $R$ ) values are good, with the lowest values observed in the SB (0.9) where the RMS is the highest (i.e.  $38.5 \mu\text{atm}$ ). The GF has the highest  $R$  value (i.e. 0.97) and the GB has the lowest RMS ( $19.5 \mu\text{atm}$ ), the latter being the region with the lowest data density. This error has an impact on the air–sea CO<sub>2</sub> flux computation. The impact of the maximum RMS on the flux is  $\pm 4 \text{ mmol m}^{-2} \text{ d}^{-1}$ . It gives a high influence of the air–sea CO<sub>2</sub> flux and our uncertainty of the air  $p\text{CO}_2$  increases this uncertainty.



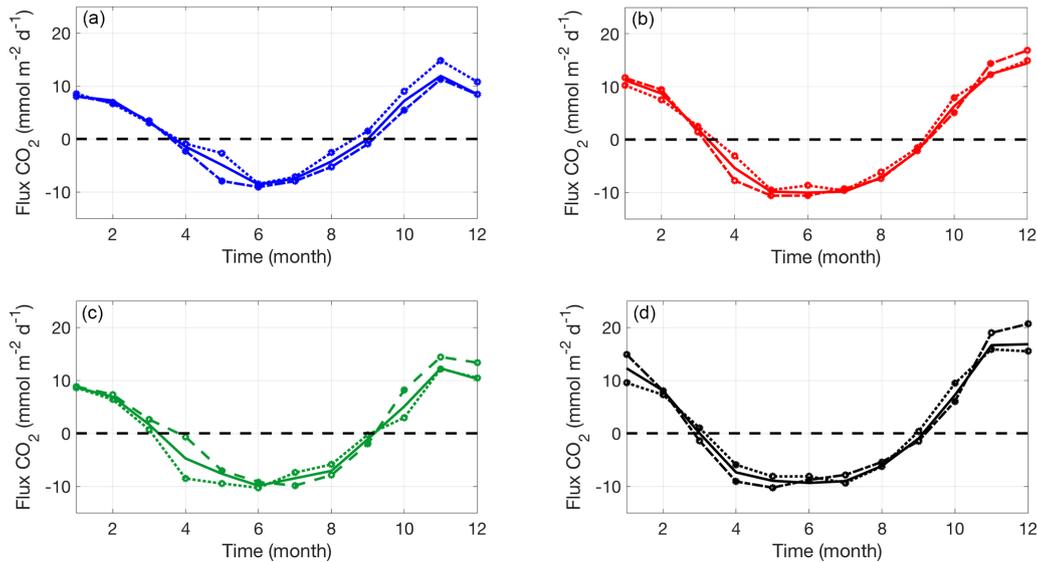
**Figure 3.** Annual evolution of the (a) transfer velocity based on Wanninkhof et al. (2009). (b)  $PCO_2$  and (c) air–sea  $CO_2$  flux based on the SMHIP wind product for each basin.



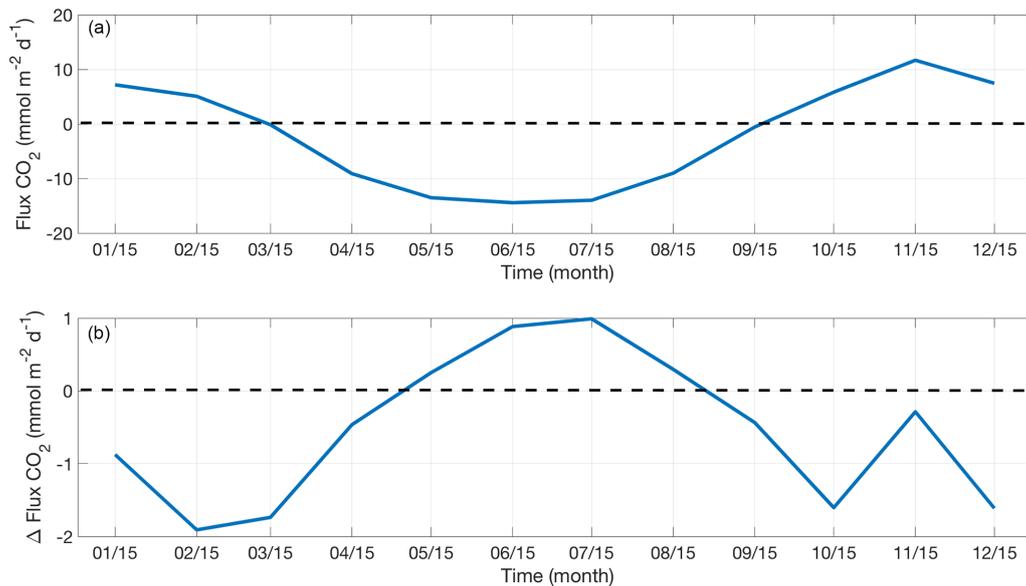
**Figure 4.** Annual evolution of the (a) transfer velocity based on Wanninkhof et al. (2009). (b)  $PCO_2$  and (c) air–sea  $CO_2$  flux based on the SMHIP wind product for each basin. In the legend: GB: Gulf of Bothnia; CB: Central Basin; GF: Gulf of Finland; SB: South Basin; and BS: Baltic Sea.

The difference between the phase before 2003 and the phase after 2007 could be explained by the repartition of the data used to calculate our results. In order to understand if this repartition of the initial data is responsible for the phase difference, we studied the representation of the data along the different years for each neuron of the SOM maps in each basin (Fig. 8). For the three first basins (Fig. 8a, b, c), all the years are present, at least in part, even if some classes

seem to be solely composed of data measured before 2002, in particular in the southern regions (the blue trend colour classes). In the north of the GB there are no data before 2008; thus, the results that we show can be affected by this lack of data. However, the results are coherent with the other basins. The distribution of the data is well spread (Fig. 8e, f, g, h) throughout the classes.



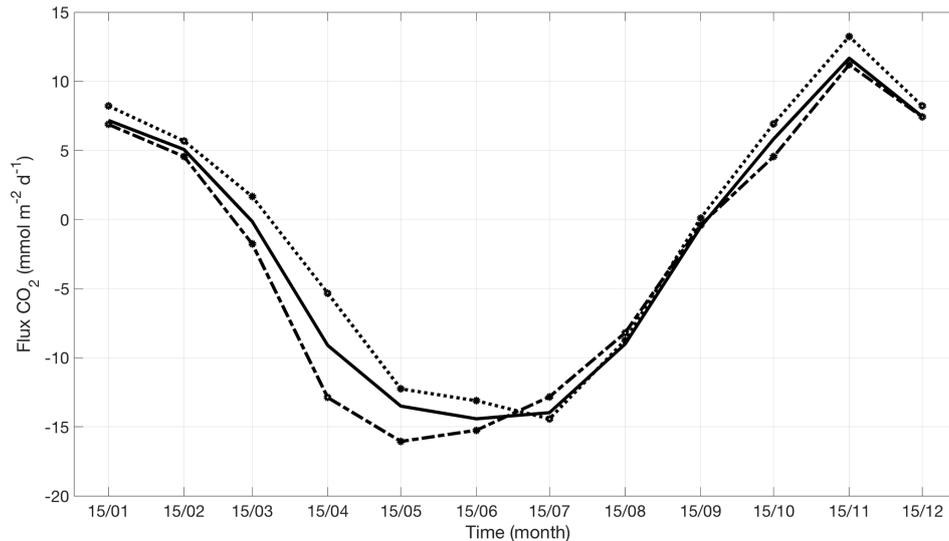
**Figure 5.** Seasonal cycle of air–sea CO<sub>2</sub> flux for the (a) Gulf of Bothnia, (b) Central Basin, (c) Gulf of Finland, and (d) South Basin. Solid lines represent the average for the full period (1998 to 2011), dotted lines with markers are for the first 5 years (1998–2002), and dashed lines are for the last 5 years (2007 to 2011). In the legend: GB: Gulf of Bothnia; CB: Central Basin; GF: Gulf of Finland; SB: South Basin; and BS: Baltic Sea.



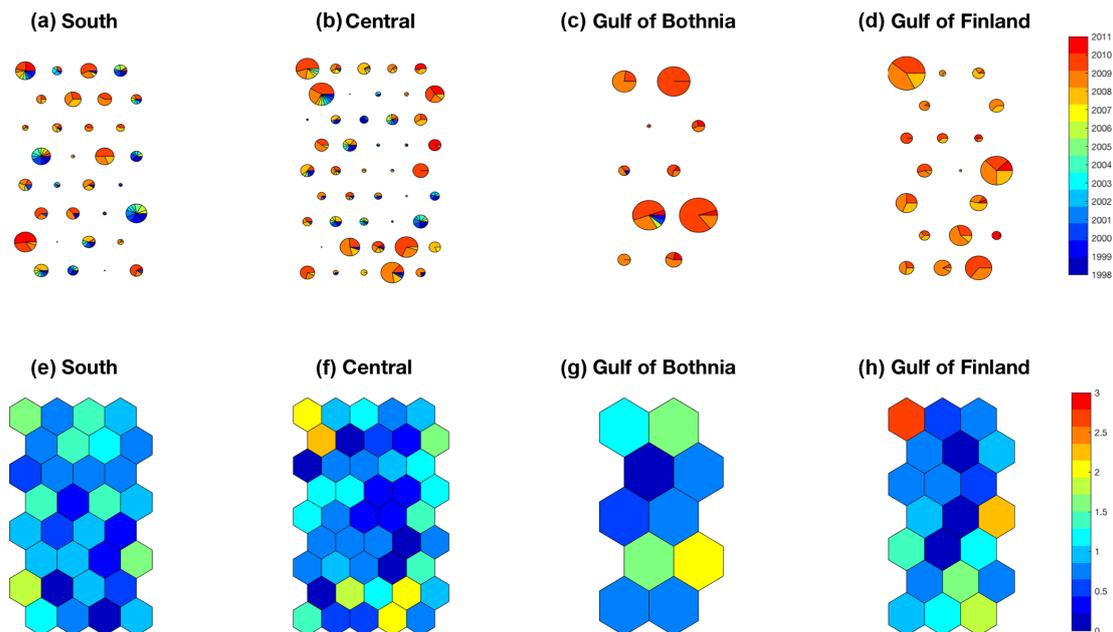
**Figure 6.** Average (a) of the air–sea CO<sub>2</sub> flux and (b) of the difference between the coastal region and open sea from 1998 to 2011.

Two tests were performed in order to estimate the error on the air–sea CO<sub>2</sub> flux. One with the SATp wind product and one with the air–sea flux estimation method Rutgersson et al. (2009) describes in Eq. (3). These results are presented in Fig. 9. The two air–sea CO<sub>2</sub> flux estimations are computed using the two sets of wind data, the SMHIp and SATp data sets. The CO<sub>2</sub> flux computed using SMHIp wind data is available from 1998 to 2011 and using SATp wind data from 2000 to 2011. We compared the two products from 2000 and 2011 (not shown here). The two flux estimations from

the wind product have the same order of magnitude. Nevertheless, the seasonal cycle from the air–sea CO<sub>2</sub> flux using SATp product is larger, with a lower value in summer and a higher value in winter. We observe the maximum difference in January (when the flux using SMHIp winds is higher) and in September (when the flux using SATp winds is higher). The monthly variability in the flux using SMHIp winds is 8.7–11.4 mmol m<sup>-2</sup> d<sup>-1</sup> versus 3.4–13.4 mmol m<sup>-2</sup> d<sup>-1</sup> using SATp winds. High variability in January using the SATp wind product can be explained by the lack of satellite data



**Figure 7.** Seasonal cycle of air–sea CO<sub>2</sub> flux for the Baltic Sea. The solid line represents the average for the full period (1998–2011). The dotted line with a marker is for the first 5 years (1998–2002) and the dashed line is for the last 5 years (2007 to 2011).

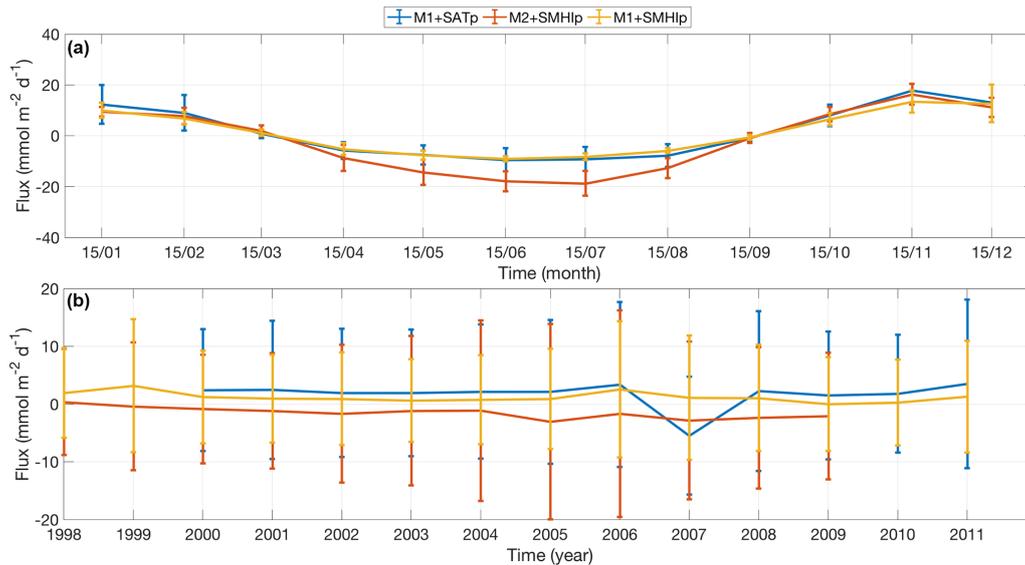


**Figure 8.** Panels (a), (b), (c), and (d) are the distributions of the years of each data in each class for each basin SOM. Panels (e), (f), (g), and (h) are the percentages of the total number of data present in each class of the different basins' SOM. The size of the circles in the top figures is also representative of the percentage of the total number of data present in each class of the different basins' SOM.

for this month. In addition, there are also interannual variations. In most years, the Baltic Sea acts as a sink. Using the SMHIp winds, the exchange ranges from  $-2.9$  to  $0.6 \text{ mmol m}^{-2} \text{ d}^{-1}$  with an average of  $-1.6 \text{ mmol m}^{-2} \text{ d}^{-1}$ ; using the SATp winds, the annual uptake is larger, being between  $-3.9$  and  $0.3 \text{ mmol m}^{-2} \text{ d}^{-1}$  with an average for 2000–2011 of  $-2.1 \text{ mmol m}^{-2} \text{ d}^{-1}$ . The trend is the same for both products, with a decrease in the flux and an increase in the absorption of  $p\text{CO}_2$  from the atmosphere. The average

difference between the wind from satellite and the SMHI wind products gives a value of  $0.98 \text{ m s}^{-2}$  and has an influence of  $0.34 \text{ mmol m}^{-2} \text{ d}^{-1}$  on the air–sea CO<sub>2</sub> flux. Our method to recompute the  $p\text{CO}_2$  gives a root mean square between 19.5 and  $38.5 \text{ } \mu\text{atm}$ , which depends on the basin; this has an effect on the air–sea CO<sub>2</sub> flux of  $-1.2 \text{ mmol m}^{-2} \text{ d}^{-1}$ .

Two methods to compute the air–sea CO<sub>2</sub> flux have been used: one from Wanninkhof et al. (2009) in which the results are described above, the second from Rutgersson



**Figure 9.** The air–sea CO<sub>2</sub> flux estimate evolution with Method 1 and the SATp product (blue); Method 2 and the SMHIp product (red); and Method 1 and the SMHIp product (yellow). (a) For a year and (b) on average for all the years.

et al. (2009). The second one used the water-side convection from a model (Norman, 2013). The mean difference between the two products is  $1.2 \text{ mmol m}^{-2} \text{ d}^{-1}$ . The higher differences are observed in 1999 ( $3.2 \text{ mmol m}^{-2} \text{ d}^{-1}$ ) and in 2006 ( $2.6 \text{ mmol m}^{-2} \text{ d}^{-1}$ ). The two methods to compute coefficient exchange give a difference on the air–sea CO<sub>2</sub> flux of 0.088. On a seasonal scale the differences of the two methods are higher in spring and summer (April to August) and range between  $4 \text{ mmol m}^{-2} \text{ d}^{-1}$  in April and  $10 \text{ mmol m}^{-2} \text{ d}^{-1}$  in June. In winter, the difference is between 0.2 and  $2.0 \text{ mmol m}^{-2} \text{ d}^{-1}$ .

To conclude, the  $p\text{CO}_2$  uncertainty gives a high variability in the air–sea CO<sub>2</sub> flux, the wind products influence the value more than the variability, and the difference is quite similar in all the time series. The method influences the variability and it does not influence all the time series in the same way.

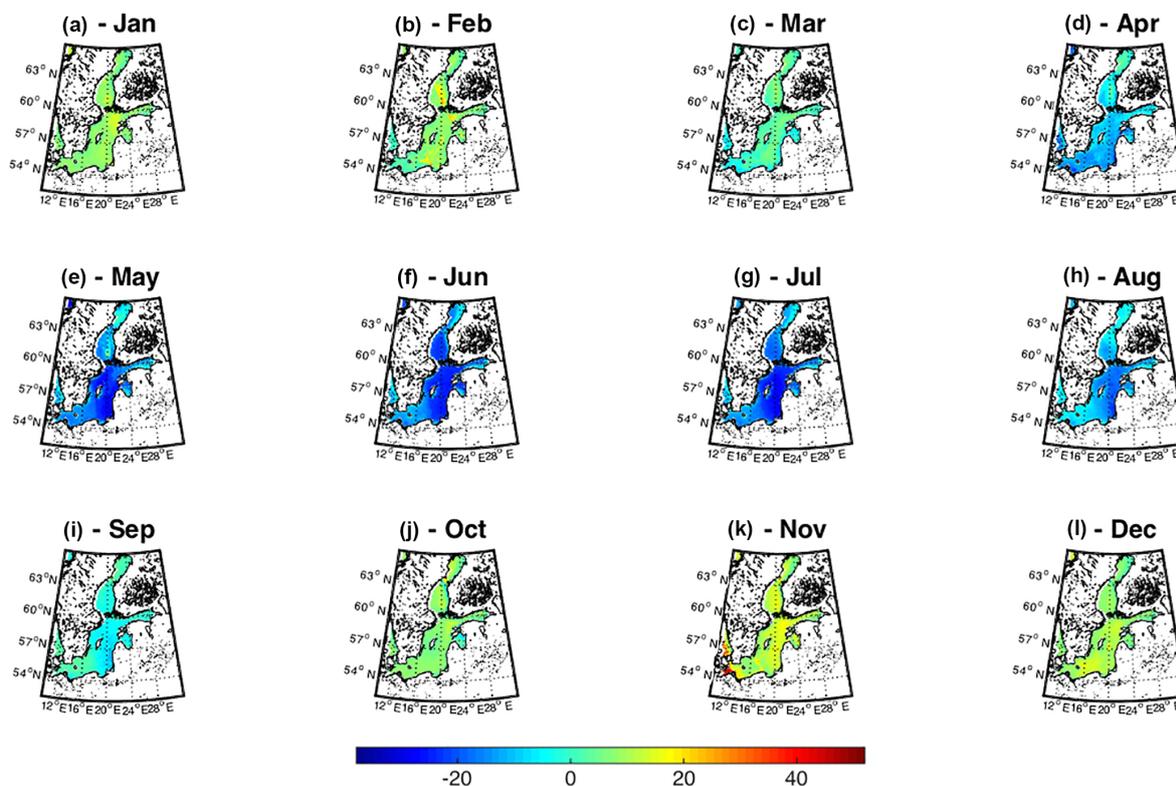
### 3.2.3 Air–sea CO<sub>2</sub> flux climatology

The climatology of the flux displays high seasonal and spatial variability, ranging from  $-13$  to  $10 \text{ mmol m}^{-2} \text{ d}^{-1}$ . On average, from 1998 to 2011, the entire Baltic Sea acted as a source of  $1.2 \text{ mmol m}^{-2} \text{ d}^{-1}$ . The results are different if we use the method from Rutgersson et al. (2009), which resulted in  $1.4 \text{ mmol m}^{-2} \text{ yr}^{-1}$  and a sink of  $-1.5 \text{ mmol m}^{-2} \text{ yr}^{-1}$  if we used the SATp winds (Fig. 10). The values observed are in agreement with those from other studies, indicating that the Baltic Sea can be, on average, a small source or a small sink of CO<sub>2</sub>. Most previous research results concerning the carbon budget of the Baltic Sea cover shorter periods, indicating a range between  $-1.16$  and  $2.9 \text{ mol m}^{-2} \text{ yr}^{-1}$  (e.g. Wesslander et al., 2010; Kulinski and Pempkowiak, 2012), though the maximum values reported in these studies are all found in

the same one or two years (Algesten et al., 2006). Half of the studies demonstrate that the Baltic Sea or certain basins of it act as sources, while the other studies demonstrate that the Baltic Sea acts as a sink for the atmosphere (Norman et al., 2013a).

## 4 Discussion and conclusions

Canadell (2003) explains that it is really challenging to precisely estimate the variation in the  $p\text{CO}_2$  in marginal seas. This is due to several aspects but mainly due to temporal and spatial sparsity of measurements. Remote sensing using applicable algorithms could certainly be an important approach, complementing shipboard observations as well as in situ buoy and wind tower measurements. Using our method, we present the first estimated CO<sub>2</sub> flux climatology based on remote sensing for the Baltic Sea. This gives an estimated annual mean air–sea CO<sub>2</sub> flux of  $1.2 \pm 0.8 \text{ mmol m}^{-2} \text{ d}^{-1}$  and a seasonal variability of between  $-13$  and  $10 \text{ mmol m}^{-2} \text{ d}^{-1}$ . The interannual variability is 1 order of magnitude lower, being between 0.01 and  $3.19 \text{ mmol m}^{-2} \text{ d}^{-1}$ . Several studies have estimated the air–sea CO<sub>2</sub> fluxes in the Baltic Sea over the last decade; most of these examine specific regions, but only a few cover the entire Baltic Sea. Kulinski and Pempkowiak (2012) demonstrate that the Baltic Sea was a source of CO<sub>2</sub> for the atmosphere between 2002 and 2008, but they use data from several time periods and sources. Using a biogeochemical model covering the 1960–2009 period, Norman et al. (2013b) suggest that the entire Baltic Sea acts as a net sink of between  $-0.22$  and  $-0.17 \text{ mol m}^{-2} \text{ yr}^{-1}$ , in agreement with our value of  $-0.02 \text{ mol m}^{-2} \text{ yr}^{-1}$ .



**Figure 10.** Temporal evolution of the air–sea CO<sub>2</sub> flux between 1998 and 2011 based on SMHIP data.

In the GF, we found the lowest source of CO<sub>2</sub> from the atmosphere ( $0.2 \text{ mol m}^{-2} \text{ yr}^{-1}$ ), which ranges between  $-0.3$  and  $0.9 \text{ mol m}^{-2} \text{ yr}^{-1}$ . These lowest values are observed in 2005 and from 2007 to 2009: during this period the GF is actually a sink for the atmosphere. The GB is a sink in 2009 in our study, but this value decreases from 1998 to 2009. This flux has a value of  $0.5 \text{ mol m}^{-2} \text{ yr}^{-1}$  in 2002, lower than the value of  $2.9 \text{ mol m}^{-2} \text{ yr}^{-1}$  from Algesten et al. (2006). This estimation is based on a few days of measurements from a few stations in the GB. Our results indicating a small source are in agreement with those of the study demonstrating a larger sink in the Bothnian Sea ( $-0.73 \text{ mol m}^{-2} \text{ yr}^{-1}$ ) and a smaller source in Bothnian Bay ( $0.14 \text{ mol m}^{-2} \text{ yr}^{-1}$ ) between 1999 and 2009; this finding could explain why the entire GB region acts as a small sink or a small source, on average.

In the CB, Schneider et al. (2014) demonstrate that in four selected years (i.e. 2003, 2004, 2009, and 2010), the surface water acts as a sink for the atmosphere, as found in our study, with the value of the uptake rates ranging between  $-0.04$  and  $-0.3 \text{ mol m}^{-2} \text{ yr}^{-1}$ . The rate explained the enhanced carbon in the sediments (Schneider et al., 2014). Our study of 2005, 2008, and 2009 finds an uptake value between  $-0.9$  and  $-1.0 \text{ mol m}^{-2} \text{ yr}^{-1}$ , slightly higher than that reported in Schneider et al. (2014), who use boat-line data. This could be because of the spatial resolution of our product, which includes the entire CB. Our mean value for the CB indicates

that it is a sink for the atmosphere. This is in contrast to the findings of Wesslander et al. (2010), who demonstrate that, for a slightly different period (i.e. 1994 to 2008), the CB acts as a source of  $1.64 \text{ mol m}^{-2} \text{ yr}^{-1}$  for the atmosphere. As we explain in Parard et al. (2014), the  $p\text{CO}_2$  data do not reproduce the spring–summer bloom in the eastern Gotland Sea described in Schneider et al. (2015). The data used for the computation contain the voluntary observing ships (VOSs) ship line, but we calculated a monthly average. Thus, we missed some higher-frequency processes. In their study, Schneider et al. (2015) explain that the spring bloom takes place around 12 February and 21 March (5 weeks); thus, the average must smooth the variability due to the bloom. In order to improve the  $p\text{CO}_2$  data set, it is better to use the daily data in order to better reproduce such processes.

To conclude, the first approximation using remote sensing data and in situ  $p\text{CO}_2$  data to compute the  $\text{FCO}_2$  gives good spatial and temporal resolutions compared with those of measurements from ships or wind towers. Indeed, the satellite data give information on  $p\text{CO}_2$  variability and on  $\text{FCO}_2$ . The in situ data set of  $p\text{CO}_2$  in the Baltic Sea is used to construct an entire map of the Baltic Sea in space and time with SOMLO methodology. SOMLO was used to accommodate the non-linearity of the mechanics driving the  $p\text{CO}_2$ . It uses artificial neural networks to classify data into situations and then performs a reconstruction by using a multiple linear regression (MLR) in each class. The process involves clas-

sifying the explicative parameters (i.e. sea surface temperature, coloured dissolved organic matter (CDOM), chlorophyll, time, net primary production, and mixed layer depth)

The first estimates of Baltic Sea air–sea exchange based on remote sensing products display reasonably good agreement with previous estimates and indicate a negative trend, with annual uptake changing from 0.6 to  $-2.8 \text{ mol m}^{-2} \text{ yr}^{-1}$  over the 1998–2007 period. After 2007, the decrease is smaller and the flux remains quite stable at around  $-2.8 \text{ mol m}^{-2} \text{ yr}^{-1}$ . The air–sea CO<sub>2</sub> flux product depends on the wind product and on the  $p\text{CO}_2$  product, but also on the water convection. For winds, the higher-resolution product gives larger flux amplitudes, and for  $p\text{CO}_2$ , chlorophyll and CDOM are essential inputs.

The air–sea CO<sub>2</sub> flux is sensitive to different parameters (wind product,  $p\text{CO}_2$ , exchange coefficient). The wind products impact differently in the Baltic Sea and the northern Baltic Sea. In the Gulf of Bothnia, the wind plays affect the inter-annual variation in air–sea CO<sub>2</sub> flux, which is higher than in the other basins. On average, the Central Basin near the South Basin is the region with the highest uptake of CO<sub>2</sub>. The coastal region has a slightly higher uptake than the open sea region.

Several parameters would be useful to improve our product as more in situ data constrain our computation more. However, other parameters such as sea surface salinity, which has a strong variability in the Baltic Sea and a higher frequency, should be used in order to better represent the different processes and better estimate the air–sea CO<sub>2</sub> flux.

**Data availability.** The air–sea flux CO<sub>2</sub> estimation will be available at <https://ecds.se/dataset/remote-sensing-data-to-estimate-pco2-and-air-sea-co2-exchange>.

**Competing interests.** The authors declare that they have no conflict of interest.

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