

The Potential of using Remote Sensing data to estimate Air-Sea CO₂ exchange in the Baltic Sea.

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Abstract.

In this article, we present the first climatological map of air-sea CO₂ flux over the Baltic Sea, based on remote-sensing data: satellite imaging derived estimates of pCO₂ using self-organizing maps classifications along with class-specific linear regressions (SOMLO methodology) and remote-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 CO₂ 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 fluxes estimations.

Furthermore, the CO₂ 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 pCO₂ unvaryingly over the whole Baltic Sea, with high winter CO₂ emissions and high pCO₂ 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⁻² d⁻¹ for the South Basin and 0.9 for the Central Basin) than in the northern regions (mean source of 0.1 mmol m⁻² d⁻¹) and the coastal areas act as a larger sink (annual uptake of -4.2 mmol m⁻² d⁻¹) than does the open sea (-4 mmol m⁻² d⁻¹). In its entirety, the Baltic Sea acts as a small source of 1.2 mmol m⁻² d⁻¹ on average and this annual uptake has increased from 1998 to 2012.

15 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 (CO₂) plays a key role in controlling Earth's climate. The oceanic uptake of anthropogenic CO₂ helps regulate atmospheric CO₂ through air-sea exchange. Coastal and marginal seas represent nutrient-rich areas with strong biological activity and are influenced by various anthropogenic factors.

20 As the oceans take up a major part of the anthropogenic emissions of CO₂, many oceanic regions are experiencing ongoing acidification. There are still major uncertainties in assessing the oceanic uptake of anthropogenic CO₂: during 2005–2014 it was estimated to 2.6 GtC yr⁻¹, an estimated 26% of the total anthropogenic CO₂ emissions (Le Quéré et al., 2015). During long time, the lack of information on the coastal seas, have 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 pCO₂ difference is significantly larger in coastal regions than open ocean (Rödenbeck et al., 2013), the variability of the exchange is high.

Various methods, both direct and indirect, are used to determine the air-sea flux of CO₂ (FCO₂) (e.g. Smith et al., 1996; 5 McGillis et al., 2001; Krasakopoulou et al., 2009). Both direct and indirect measures of FCO₂ were used in this study (McGillis et al., 2001; Rutgersson and Smedman, 2009; Gutiérrez-Loza and Ocampo-Torres, 2016).

Other studies have calculated FCO₂ 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 pCO₂ of 10 the surface ocean or wind fields). The wide range of values of in situ coastal FCO₂ entails even wider uncertainties in global estimates of FCO₂, as there is the potential to under- or overestimate FCO₂ 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 FCO₂ along each coastal setting of continental margins will therefore lead to a better constrained set of global FCO₂ estimates.

Since the year 2000, many different FCO₂ estimates and measurements have been reported for various near-shore, coastal, and 15 inner-shelf environments. The question about the coastal seas which can be a source or a sink remained open until recently, in Chen et al. (2013) the coastal sea act as a sink with a mean value of air to sea flux is $-1.09 \pm 2.9 \text{ mol C m}^{-2} \text{ yr}^{-1}$. The study shows that most of the shelves absorb CO₂ from the atmosphere except at the low latitudes where they act as a source (0.11 Pg C yr⁻¹) compare to high and temperate latitude ($-0.33 \text{ pG C yr}^{-1}$). The study shows that the shelves in the Atlantic Ocean have the highest total absorption, which represent 33% of the total of the absorption which correspond to a mean air sea CO₂ 20 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 CO₂ along the continental margins (Alin et al., 2012), due to their high social and ecological impact (Vargas et al., 2012).

The high biological activity is the major cause of high CO₂ fluxes between the coastal and marginal seas. With this information, 25 coastal seas may contribute disproportionately to the open-ocean storage of CO₂ (Thomas et al., 2004) via a mechanism called the continental shelf pump (Tsunogai et al., 1999). In recent years, detailed field studies of CO₂ fluxes take place in a few areas, such as the East China Sea, North-west 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 is available on a global scale about these CO₂ fluxes (Liu et al., 2000a, b; Cai et al., 30 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) which has been relatively well studied (e.g. Omstedt et al., 2004; Hjalmarsson 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 are in 2015 estimated at $17241.9 \text{ m}^3 \text{ s}^{-1}$ (Johansson, 2017). In the Baltic Sea, (Siegel and Gerth, 2012) shows that 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 are the limiting factors 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 as well as the sea currents.

In recent years, the Baltic Sea has also been paid more attention as a coastal system affecting both the uptake/release of 5 anthropogenic CO₂ and the natural CO₂ cycle (Thomas and Schneider, 1999; Lansøet al., 2015). Between 1994 and 2008 direct CO₂ measurements from a cargo ship has been recorded, with a monthly resolution.

The net annual air-sea exchange of CO₂ in the central Baltic Sea and the Kattegat varied both regionally and inter-annually. In Wesslander et al. (2010), show that the Kattegat sea was, a sink of CO₂ while the East 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 10 is due to the CO₂-enriched water mixes in winter in 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 CO₂ source. In opposite in the Kattegat, is highly influenced by oceanic conditions.

The balance between mineralization and production, as well as 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)

15 The goal of the present study is to develop an air-sea CO₂ 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 CO₂ from 1998 to 2011 in the entire Baltic Sea and discuss the advantage and the limit of the method.

In this study, the air sea CO₂ flux is estimated, with the ocean-surface pCO₂ in the Baltic Sea estimate from satellite-data 20 derived products 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 CO₂ in the Baltic Sea have produced a wide range of results, with net exchange varying between -3.6 and +2.9 mol CO₂ m⁻² y⁻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 25 wind products used to estimate the exchange (based on satellite data and reanalysis data). In Section 4, we analyze the wind products' quality, as well as various aspects of the estimated fluxes , and in Section 5 we present our conclusions.

2 Data and method

2.1 pCO₂ map

We used the SOMLO methodology (Sasse et al., 2013), to reconstruct the sea-surface pCO₂ concentrations. The SOMLO 30 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 dataset of explanatory parameters into classes and then locally

learns a set of linear regression coefficients to infer the pCO₂ 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 pCO₂.

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

5 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 center of each region.

The covariance of the explicative variables with the pCO₂ 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 where the pCO₂ is more correlated with these values.

10 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 meso-scale reanalysis product. A reanalysis is 15 a combination of measurements and a model in which the available data are assimilated into a high-quality numeric modeling 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 20 assimilated into the system are from four databases: the Integrated Surface Database Station History (ISH) database maintained by NOAA's National Climatic Data Center (NCDC), the MARS archive 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 of 6 hours. 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 x 25 4 km pCO₂ pixels.

In order to estimate the impact of the wind product on the air-sea CO₂ flux, we computed the flux with a remote sensing product at daily scale. The wind data are reprocessed QuikSCAT (QSCAT) and ASCAT data (Bentamy and Croizé-Fillon, 2013) with a spatial resolution of 25x 25 km. The data are available from 2000 to 2011.

2.3 Calculation of CO₂ flux

30 The flux of CO₂ (FCO₂) from sea to air (positive value) or air to sea (negative value) is often calculated using the difference in the partial pressure of CO₂ between the surface water and the atmosphere ($\Delta p\text{CO}_2$).

Here, the atmospheric pCO₂ was estimated using the method from Rutgersson et al. (2009) and the sea-surface pCO₂ concentrations are reconstructed with the SOMLO methodology (Sasse et al., 2013), as done 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:

$$FCO_2 = kK_0\Delta pCO_2 \quad (1)$$

where K_0 is the salinity- and temperature-dependent solubility constant (Weiss et al., 1982). The gas transfer velocity was 5 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 define as Method 1.

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

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

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

3 Results

15 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 one based on remote-sensing data at daily scale 10 m wind data are reprocessed QuikSCAT (QSCAT) and ASCAT data (Bentamy and Croizé-Fillon, 2013) with a spatial resolution of 25x 25 km here called SATp. The two products are quite coherent when compared to all the station data used here, though 20 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 following we decided to used the SMHI product to compute the transfer velocity.

The wind product SMHIp used here to compute the air-sea CO_2 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. 25 (2008). Here, a micro-meteorological 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 agree quite well with the station data (Table 1). Most of the synoptic stations are very close to the coast, so 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).

5 The root-mean-square differences (RMSDs) is given in Table 1.

We increase the resolution of the wind products by means of linear interpolation to compute the air–sea CO_2 flux. This was done to provide coherency between our datasets.

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 (Figs. 2) for the twelve 10 months during 13 years from 1998 to 2011. Fig. 2 shows the wind speed in colors and the annual wind variability in contours at the seasonal time scale. The mean winds are higher in the Central Basin (CB) than the Gulf of Bothnia (GB), i.e. about 7–7.4 m s^{-1} versus 5–6 m s^{-1} . In terms of variability, the wind can vary by as much as 1.5–2.1 m s^{-1} in both CB and 1.4–1.9 m s^{-1} in GB. On the monthly scale, high mean winds (8–9 m s^{-1}) are seen in the Baltic Sea from November to February (Fig. 2). Of 15 the four regions, CB experiences the highest winds in winter months. March and September are transition months with winds generally between 7 and 8 m s^{-1} . May and June are the months when the winds are generally low, 4–5 m s^{-1} . The largest variability in the winds, as represented by the contours (Figure 2), is observable from September to December. The variability remains strong from December to February (1.2–2.4 m s^{-1}) in all the basins, while the lowest variability is observed in July (< 0.8 m s^{-1}).

3.2 Air–sea CO_2 flux

20 3.2.1 Air-sea CO_2 flux estimation and variability

The air–sea CO_2 flux estimations are shown in Figure 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 25 surface pCO_2 partial pressure (the air-sea difference in partial pressure) previously seen in (Parard et al., 2016). April to August represents an uptake and October to February an outgassing. The interannual variability is slightly larger during the the biologically active period. For example, in April the northern basins act as a source area while the southern basins represent 30 an uptake of the atmospheric CO_2 . Transfer velocity is largest in the southern basin and during winter following the wind-speed pattern. In Figure 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 to 2011, the entire Baltic Sea acts as a sink of $-1.2 \text{ mmol m}^{-2} \text{ d}^{-1}$ (Figure 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_2 . Most previous research results concerning the carbon budget cover shorter periods, indicating a range between -1.16 and $2.9 \text{ mol m}^{-2} \text{ y}^{-1}$ (Wesslander, 2011; Kulinski and

Pempkowiak, 2012, e.g.), 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 Baltic Sea or certain basins of it act as sources, while the others demonstrate that it acts as a sink for the atmosphere (Norman et al., 2013a). In (Chen et al., 2013), the Baltic Sea show a air-sea CO_2 flux of $-1.95 \text{ mol m}^{-2} \text{ yr}^{-1}$ which is also in agreement with the result of our method.

5 The annual mean values for transfer velocity, pCO_2 and fluxes for these four regions are presented in Fig 4.

During all the study period, the four basin act in general as a source. The Central Basin acts as a source except for 4 years 2003, 2004, 2009 and 2010 with a lower value in 2009($-0.8 \text{ mmol m}^{-2} \text{ d}^{-1}$). The Gulf of Finland acts as a source of the same order of magnitude as the Central Basin with 4 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 South Basin and the Gulf of Bothnia act as a source in all the years except respectively 2010 with a low 10 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 the same order of magnitude for all the basins however the largest variability is seen in the Gulf of Bothnia, 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) (Figure 3).

Between 1998 and 2011, the annual air-sea CO_2 flux in the Baltic Sea is always positive (Figure 4) but we observed higher 15 flux before 2003 and after 2007. The four basins display a decrease in the flux from 1998 to 2011 (Figure 4). The decrease is larger in the Gulf of Bothnia, after 2008 the value are half of the value before. A smaller decrease is observed in the Gulf of Finland. A decreasing trend can be explained by transfer velocity or pCO_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 seasonal distribution 20 of parameters. The seasonal cycle shows a shift in time when the first five years (1998 to 2002) are compared to the last five years (2007 to 2011) in Figure 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 GB and GF for a 25 reduced outgassing in early winter. As the data is not entirely homogeneous as it describes in Parard et al. (2015) one should not draw too far conclusions from the suggested trend. It could, however, be related to the higher pCO_2 concentrations in the atmosphere due to anthropogenic emissions, the corresponding increase in CO_2 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 spring-time "uptake" differences in temperature and ice cover might be a more likely explanation.

The coastal region is defined by a distance of 0.5° in latitude and longitude from the coast. Farther than 0.5° in latitude and longitude from the closest coast is defined as the open sea. The CO_2 flux computes 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_2 flux is $-0.5 \text{ mmol m}^{-2} \text{ d}^{-1}$ with a 30 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 coastal region. The air-sea CO_2 fluxes are lower for all the 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). The biological activity is one of the explanation of the lower air-sea CO_2 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 35 (Omstedt et al., 2009; Norman et al., 2013a); this has the effect of reducing the CO_2 emitted to the atmosphere. In the coastal

region we observed a change in the sink between the first five years between 1998 and 2002 and the last five years between 2007 and 2011 (Figure 7), The lower air-sea CO₂ flux is observed during the last years and the the minimum of the air-sea CO₂ flux is in April and May. It correlates with the observation in the Figure 5. The sink increase in April from -2.9 mmol m⁻² d⁻¹ and in May from - 1.8 mmol m⁻² d⁻¹. The monthly difference is small compared with that observed at the seasonal scale, though we may be underestimating the effect of the upwelling at the monthly scale. A review of Baltic Sea upwelling (Lehmann and Myrberg, 2008) demonstrates that the typical upwelling lasts from several days to one month at 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 pCO₂ has an advantage to compute a monthly map pCO₂ for the entire Baltic Sea at monthly scale from 1998 to 2011 from a data set of in situ data present in figure 1. As it explained in (Parard et al., 2016) for the reconstructed pCO₂ values. The correlation coefficient (*R*) values are good, the lowest being observed in the Southern Basin (0.9) where the RMS is the highest (i.e., 38.5 μ atm). The Gulf of Finland has the highest *R* value (i.e., 0.97) and the Gulf of Bothnia the lowest RMS (19.5 μ atm), the latter being the region with the lowest data density. This error has an impact on the air-sea CO₂ flux computation. The impact of the maximum RMS on the flux is \pm 4 mmol m⁻² d⁻¹. It gives a high influence of the air-sea CO₂ flux and our incertitude on the air pCO₂ increase this incertitude.

The difference between the phase before 2003 and 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 (Figure 8). For the three first basins (Figure 8,a.,b.,c.), all the years are present at least in part, even if some classes seem to be solely composed from data measured before 2002, in particular in the Southern regions (the blue trend color classes). In the North of the Gulf of Bothnia there is no data before 2008 so the results that we show can be affected by this lack of data, yet are coherent with the other basins. The distribution of the data is well spread (Figure 8,e.,f.,g.,h.) throughout the classes.

Two tests were performed in order to estimate the error on the air-sea CO₂ flux. One with SATp wind product and one with the air-sea flux estimations method Rutgersson et al. (2009) describes in eq. 3. These results are presented in Figure 9. The two air-sea CO₂ flux estimations are computed using the two sets of wind data, the SMHIp and SATp datasets. The CO₂ 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 shows here). the two flux estimations from the wind product have the same order of magnitude. Nevertheless, the seasonal cycle from air-sea CO₂ flux using SATp product is larger, with lower value in summer and higher 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 of the flux using SMHIp winds is 8.7-11.4 mmol m⁻² d⁻¹ versus 3.4-13.4 mmol m⁻² d⁻¹ using SATp winds. High variability in January using the SATp wind product can be explained by the lack of satellite data during 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 mmol m⁻² d⁻¹ with an average of -1.6 mmol m⁻² d⁻¹; using the SATp winds, the annual uptake is larger, being between -3.9 and 0.3 mmol m⁻² 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 pCO_2 from the atmosphere. The average difference between the wind from satellite and the SMHI wind products give a value of 0.98 m s^{-2} and have an influence of $0.34 \text{ mmol m}^{-2} \text{ d}^{-1}$ on the air-sea CO_2 flux. Our method to recompute the pCO_2 gives a root mean square between 19.5 and $38.5 \mu\text{atm}$, which depend on the basin, this has an
5 effect on the air-sea CO_2 flux of $-1.2 \text{ mmol m}^{-2} \text{ d}^{-1}$.

The two methods to compute the air-sea CO_2 flux have been used, one from (Wanninkhof et al., 2009) where the results are described above, the second from Rutgersson 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 difference is 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 method to compute coefficient exchange give a
10 difference on the air-sea CO_2 flux of 0.088. At seasonal scale the difference on the two methods are higher in spring and summer (April to August) 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 pCO_2 incertitude gives a high variability in the air-sea CO_2 flux, the wind products influence the value more than the variability, and the difference is quite similar in all the time serie. The method influence the variability and it
15 does not influence all the time serie in the same way.

3.2.3 Air-sea CO_2 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 acts as a source of $1.2 \text{ mmol m}^{-2} \text{ d}^{-1}$. The result are different if we used the method from Rutgersson et al. (2009) which give $1.4 \text{ mmol m}^{-2} \text{ y}^{-1}$ and give a sink if we used the SATp winds -1.5 mmol m^{-2}
20 y^{-1} (Fig. 10). The values observed 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_2 . Most previous research results concerning the carbon budget of the Baltic Sea cover shorter periods, indicating a range between -1.16 and $2.9 \text{ mol m}^{-2} \text{ y}^{-1}$ (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
25 et al. (2006). Half of the studies demonstrate that Baltic Sea or certain basins of it act as sources, while the others demonstrate that it acts as a sink for the atmosphere (Norman et al., 2013a).

4 Discussion and Conclusions

Canadell (2003) explain that it is really challenging to estimate precisely the variation of the pCO_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 ship-board observations as well as in situ buoy and wind-tower measurements. Using our method, we present the first estimated CO_2 flux climatology based on remote sensing for the
30 Baltic Sea. This gives an estimated annual mean air-sea CO_2 flux of $1.2 \pm 0.8 \text{ mmol m}^{-2} \text{ d}^{-1}$ and a seasonal variability of between $-13.$ to $10 \text{ mmol m}^{-2} \text{ d}^{-1}$. The interannual variability is one order of magnitude lower, being between 0.01 and 3.19

mmol m⁻² d⁻¹. Several studies have estimated the air-sea CO₂ fluxes in the Baltic Sea over the last decade; most of these examine specific regions, but only a few treat the entire Baltic Sea. Kulinski and Pempkowiak (2012) demonstrate that the Baltic Sea was a source of CO₂ 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
5 Sea acts as a net sink of between −0.22 and −0.17 mol m⁻² yr⁻¹, in agreement with our value of −0.02 mol m⁻² yr⁻¹.

In the Gulf of Finland, we found the lowest source of CO₂ from the atmosphere (0.2 mol m⁻² yr⁻¹), which ranges between −0.3 to 0.9 mol m⁻² yr⁻¹. These lowest value are observed in 2005 and 2007 to 2009: during this period it is actually a sink for the atmosphere. The gulf of Bothnia is a sink in 2009 in our study but this value decreases from 1998 to 2009. This flux has a value of 0.5 mmol m⁻² yr⁻¹ in 2002, lower than the value of 2.9 mol m⁻² yr⁻¹ from Algesten et al. (Algesten et al., 2006).
10 This estimation is based on a few days of measurements from a few stations in the Gulf of Bothnia. Our results indicating a small source are in agreement with those of the study demonstrating a larger sink in the Bothnian Sea (−0.73 mol m⁻² yr⁻¹) and a smaller source in Bothnian Bay (0.14 mol m⁻² yr⁻¹) between 1999 and 2009; this finding could explain why the entire Gulf of Bothnia region acts as a small sink or a small source on average.

In the Central Basin, Schneider et al. (2014) demonstrate that in four selected years (i.e. 2003, 2004, 2009, and 2010), the
15 surface water acts as a sink for the atmosphere, as found in our study, the value of the uptake rates ranging between −0.04 and −0.3 mol m⁻² yr⁻¹, the rate explained the enhance 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 mol m⁻² yr⁻¹, slightly higher than that reported Schneider et al. (2014), who use boat-line data. This could be because of the spatial resolution of our product, which includes the entire Central
20 Basin. Our mean value for the Central Basin 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 Central Basin acts as a source for the atmosphere of 1.64 mol m⁻² yr⁻¹. As we explain in Parard et al. (2014), the pCO₂ data set obtain 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 VOS ship line but we made monthly average so we missed some higher frequency processes. In the study, they explain that the spring bloom takes place around February 12 and March 21 (5 weeks), so the average must smooth
25 the variability due to the bloom. In order to improve the pCO₂ data set, it will be better to use the daily data in order to better reproduce such processes.

To conclude, in first approximation used remote sensing data and in-situ pCO₂ data to compute the FCO₂ gives good spatial and temporal resolutions compared with those of measurements from ships or wind-towers. Indeed the satellite data gives information on pCO₂ variability and on FCO₂. The in-situ data set of pCO₂ in the Baltic Sea are used to construct entire
30 map in Baltic Sea in space and time, with SOMLO methodology. SOMLO was used to accommodate the nonlinearity of the mechanics driving the pCO₂. It uses artificial neural networks to classify data into situations, and then performs a reconstruction by using an MLR in each class. The process involves classifying the explicative parameters (i.e., SST, CDOM, Chl, time, NPP, and MLD)

The first estimates of Baltic Sea air-sea exchange based on remote-sensing products display reasonably good agreement
35 with previous estimates and indicate a negative trend, with annual uptake changing from 0.6 to −2.8 mol m⁻² 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 CO₂ flux product depends on the wind product and on the pCO₂ product but also on the water convection. For winds, the higher-resolution product gives larger flux amplitudes, and for pCO₂, chlorophyll and CDOM are essential inputs.

The air-sea CO₂ flux is sensitive to different parameters (wind product, pCO₂, exchange coefficient). The wind products 5 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 CO₂ 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 CO₂. The coastal region has a slightly higher uptake than does the open-sea region.

Several parameters would be useful to improve our product as more in-situ data constrains more our computation, but also used other parameters as the sea surface salinity which has a strong variability in the Baltic Sea and a higher frequency in order 10 to better represent the different processes to better estimate the air-sea CO₂ flux.

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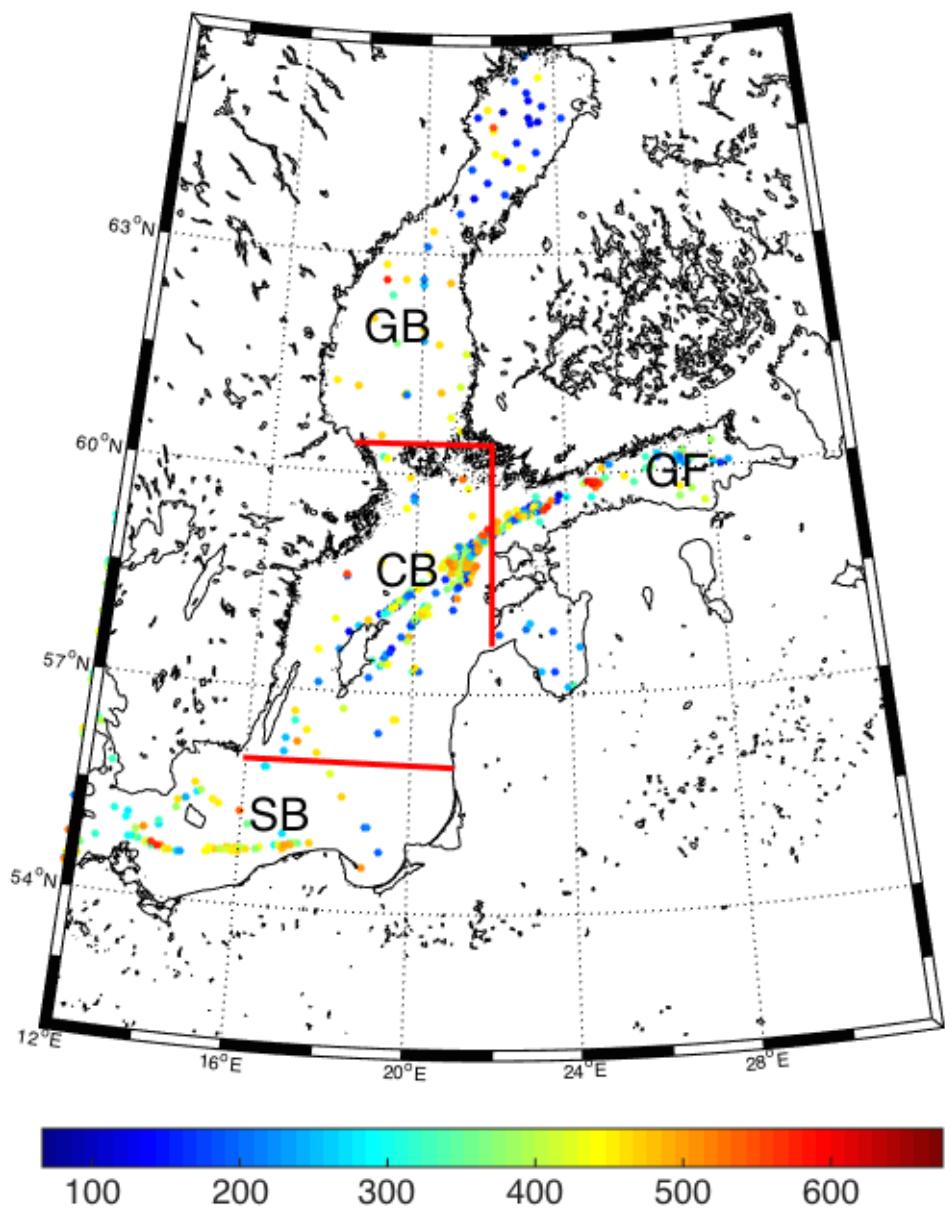


Figure 1. Data available for the Baltic Sea, 1998–2011. The dashed red lines indicate the division into the Central Basin (CB), Gulf of Finland (GF), Gulf of Bothnia (GB), and South Basin (SB). The colorbar shows the pCO₂ values in μatm .

Table 1. RMS (ms^{-1}), bias (ms^{-1}), and correlation coefficients for in situ data from SMHI, 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
ÖLAND SÖDRA	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
ÖLAND NORRA	-0.29	1.52	0.87
VISBY	-1.88	2.56	0.87
MASESKAR	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	-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
LULEÅAIRPORT	-2.32	3.17	0.68

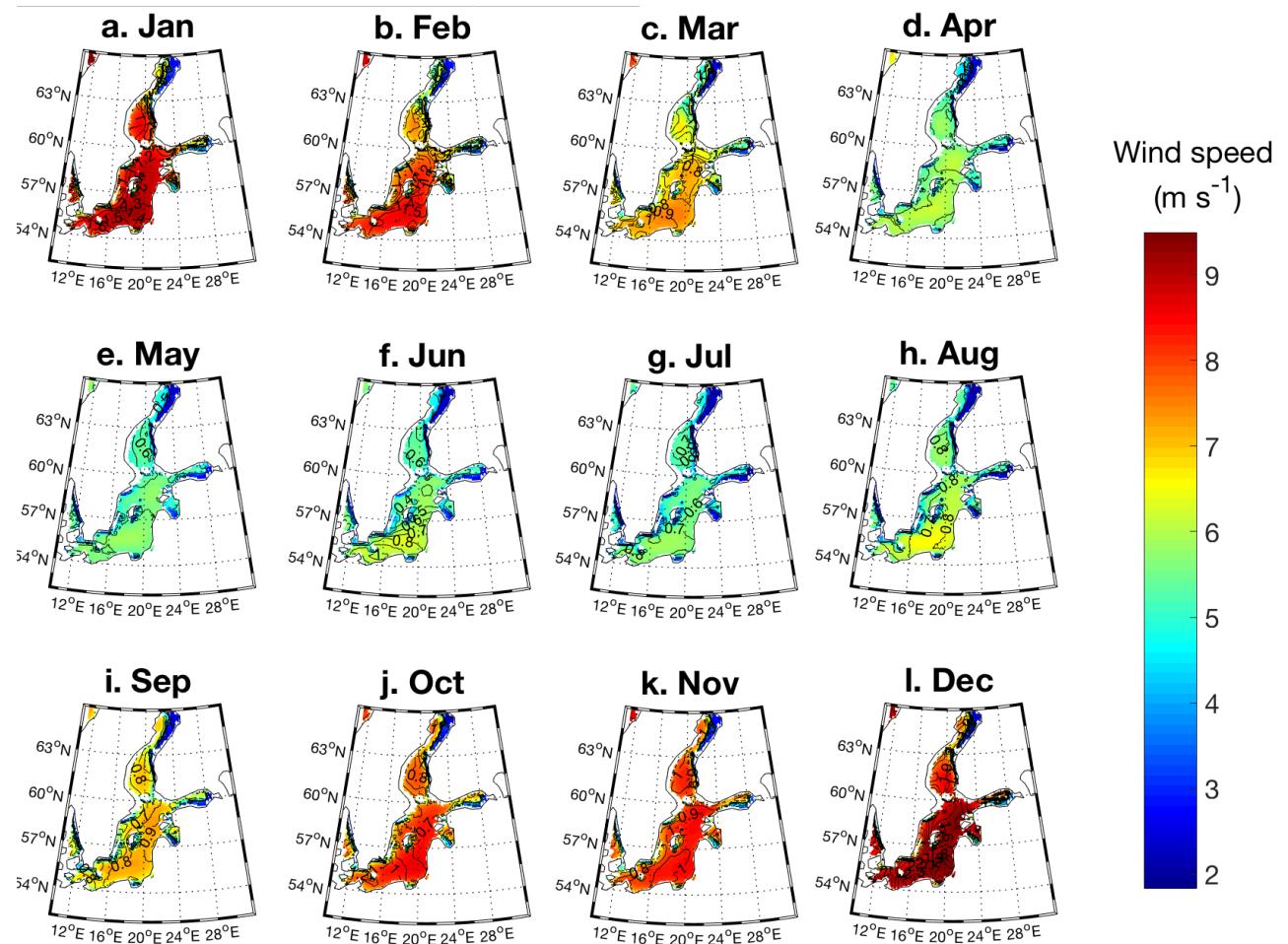


Figure 2. Monthly mean wind speed (indicated by colour bar) and annual variability (indicated by contours).

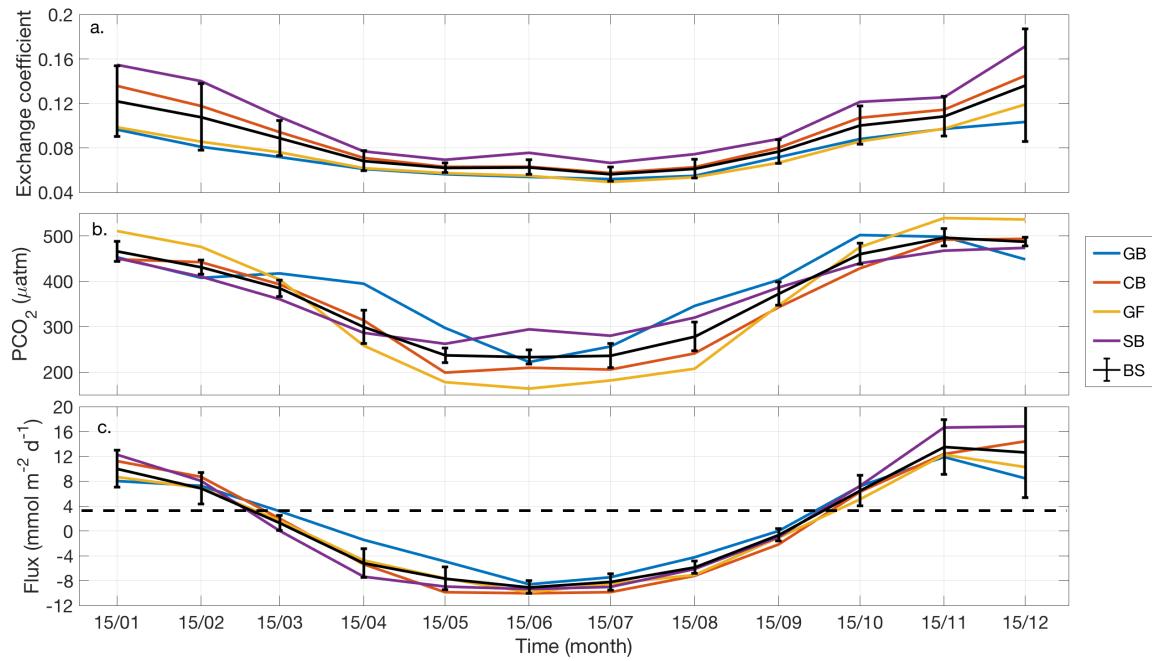


Figure 3. Evolution annual of the a.) transfer velocity based on Wanninkhof et al. (2009). b.) PCO₂ and c.) air-sea CO₂ flux based on the SMHIp wind product for each basin.

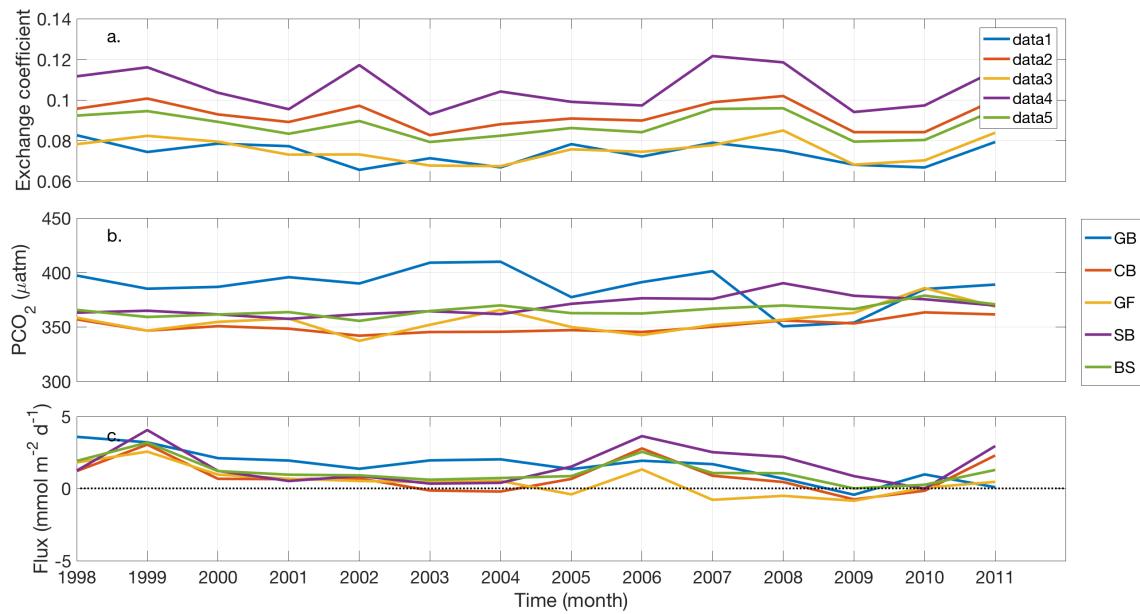


Figure 4. Evolution annual of the a.) transfer velocity based on Wanninkhof et al. (2009). b.) PCO₂ and c.) air-sea CO₂ flux based on the SMHIp wind product for each basin. In the legend : GB : Gulf of Bothnia, CB : Central Basin; GF Gulf of Finland; SB : South Basin and BS: Baltic Sea.

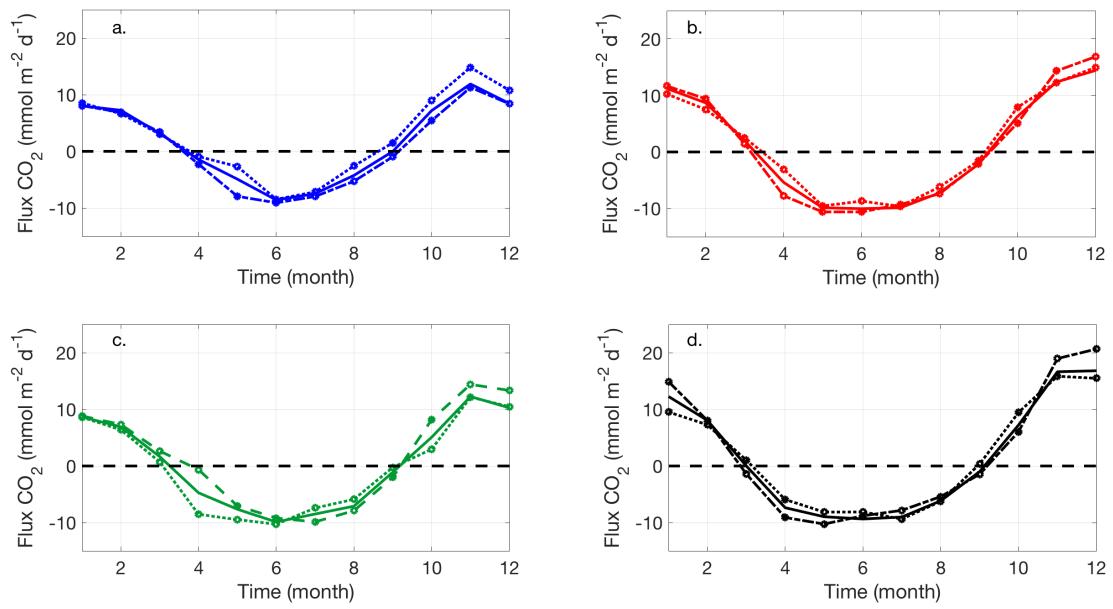


Figure 5. Seasonal cycle of air-sea CO_2 flux for a) Gulf of Bothnia, b) Central Baltic c) Gulf of Finland and d) Southern Baltic. Solid lines represent the average for the full period (1998 to 2011), dotted lines with markers are for the first 5 years (1998-2002) and dashed lines are for the last five 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.

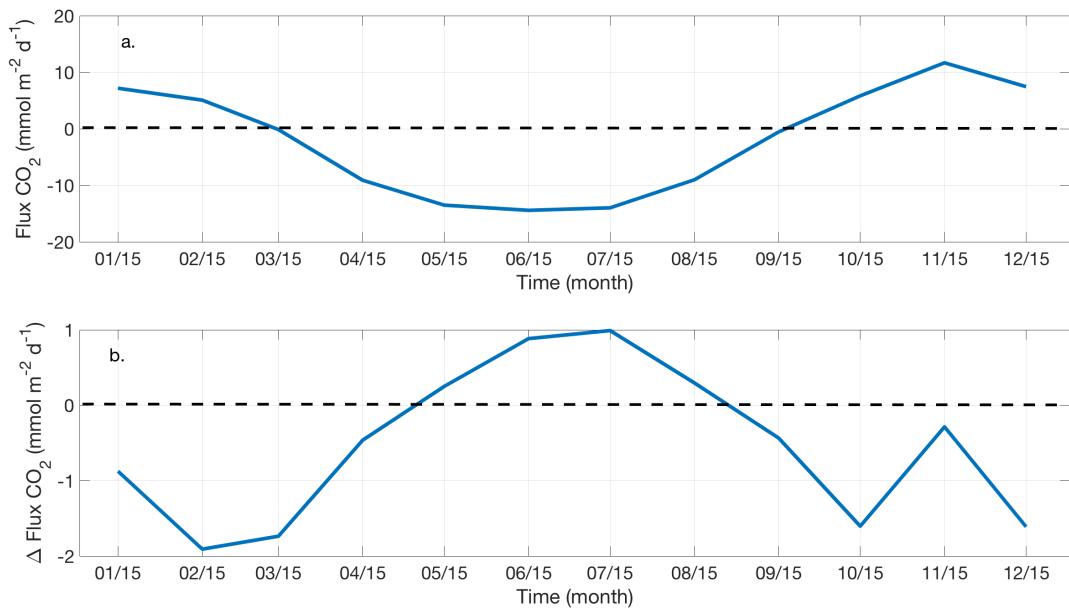


Figure 6. Average, 1998–2011, a) of the air-sea CO_2 flux and b) of the difference between the coastal region and open sea.

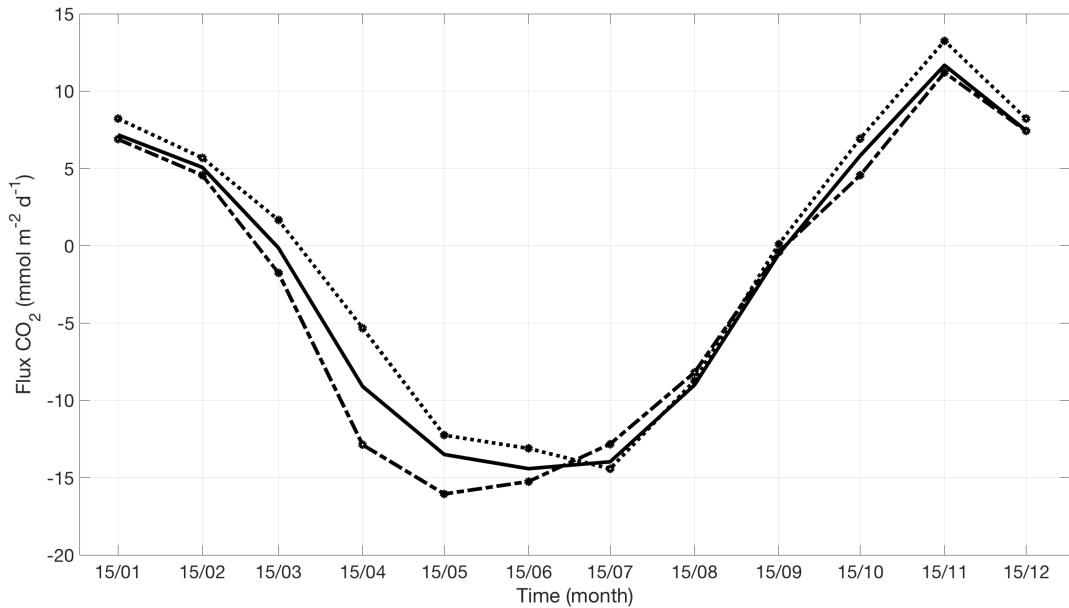


Figure 7. Seasonal cycle of air-sea CO_2 flux for Baltic Sea. Solid line represent the average for the full period (1998–2011), dotted line with marker is for the first 5 years (1998–2002) and dashed line is for the last five years (2007 to 2011).

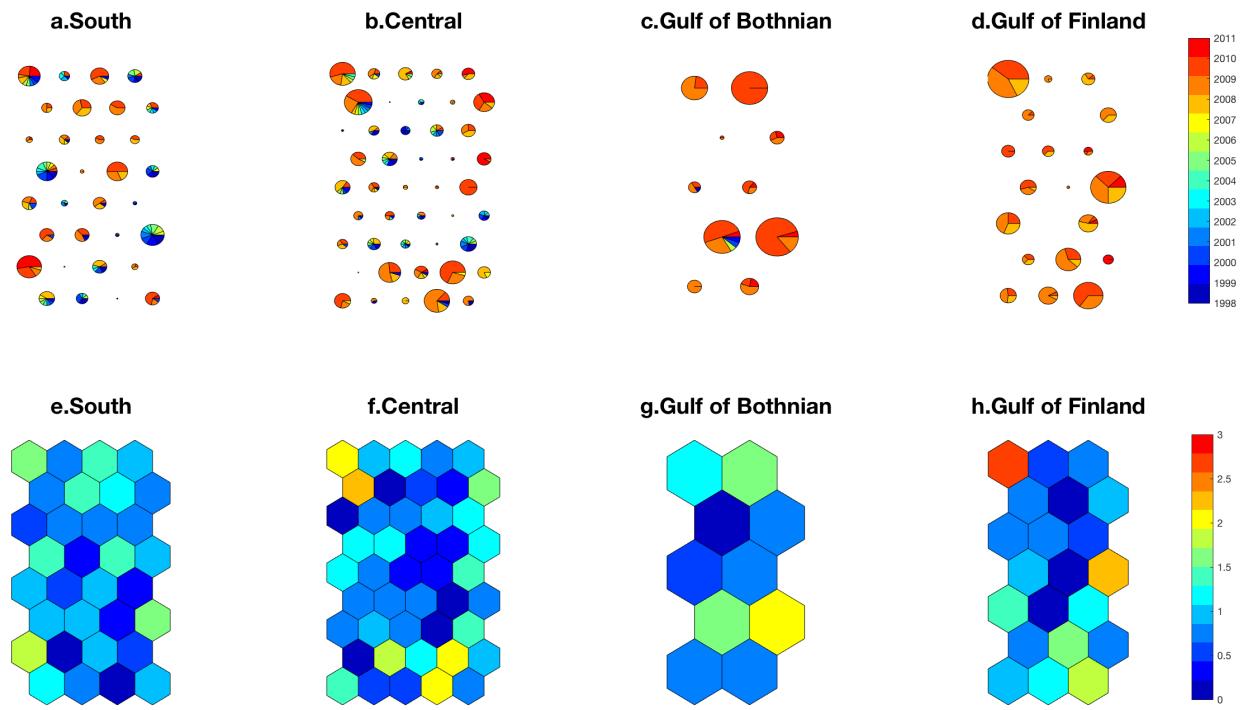


Figure 8. a.,b.,c. and d. are the distribution of the years of each data in each class for each basin SOM e.,f.,g. and h. are the percentage of the total data present in each class of the different basins' SOM. The size of the circles in the top figures is also representative of the percentage of the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

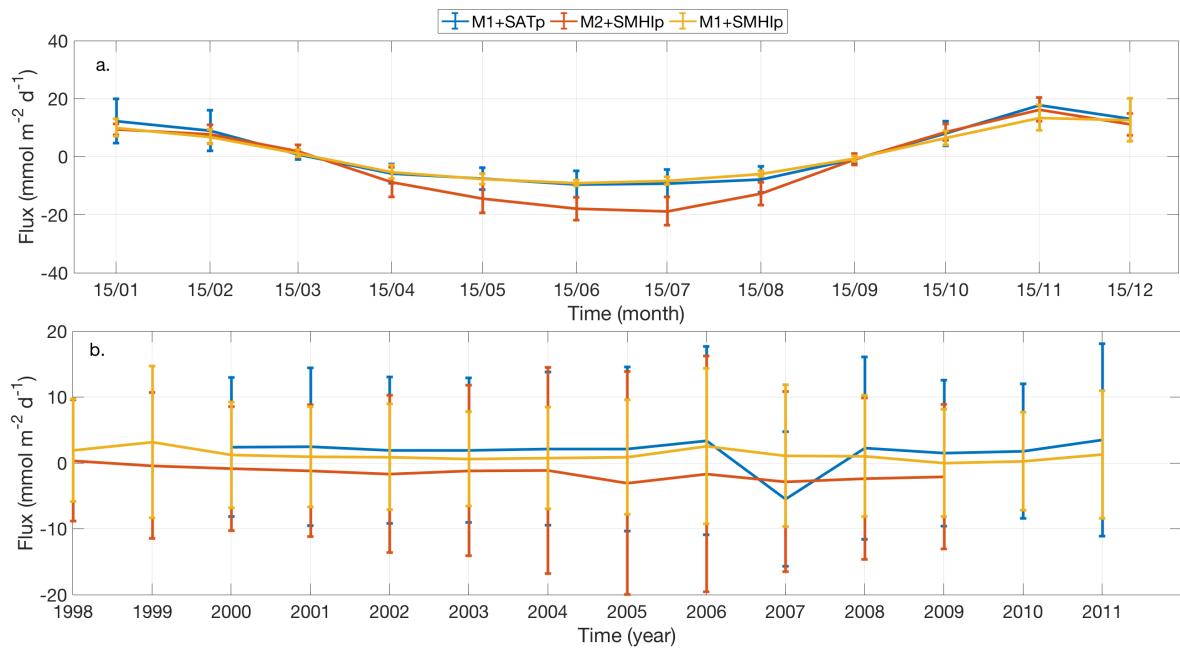


Figure 9. The air-sea CO_2 flux estimate evolution with method 1 and the SATp product (Blue); method 2 and the SMHIp product (Red); method 1 and the SMHIp product (Yellow). a. for a year b. in average for all the years.

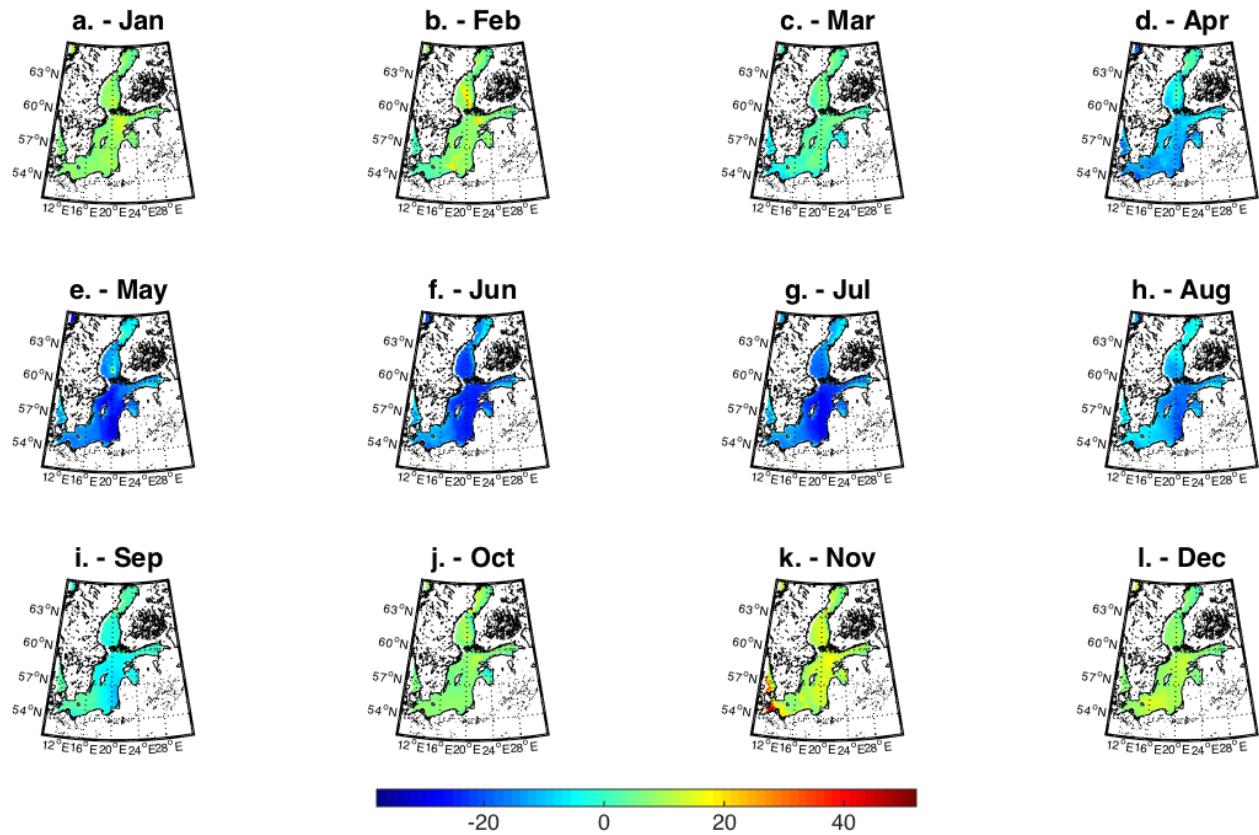


Figure 10. Temporal evolution of the air-sea CO_2 flux between 1998 and 2011 based on SMHIP data.