

Response to the Editor

Dear Prof. Marilaure Grégoire,

We have carefully addressed the comments from Referee #2 in the revised manuscript and the attached point-by-point response.

As requested, we have now made the full biogeochemical simulation in the ORCA2 model as requested by Referee #2. Furthermore, we have also thoroughly modified Section 4.2 to carefully address their criticism.

In addition, we include a new Appendix demonstrating that the perturbation approach agrees with the full approach within 3%, and we include a short paragraph at the beginning of the Methods section to provide a general overview of the simulations that were made.

Thank you for the effort that you have put into evaluating and improving this work.

Sincerely,

Jens Terhaar

Response to Referee # 2

We thank Referee #2 for their comments. Below is a point-by-point response, which accompanies a thoroughly revised manuscript.

Referee # 2

First of all I appreciate that the authors carefully considered most of previous comments. As I already mentioned the paper is clear and provides interesting results. However, some concerns remain in addition to some errors in the revised version. Were these concerns lifted, the paper would then be fit for publication.

Major comments

Reviewer Question 1 — In my first review I had questioned the adequacy of the initialization with ORCA05 results in 1958 and suggested that this method be compared for ORCA2 with an experiment identical to that performed with ORCA05 over the entire period. I expected results of this sensitivity experiment to be presented in Figures 2, 7, and 9 and discussed in the text. However, the authors only performed a simplified perturbation simulation with ORCA2. This does not allow assessing the method consisting in the initialization of ORCA2 and ORCA025 with ORCA05 results in 1958. In addition, except for the mention in Section 4.2, there is no discussion of that additional experiment.

Reply: As requested, we have now made the full biogeochemical simulation in ORCA2, i.e. the one that is analogous to that performed with ORCA05 (1870–2012). The results are now presented in Figures 2, 7, and 9. These results are discussed in sections 3.3 and 4.5. Section 4.2 has been thoroughly modified.

Reviewer Question 2 — The discussion of model data comparison (Section 4.2) is mostly speculative and relies on a misunderstanding of the TTD method.

The fact that models predict lower values than data-based C_{ant} reconstructions is no proof that reconstructions overestimate C_{ant} as sentence on lines 2-3 page 14 suggests. The different model versions clearly underestimate CFC-12 invasion (Sections 3.2 and 4.1). In consequences one would also expect C_{ant} to be underestimated. In that respect why do the authors insist on lowering data-based estimates?

While the TTD method as applied by Tanhua et al. (2009) or in GLODAP-v2 is less well constrained for large transit times (since this method relies on tracers with a short atmospheric history) it does not follow that C_{ant} should be set to zero whenever CFC concentrations are very low. Any water parcel in the ocean is characterized by a distribution of transit times (TTD) which differs from a delta-function due to the presence of mixing (e.g., Waugh et al., 2006). The mean of that distribution corresponds to the mean water age and its width depends on mixing strength and pathways. The assumption that the TTD width is equal to its mean seems to be adequate enough for most ocean areas (Waugh et al., 2006; Tanhua et al., 2009). Hence the water body

under consideration is characterized by ages ranging from zero to the mean age and beyond. Taking that into account, and acknowledging that CFC's and CO₂ do not have the same atmospheric history (CFC-12 concentrations in the atmosphere started to rise significantly after 1950 while the anthropogenic carbon perturbation started 2 centuries earlier) a mean age of 300 to 400 years does not preclude any C_{ant} contribution at depth.

Reply: In Section 4.2, we have removed any speculation and no longer attempt to adjust the data-based estimates downward for an improved model-data comparison. We clearly state that our models underestimate C_{ant} concentrations in the deep waters of the Arctic Ocean, while emphasizing the importance of the CFC-12 model evaluation for that conclusion. In the revised version of section 4.2, we also mention that the different data-based approaches that have been used to estimate C_{ant} generally yield different results in deep waters and that the TTD approach tends to be on the high end (Khatiwala et al., 2013).

Additionally there is no rationale for assuming that “there is a symmetry during 1765–2005 about the ORCA05 result with ORCA2 being lower and ORCA025 being higher” even if it happens to be the case after 1958. There are many processes at stake (air-sea exchange, lateral transport, mixing, atmospheric increase...). Therefore the response of the system is expected to be non-linear. In consequence there is no justification for assuming that the C_{ant} inventory with ORCA025 would be larger by 0.4 Pg C had the experiment started in 1765.

Reply: We have now removed the quantitative argument, based on symmetry, that the Arctic Ocean inventory in ORCA025 would have been 0.4 Pg C higher had that simulation started in 1765 instead of being initialized in 1958 with results from ORCA05. We now emphasize simply the direction of the expected change, an increase based on (1) the greater penetration of CFC-12 into intermediate waters in ORCA025 relative to ORCA05 (Figure 5) and (2) the weaker penetration of CFC-12 for ORCA2 along with the corresponding lower simulated C_{ant} inventory in ORCA2 when it was integrated since 1765 compared to it being initialized in 1958 with results from ORCA05. Moreover, the sign of the divergence of ORCA2 from ORCA05 is always consistent (negative), whether initialized from ORCA05 in 1958 or in the two perturbation simulations (initialized in 1765 and in 1870). The same consistency must hold for ORCA025 relative to ORCA05, although the sign of its divergence is opposite.

In regards to the physical processes that are mentioned by Referee #2, they are nonlinear, but there is no substantial trend nor shift in trend in the physical forcing that was imposed on the model throughout the simulation. That is, the DFS forcing has little trend as pointed out in the revised manuscript.

In summary, all of our transient tracer simulations indicate that the ventilation of subsurface waters is stronger in ORCA025 than in ORCA05. The former absorbs more CFC-12 and C_{ant} than the latter. It follows that any “partial” simulation where ORCA025 is initialized with ORCA05 output partway through, as we have done due to computational limitations, will absorb less C_{ant} than a full ORCA025 simulation. We see no way to escape this logic. Moreover it is supported by the consistent divergence of ORCA2 from ORCA05 in three simulations (initialized at different times along the way), all in the opposite direction since subsurface ventilation in ORCA2 is weaker than in ORCA05.

Further, the ORCA2 experiment which result in a lower inventory is not a biogeochemical experiment but a perturbation one which relies on a simplified carbon cycle. Inventories should not be corrected on that basis. Results of a complete biogeochemical experiment with ORCA2 starting in 1765 would be needed for such assessment. I am rather surprised that no such experiment seems

to be available?

Reply: We have now made an ORCA2 biogeochemical simulation starting in 1870, identical to the ORCA05 simulation to test the effect of branching. It would not have been consistent to start that new full biogeochemical simulation in ORCA2 in 1765 because the full biogeochemical simulation in ORCA05 only started in 1870.

The assertion by Referee #2 that inventories should not be corrected based on the perturbation approach is founded on the assumption that it gives very different results than does the full biogeochemical approach. Because a rigorous comparison of the two approaches has never been published, we now offer this comparison as an Appendix in the revised manuscript. It shows that the C_{ant} inventory simulated with the perturbation approach agrees within 3% of that from the full biogeochemical approach, both globally and regional. Moreover, there is remarkable overall structural agreement. Therefore, we still use the difference between two perturbation simulations (one starting in 1765 and another in 1870) to correct for the late starting date (1870) of the full biogeochemical simulations, each made in a consistent fashion at the different resolutions (ORCA025, ORCA05, ORCA2, and ORCA2*).

Rather than aiming at reconciling modeled C_{ant} and data-based reconstruction this section should be devoted to discussing C_{ant} in view of the CFC-12 results.

Reply: As mentioned above, Section 4.2 no longer attempts to reconcile model and data-based C_{ant} by adjusting the deep-water, data-based estimates downward. It now offers a more refined discussion of C_{ant} in light of the CFC-12 results while also mentioning some literature that discusses uncertainties associated with the data-based C_{ant} estimates in deep waters.

Reviewer Question 3 — Conclusions need to be revised along the preceding lines (Page 17 lines 29-32 and page 18, line 1-4). The conclusions should also mention that all model versions underestimate CFC inventories in the Arctic, hence underestimated C_{ant} inventories.

Reply: As in section 4.2, the revised Conclusions no longer mention any downward adjustment of the TTD data-based estimates. They also now mention that all model configurations underestimate C_{ant} inventories because they generally underestimate CFC-12.

Reviewer Question 4 — Model vertical resolution and mixing schemes.

a. In the model description on page 4 the authors state “Vertically, all three model configurations have the same discretization, where the full- depth water column is divided into 46 depths levels, whose thicknesses increase from 6 m at the surface to 500 m in the deepest grid box” If I am well informed the 500 m box thickness for deep boxes is typical of ORCA2 with 31 levels while a thickness of 6 m at the surface is typical of the 46 levels versions. Is the vertical grid spacing actually the same for all 3 model versions?

Reply: There was confusion on this point because we did not mention in this sentence that the “partial steps” formulation can essentially double the thickness of the the bottom layer. In section 2.1 of the revised manuscript we eliminate this confusion, writing the following: “Vertically, all three model configurations have the same discretization, where the full-depth water column is divided into 46 levels

whose thicknesses vary from 6 m (top level) to 249 m (level 45), but that the latter can reach up to 498 m, being extended into level 46 as a function of the bathymetry (partial steps)."

b. Additionally, one may wonder if the vertical diffusivity and viscosity are represented the same way in all 3 versions? Could the authors add information on that aspect?

Reply: We have now added the following sentence to section 2.1: "Vertically, the same eddy viscosity ($1.2 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$) and diffusivity coefficients ($1.2 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$) were used in all three resolutions."

Minor comments

Reviewer Question 5 — The CFC model-data misfits quoted on page 11 (lines 13-15) do not agree with the values in Table 6. In the later ORCA2 displays the best agreement with data!

Reply: The labels for ORCA2 and ORCA025 were reversed and have been corrected.

Reviewer Question 6 — Figure 4: the bottom right panel displays CFC-12 results and not temperature.

Reply: That erroneous label has been corrected.

Reviewer Question 7 — Page 7, line 2: "...using ORCA05 until 1957 and then all three configurations from 1858 1958 to 2012..."

Reply: This correction has been made.

Reviewer Question 8 — Page 11, line 13: Table 6 does not come into order; should be Table 5

Reply: Table 5 and Table 6 have now been put into the proper order.

Reviewer Question 9 — Page 11, line 29: "In 2002, the upper limit of the modeled Cant inventory range..."

Reply: Done

Reviewer Question 10 — Page 11, line 32: "These This correction is 0.4 Pg C in 2005 for each resolution..."

Reply: Done

Reviewer Question 11 — Page 12, line 25: "... flow fields is 0.05 Pg C (3%) is smaller than..."

Reply: Done

Reviewer Question 12 — Page 14, line 15: "Although we cannot assess this affect effect directly"

Reply: Done

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Model constraints on the anthropogenic carbon budget of the Arctic Ocean

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Abstract. The Arctic Ocean is projected to experience not only amplified climate change but also amplified ocean acidification. Modeling future acidification depends on our ability to simulate baseline conditions and changes over the industrial era. Such centennial-scale changes require a global model to account for exchange between the Arctic and surrounding regions. Yet the coarse resolution of typical global models may poorly resolve that exchange as well as critical features of Arctic Ocean circulation. Here we assess how simulations of Arctic Ocean storage of anthropogenic carbon (C_{ant}), the main driver of open-ocean acidification, differ when moving from coarse to eddy-admitting resolution in a global ocean circulation-biogeochemistry model (NEMO-PISCES). The Arctic's regional storage of C_{ant} is enhanced as model resolution increases. While the coarse-resolution model configuration ORCA2 (2°) stores 2.0 Pg C in the Arctic Ocean between 1765 and 2005, the eddy-admitting versions ORCA05 and ORCA025 (1/2° and 1/4°) store 2.4 and 2.6 Pg C. The difference in inventory between model resolutions ~~is only due to that is accounted for is only from their divergence after 1958. It would be 1958, when ORCA2 and ORCA025 were initialized with output from the intermediate resolution ORCA05. The difference would have been~~ larger had all model resolutions been initialized in 1765 as was ~~the intermediate resolution model (ORCA05)~~. The ORCA25 ~~results falls within the uncertainty range Arctic C_{ant} storage estimate of 2.6 Pg C should be considered a lower limit because that model generally underestimates observed CFC-12 concentrations. It reinforces the lower limit~~ from a previous data-based C_{ant} ~~storage estimate approach~~ (2.5 to 3.3 Pg C). ~~Across the three resolutions~~ Independent of model resolution, there was roughly three times as much C_{ant} that entered the Arctic Ocean through lateral transport than via the flux of CO₂ across the air-sea interface. Wider comparison to nine earth system models that participated in the Coupled Model Intercomparison Project Phase 5 (CMIP5) reveals much larger diversity of stored C_{ant} and lateral transport. Only the CMIP5 models with higher lateral transport obtain C_{ant} inventories that are close to the data-based estimates. Increasing resolution also enhances acidification, e.g., with greater shoaling of the Arctic's average depth of the aragonite saturation horizon during 1960–2012, from 50 m in ORCA2 to 210 m in ORCA025. ~~To assess the potential to further refine modeled estimates of the Arctic Ocean's C_{ant} storage and acidification, sensitivity tests that adjust model parameters are needed given that century-scale global ocean biogeochemical simulations still cannot be run routinely at high resolution.~~

Even higher model resolution would likely further improve such estimates but its prohibitive costs also call for other more practical avenues for improvement, e.g., through model nesting, addition of coastal processes, and refinement of subgrid-scale parameterizations.

1 Introduction

5 The Arctic is experiencing amplified ocean acidification (Steinacher et al., 2009) and amplified climate change (Bekryaev et al., 2010), both of which may affect the marine ecosystem (Gattuso and Hansson, 2011). The main driver of the ongoing acidification of the open ocean is the increase in atmospheric CO₂ during the industrial era and the ensuing uptake of anthropogenic carbon from the atmosphere. Although this absorbed anthropogenic carbon cannot be measured directly, being dominated by the natural component, it has been estimated from other oceanographic data.

10 For instance, Gruber et al. (1996) developed the ΔC^* method, building on seminal studies (Brewer, 1978; Chen and Millero, 1979) and their criticism (Broecker et al., 1985) as well as large new global data sets with improved CO₂ system measurements. That back-calculation method first calculates the total dissolved inorganic carbon (C_T) at equilibrium with atmosphere before the water parcel is subducted. ~~That~~ The preformed C_T is then corrected for changes due to biological activity, as estimated from measurements of dissolved oxygen, total alkalinity (A_T), and nutrients, after which an estimate of preindustrial carbon is

15 removed, finally yielding ΔC^* . Yet the ΔC^* method's assumption of a constant air-sea CO₂ disequilibrium appears problematic in the high latitudes (Orr et al., 2001).

A second approach approximates the invasion of anthropogenic CO₂ into the interior ocean by a Transient Time Distribution (TTD) method, itself constrained by observations of transient-tracers such as CFC-12 or SF₆ (Hall et al., 2002; Waugh et al., 2004). A third approach uses a Green's function instead of a TTD while also exploiting multiple transient tracers to

20 assess the ocean's temporally changing distribution of anthropogenic carbon (Khatiwala et al., 2009). A comparison of these methods suggests that by 2010 the ocean had absorbed 155 ± 31 Pg C of anthropogenic carbon, around one-third of all emitted anthropogenic carbon (Khatiwala et al., 2013)

Less attention has been paid to anthropogenic carbon storage in the Arctic. Sabine et al. (2004) estimated that the Arctic Ocean had absorbed 4.9 Pg C by 1994. Yet without estimates for anthropogenic carbon in the Arctic itself, Sabine et al. scaled

25 the Arctic inventory to be 5% of their ΔC^* -based estimate for global anthropogenic carbon storage, assuming the same Arctic:Global ocean ratio as in the global gridded distribution of observed CFC-12 (Willey et al., 2004). More recently, Tanhua et al. (2009) used Arctic observations of CFC-11, CFC-12, and SF₆ and the TTD approach, revising the former Arctic anthropogenic carbon storage estimate downward to a range of 2.5 to 3.3 Pg C for year 2005. With that estimate, they emphasized that while the Arctic Ocean represents only 1% of the global ocean volume, it stores 2% of the global ocean's anthropogenic

30 carbon. Although these numbers are relatively small, Arctic concentrations of anthropogenic C_T must be relatively large, thus driving enhanced acidification in the Arctic Ocean. No other approaches have been used in the Arctic.

To provide an alternate approach to estimate anthropogenic carbon in the Arctic and to assess its budget and the mechanisms that control it, ~~we made one could make~~ carbon cycle simulations over the industrial era ~~using with~~ a coupled ocean circulation-

biogeochemical model. A global-scale ~~configuration is used~~ model configuration would be needed to account for the Arctic in the context of the global carbon cycle, while avoiding ~~artifacts~~ artefacts from lateral boundary conditions that ~~are needed for~~ must be imposed in regional models. ~~Simulations with the same model (NEMO-PISCES) are made at three resolutions, from coarse to eddy admitting, to help assess the extent to which coarse-resolution models may need to be improved to adequately~~ simulate anthropogenic carbon storage in the Arctic Ocean.

~~Coarse-resolution may be~~ Yet typical ocean general circulation models have coarse resolution, which may be insufficient to adequately represent Arctic Ocean bathymetry, shelf, slopes, and ridges, all of which affect Arctic Ocean circulation (Rudels et al., 1994).

The bathymetry of the Arctic Ocean differs from that in other oceans in part because of the preponderance of shelf seas, comprising 53% of the total surface area (Jakobsson, 2002) (Figure 1). The remaining 47% of ~~the-its~~ surface area covers 95% of the total volume of the Arctic Ocean, split across four deep basins: the Nansen basin, the Amundsen basin, the Makarov basin, and the Canadian basin. Water masses enter these deep basins via (1) ~~via~~ deep inflow from the Atlantic through the Fram Strait into the Nansen basin, (2) ~~via deep-water~~ inflow from the Barents Sea ~~to that sinks into~~ the Nansen basin through the St. Anna Trough, as cooling increases density, and (3) ~~and via transport from~~ density flows along the continental shelves that are driven by brine rejection from sea-ice formation (Jones et al., 1995). These three local ~~processes-transfers~~ are difficult to resolve in coarse-resolution models, e.g., local density flows necessitate much higher resolution (Proshutinsky et al., 2016). Model resolution also affects the simulated interior circulation of the Arctic Ocean by its connection to the global ocean circulation via four relatively narrow and shallow passages: (1) the Canadian Archipelago, (2) the Fram Strait, (3) the Barents Sea Opening, and (4) the Bering Strait (Aksenov et al., 2016). Lateral exchange of water, carbon, and nutrients across these sections also affects Arctic Ocean primary production and acidification (Popova et al., 2013; Luo et al., 2016).

Here ~~we-our aim is to~~ use a three-dimensional model to help refine the estimate of the total anthropogenic carbon in the Arctic Ocean while assessing the dominant pathways by which anthropogenic carbon enters the Arctic Ocean and the relative importance of that lateral input relative to the air-sea flux. Three simulations made at increasingly higher grid resolution ~~allow us to assess~~ are aimed at assessing the extent to which the coarse resolution used by typical global ocean models may need to be improved to adequately estimate storage of anthropogenic carbon in the Arctic Ocean and associated ocean acidification.

2 Methods

Multiple global simulations were made to assess C_{ant} in the ocean. Simulations were made with a state-of-the-art ocean circulation-biogeochemical model at three resolutions over the industrial period since the mid-19th century, i.e., as typical of recent model comparison efforts. Longer simulations were also made at the same resolutions with a less costly (and less precise) perturbation approach to correct for the missing anthropogenic carbon given that the actual industrial era began about a century earlier (mid-18th century). The highest resolution configuration used an unprecedented lateral grid spacing for such long, global, biogeochemical simulations, although its cost meant its effect could be assessed only over 1958–2012.

2.1 Models

For our study, we used the global ocean circulation model NEMO-v3.2 (Nucleus for European Modeling of the Ocean - version 3.2) ~~including the biogeochemical component PISCES-v1 (Pelagic Interactions Scheme for Carbon and Ecosystem Studies) (Aumont and Bopp, 2006)~~. The NEMO model has three parts: (1) the ocean dynamics and thermodynamics model OPA (Madec, 2008), (2) the sea-ice model LIM (Vancoppenolle et al., 2009), and (3) the passive tracer module TOP. This physical model is coupled via TOP to version 1 of PISCES ([Pelagic Interactions Scheme for Carbon and Ecosystem Studies](#)) ([Aumont and Bopp, 2006](#)). For this study we used NEMO at three resolutions: a laminar 2°-configuration (ORCA2) typical of coarse-resolution ocean models (Madec et al., 1998), which does not resolve eddies; an intermediate 0.5°-configuration (ORCA05) that just begins to allow eddies to appear spontaneously (Bourgeois et al., 2016); and a higher-resolution, eddy admitting version, i.e., 0.25°-configuration (ORCA025), which is still not eddy resolving (Barnier et al., 2006). [The highest resolution simulation is referred to as ORCA025-G70 in the DRAKKAR ensemble.](#)

All three configurations have a tripolar, curvilinear horizontal grid. One grid pole (singularity) is located at the geographical South Pole while the conventional North-Pole grid singularity over the Arctic Ocean has been replaced by two [grid](#) singularities, both displaced over land, one over Canada and the other over Russia (Madec et al., 1998), thereby saving computational costs and avoiding numerical ~~artifacts.~~

[artefacts.](#) From 90°S to 20°N, the grid is a normal Mercator grid; north of 20°N, it is distorted into ellipses to create the two northern singularities (Barnier et al., 2006; Madec, 2008). The grid size changes depending on resolution and location (Table 1). The mean horizontal grid size in the Arctic Ocean (average length of the 4 horizontal edges of surface grid cells in the Arctic Ocean) is 121 km in ORCA2, 29 km in ORCA05, and 14 km in ORCA025. The ~~minimum-smallest~~ horizontal grid size in the Arctic is 63 km in ORCA2, 9 km in ORCA05, and 3 km in ORCA025.

Vertically, all three model configurations have the same discretization, where the full-depth water column is divided into ~~46 depths levels, whose thicknesses increase~~ [levels whose thicknesses vary](#) from 6 ~~m at the surface to 500 m in the deepest grid box~~ [m \(top level\) to 249 m \(level 45\), but the latter can reach up to 498 m, being extended into level 46 as a function of the bathymetry \(partial steps\).](#) For its bathymetry, the ocean model relies on the 2-minute bathymetry file ETOPO2 from the National Geophysical Data Center, which is based on satellite derived data (Smith and Sandwell, 1997) except for the highest latitudes: the IBCAO bathymetric data is used in the Arctic (Jakobsson et al., 2000) and BEDMAP bathymetric data is used for the Southern Ocean south of 72°S (Lythe and Vaughan, 2001). To interpolate the bathymetry on the model grid, the median of all data points in one model grid cell was computed. NEMO uses the partial-step approach for the model to better match the observed topography. In this approach, the bathymetry of the model is not tied directly to the bottom edge of the deepest ocean grid level, which varies with latitude and longitude; rather, the deepest ocean grid level for each column of grid cells is partially filled in to better match the observed ocean bathymetry. ~~For the parameter values and numerical characteristics of the ORCA025 configuration, we followed Barnier et al. (2006).~~ ([Barnier et al., 2006](#)).

The lateral isopycnal diffusion and viscosity coefficients were chosen depending on the resolution (Table 2). In ORCA2, a Laplacian viscosity operator was used, whereas a bi-Laplacian operator was used in ORCA05 and ORCA025. To simulate the

effect of eddies on the mean advective transport in the two coarser resolution configurations, the eddy parameterization scheme of Gent and McWilliams (1990) was applied with eddy diffusion coefficients indicated in Table 2. Vertically, the same eddy viscosity ($1.2 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$) and diffusivity coefficients ($1.2 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$) were used in all three resolutions.

The biogeochemical model PISCES (Aumont and Bopp, 2006) includes four plankton functional types: two phytoplankton (nanophytoplankton and diatoms) and two zooplankton (micro- and meso-zooplankton). The growth of phytoplankton is limited by the availability of five nutrients: nitrate, ammonium, total dissolved inorganic phosphorus P_T , total dissolved silicon Si_T , and iron. The nanophytoplankton and diatoms are distinguished by their need for all nutrients, with only diatoms requiring silicon. While the Fe:C and Chl:C ratios of both phytoplankton groups as well as the Si:C ratio of diatoms are predicted prognostically by PISCES, the remaining macronutrient ratios are held constant at C:N:P = 122:16:1 (Takahashi et al., 1985). The same ratio holds for nonliving compartments: dissolved organic matter (DOM) and both small and large sinking particles, which differ in their sinking velocity. In PISCES, nutrients are supplied by three external pathways: atmospheric dust deposition, river delivery, and sediment mobilization of iron. Dust deposition was taken from a simulation by Tegen and Fung (1995). River discharge of C_T and dissolved organic carbon (DOC) is based on the Global Erosion Model (GEM) by Ludwig et al. (1998). Riverine DOC was assumed to be entirely labile, being instantaneously transformed into C_T as soon as it enters the ocean. River delivery of the other four nutrients (Fe, N, P, and Si) were calculated from riverine C_T delivery, assuming constant ratios of C:N:P:Si:Fe = 320:16:1:53.3:3.64 $\times 10^{-3}$ (Meybeck, 1982). For sediment mobilization, dissolved iron input was parameterized as $2 \mu\text{mol Fe m}^{-2} \text{ day}^{-1}$ for depths shallower than 1100 m following Moore et al. (2004).

2.2 Biogeochemical simulations

For initial conditions, we used observational climatologies for temperature and salinity combined from three sources (Barnier et al., 2006), for dissolved oxygen and nutrients (nitrate, P_T , and Si_T) from the 2001 World Ocean Atlas (Conkright et al., 2002), and for preindustrial C_T and A_T from the observation-based Global Data Analysis Product (GLODAP) (Key et al., 2004). As comparable observational climatologies for DOC and iron are lacking, those variables were initialized from output of a 3000-year spin up of an ORCA2 simulation including PISCES. Other tracers have short recycling times and were thus ~~initialized~~ initialized with globally uniform constants.

For physical boundary conditions, all simulations were forced with the same DRAKKAR Forcing Set (DFS) constructed ~~by Brodeau et al. (2010)~~ originally by Brodeau et al. (2010) and routinely updated. This historical ~~reanalysis~~ reanalysis-based forcing data set provides surface air temperature and humidity at 2 m, wind fields at 10 m, shortwave and longwave radiation, and the net surface freshwater flux (evaporation minus precipitation). ~~This data set~~ It covers 55 years, including 1958–2001 from version 4.2 and ~~then~~ 2002–2012 from version 4.4.

A 50-year spin up was first made from rest in the ORCA05 NEMO-PISCES model (coupled circulation-biogeochemistry), after initializing the model variables with the above-mentioned fields. The resulting simulated physical and biogeochemical fields were then used to initialize the ORCA05 NEMO-PISCES simulations in 1870, and that model was subsequently integrated ~~during 1870–1957~~ over 1870–1957. Since no atmospheric reanalysis is available during that period, we simply looped the DRAKKAR Forcing Set. Then at the beginning of 1958, the ORCA05 simulated fields were interpolated to the ORCA2

and ORCA025 grids, and simulations were continued in each of the three configurations during 1958 to 2012 (Fig. 2). We refer to these simulations as B1870-ORCA2, B1870-ORCA05, and B1870-ORCA025 (Table 3), where the first letter refers to the type of simulation (B for biogeochemical), the following four numbers refer to the initialization year, and the remainder refers to the resolution used over 1958–2012.

- 5 ~~For this study we~~ The initialization of the ORCA025 and ORCA2 models in 1958 with interpolated fields from ORCA05 introduces an error in the results from B1870-ORCA2 and B1870-ORCA025. To estimate this branching error in the low resolution model, we also made a simulation using ORCA2 from 1870 to 2012 (referred to as B1870-ORCA2*) and compared it to B1870-ORCA2 (initialized in 1958 with output from ORCA05). This strategy was not possible for ORCA025, because running ORCA025 with PISCES over 1870–2012 is too costly.
- 10 For each member of this “B” class of simulations, we actually made two types of ~~simulations~~ runs, historical and control, both forced with the same reanalysis fields. ~~In addition, the~~ Those two runs differ only in their atmospheric CO₂ concentrations. The control simulations were forced with the preindustrial CO₂ concentration of 287 ppm in the atmosphere over the entire period from 1870 to 2012. The historical simulations were forced with yearly averaged historical atmospheric CO₂ concentrations reconstructed from ice cores and atmospheric records over 1870 to 2012 (Le Quéré et al., 2015). ~~Both, starting at the same~~
- 15 ~~reference of 287 ppm (Le Quéré et al., 2015). Making both~~ the control and the historical ~~simulations, were made for all three~~ resolutions between 1958 to 2012 to correct potential model drifts. ~~We defined the difference between the historical simulation and the control simulation~~ runs for each of the B class of simulations and taking the difference automatically corrects for model drift. Indeed, that difference is defined as the anthropogenic component. ~~While the ORCA2 and ORCA025 simulations are presented for the first time, the ORCA05 simulations were previously used by Bourgeois et al. (2016) to assess the budget of anthropogenic carbon in the coastal ocean.~~
- 20

2.3 C_{ant} - C_{ant} perturbation ~~simulations~~ simulations

- Because of computational limitations, it was necessary to start the anthropogenic CO₂ perturbation of our reference ORCA05-PISCES simulation in 1870 as opposed to the traditional earlier reference of 1765 (Sarmiento et al., 1992), a more realistic approximation of the start of the industrial-era CO₂ increase. A similar compromise was adopted for CMIP5 (Taylor et al.,
- 25 2012). During that missing 105 years, atmospheric xCO₂ increased from ~~277.86 to 287.29~~ 278 to 287 ppm, a 9 ppm difference that seems small relative to today’s total perturbation with atmospheric xCO₂ now above 400 ppm. However, Bronselaer et al. (2017) estimated that global ocean uptake of C_{ant} - C_{ant} in 1995 is actually underestimated by ~30% (29 Pg C) for simulations that reference the natural preindustrial state to 1850 rather than 1765. The cause is partly due to ocean carbon uptake during the missing 1765–1850 period, but mostly it is due to the higher preindustrial reference for atmospheric xCO₂ that results in
- 30 the air-sea flux of C_{ant} - C_{ant} being underestimated throughout the entire simulation. Unfortunately, we cannot use Bronselaer et al.’s results to correct our biogeochemical simulations because ~~their~~ they do not include the Arctic Ocean in their global data-based assessment. Furthermore, their reference date in the mid 19th century is 20 years earlier than ours ~~and because they do not include the Arctic Ocean in their global data-based assessment.~~

Instead, to correct for the late starting date of our biogeochemical simulations, we made additional simulations using the more efficient single-tracer perturbation approach (Sarmiento et al., 1992) rather than the full PISCES biogeochemical model (24 tracers). To account for the missing carbon, we added the difference between two perturbation simulations, denoted as P rather than B, one starting in 1765 (P1765) and the other ~~one starting~~ in 1870 (P1870). For consistency, we applied the same initialization strategy as for the biogeochemical simulations, i.e. using ORCA05 until the end of 1957 with that output serving as the initial fields for subsequent 1958–2012 simulations in all three configurations. The ~~difference of C_{ant} naming convention for the “P” class of simulations is like that for the “B” class (indicated by the first letter). The difference is that in each P simulation there is only one tracer and one run for each (no need for a control and a historical run). However initializing a set of P simulations in 1765 as well as in 1870 implies twice the number of simulations (Table 3). The difference in C_{ant} between P1765 and P1870 simulations was later added to the NEMO-PISCES simulations as a late-start correction to the biogeochemical simulations (B1870)~~, for each resolution separately.

The perturbation approach of Sarmiento et al. (1992) avoids the computationally intensive standard CO₂ system calculations by ~~only accounting for~~ accounting for only the perturbation ($C_{ant}C_{ant}$), assuming it is independent of the natural carbon cycle. By focusing only on anthropogenic carbon, this approach exploits a linear relationship between the anthropogenic change in ~~oceanic surface ocean~~ ocean’s pCO₂ [μatm] and its ratio with the ~~ocean’s~~ corresponding change in C_T (~~C_{ant} surface ocean dissolved inorganic carbon~~ δC_T):

$$\frac{\delta p\text{CO}_{2o}}{C_{ant}} \frac{\delta p\text{CO}_{2o}}{\delta C_T} = z_0 + z_1 \delta p\text{CO}_{2o}, \quad (1)$$

where $\delta p\text{CO}_{2o}$ is the perturbation in ~~oceanic surface ocean~~ pCO₂ and the coefficients z_0 and z_1 are each quadratic functions of surface temperature [$^{\circ}\text{C}$],

$$z_0 = a_0 + a_1 T + a_2 T^2 \quad (2)$$

$$z_1 = b_0 + b_1 T + b_2 T^2. \quad (3)$$

In the model, Eq. (1) was rearranged to solve for ~~surface ocean~~ $\delta p\text{CO}_{2o}$ in terms of $C_{ant} \delta C_T$ (Sarmiento et al., 1992, Eq. (11)), as needed to compute the air-sea flux (Sarmiento et al., 1992, Eq. (2)). In the air-sea flux equation, the atmospheric xCO₂ was corrected for humidity and atmospheric pressure to convert to $p\text{CO}_{2\text{atm}}$, which thus varies spatially while atmospheric xCO₂ does not (in the model). The atmospheric xCO₂ history for 1765–1869 is from Meinshausen et al. (2017), while the history for 1870 and beyond is the same as used in the NEMO-PISCES simulations. One set of coefficients was derived for our reference atmospheric xCO₂ in 1765; another set was derived for our reference atmospheric xCO₂ in 1870 (Table 4). The original approach was only updated to use the equilibrium constants recommended for best practices (Dickson et al., 2007) and to cover a perturbation of up to 280 ppm (see Supplement). The relative ~~error uncertainty~~ introduced by approximating the perturbation to the ocean CO₂ system equilibria with Eq. (1) remains less than $\pm 0.3\%$ across the global ocean’s observed temperature range when $\delta p\text{CO}_2^{\text{oc}} < 280$ ppm.

~~To correct our biogeochemical simulations for~~ The Arctic C_{ant} inventory in 2012 simulated by the perturbation approach (P1870-ORCA2*) underestimates that simulated by the full biogeochemical approach (B1870-ORCA2*) by 3% (Appendix A).

The corresponding underestimation by P1765-ORCA2* is expected to be similar. The similar bias of P1765-ORCA2* and P1870-ORCA2* means that the bias in their difference is probably much less than 3%. The same holds for P1765-ORCA2 vs. P1870-ORCA2. Thus using their difference to correct for the late start of B1870-ORCA2 is not only practical but also sufficiently accurate for our purposes. In contrast, not correcting B1870-ORCA2 for its late starting date would lead to a 19% underestimation of its Arctic C_{ant} inventory for the full industrial era.

In practice, to make the late-start correction, at each grid cell we added the late-starting date, we used the time-varying difference in C_{ant} for every grid cell C_{ant} between the two perturbation simulations (P1765 – P1870), adding that to the C_{ant} simulated in the biogeochemical simulations.

Lastly, we also made a perturbation simulation with using only ORCA2 from 1765 to 2012, which enables us to evaluate our simulation strategy, i.e. using ORCA05 until 1957 and then all three configurations from 1858 to 2012 (the C_{ant} simulated with B1870 for each resolution separately. From here on, we refer only to these corrected biogeochemical simulations, denoting them as ORCA2, ORCA05, and ORCA025), and ORCA2*.

2.4 CFC-12 simulation

CFC-12 is a purely anthropogenic tracer, a sparingly soluble gas whose concentration began to increase in the atmosphere in the early 1930's, part of which has been transferred to the ocean via air-sea gas exchange. Its uptake and redistribution in the ocean has been simulated following OCMIP-2 protocols (Dutay et al., 2002). The CFC-12 flux (F_{CFC}) at the air-sea interface was calculated as follows:

$$F_{\text{CFC}} = k_w(\alpha_{\text{CFC}} p\text{CFC} - C_s)(1 - I), \quad (4)$$

where k_w is the gas-transfer velocity (piston velocity) in m s^{-1} (Wanninkhof, 1992), $p\text{CFC}$ is the atmospheric partial pressure of CFC-12 in atm from the reconstructed atmospheric history by Bullister (2015), C_s is the sea surface concentration of CFC-12 (mol m^{-3}), α_{CFC} is the solubility of CFC-12 ($\text{mol m}^{-3}\text{atm}^{-1}$) from Warner and Weiss (1985), and I is the model's fractional sea-ice cover. Once in the ocean, CFC-12 is an inert tracer that is distributed by advection and diffusion; it has no internal sources and sinks. Many high-precision measurements of CFC-12 are available throughout the ocean, in sharp contrast to C_{ant} which cannot be measured directly.

As for the other simulations, those for CFC-12 were made using ORCA05 until 1957, at which point those results were interpolated to the ORCA2 and ORCA025 grids. The ORCA05 CFC-12 simulation began in 1932. From 1958 to 2012, CFC-12 was simulated for each resolution separately.

2.5 Arctic Ocean

To assess the anthropogenic carbon budget in the Arctic Ocean, we adopt the regional domain defined by Bates and Mathis (2009) (delineated in Fig. 1). That domain's lateral boundaries and the volume of water contained within them vary slightly among the three model versions due to their different resolutions and bathymetries (Table 1). The signature of these different

volumes is also apparent in the integrated quantity of anthropogenic carbon that is stored in the Arctic in 1958, although the fields for all three models are based on the same 1957 field from the ORCA05 model (Fig. 2).

2.6 Transport across boundaries

Transects are defined (Fig. 1) along the four boundaries as consistently as possible for the three resolutions. Water transport across each of the four boundaries is calculated for each model configuration by using monthly average water velocities at each boundary grid cell along a transect multiplied by the corresponding area of the face of the grid cell through which the water flows. For boundaries defined by a row of cells (Fram Strait, Canadian Arctic Archipelago [CAA], and Bering Strait), the transport is calculated across the northern face of each cell. Conversely, for the boundary that is a jagged line (jagged boundary of the Barents Sea Opening), ~~for each cell the~~ transport is calculated at the northern and eastern faces of each cell and the two transports are summed. ~~Then for~~ For each transect, transport across all of its cells are summed to obtain the transect's monthly net transport. ~~For the C_{ant}~~

For the C_{ant} transport, we do the same but also multiply the water transport at the boundary between two grid cells with their volume-weighted monthly-average concentration. This multiplication of monthly means introduces an error into the transport ~~calculation~~ calculations owing to neglect of shorter term variability. To ~~shed light on~~ elucidate that error, we sum results from those monthly calculations across all four sections, integrate them over time from 1960 to 2012, and compare that to the net transport of C_{ant} C_{apt} into the Arctic Ocean implied by the inventory change minus the cumulative air-sea flux over the same time period. The inventory of C_{ant} C_{apt} is the total mass of C_{ant} C_{apt} inside the Arctic Ocean at a given time, while the cumulative flux is the time-integrated air-sea flux of anthropogenic CO₂ over the Arctic Ocean since the beginning of the simulation. The difference between these two spatially integrated values is the reference value for the net lateral flux into the Arctic Ocean to which is compared the less exact total lateral flux of anthropogenic carbon computed from monthly mean velocity and concentration fields integrated over time. The relative error for transport of C_{ant} ~~across~~ C_{ant} across all the separate boundaries introduced by the monthly average calculations is 28% for ORCA2, 7% for ORCA05, and 3% for ORCA025. ~~Note that this~~ This error applies neither to the C_{ant} C_{apt} inventory, nor to the cumulative C_{ant} air-sea flux or the lateral C_{ant} fluxes, which are all calculated ~~'online'~~ online, i.e. during the simulations.

2.7 CFC-12 observational data

Model simulations were evaluated indirectly by comparing simulated to observed CFC-12. We choose CFC-12 to evaluate the model, because it is an anthropogenic, passive, conservative, and inert tracer, and in contrast to anthropogenic carbon, it is directly measurable. The CFC-12 atmospheric concentration increased from zero in the 1930s to its peak in the 2000s, since declining as a result of the Montreal protocol. Thus CFC-12 is a transient tracer similar to anthropogenic carbon but for which there exist extensive direct measurements, all carried out with high precision during WOCE (World Ocean Circulation Experiment) and CLIVAR (Climate and Ocean - Variability, Predictability and Change) era. Nowadays, ocean models are often evaluated with CFC-11 or CFC-12, especially those destined to be used to assess anthropogenic carbon uptake (Dutay et al., 2002; Orr et al., 2017).

The CFC-12 observations used in this study come from two trans-Arctic cruises: the 1994 Arctic Ocean Section (AOS94) (Jones et al., 2007) and the Beringia 2005 expedition (Anderson et al., 2011) (Fig. 1). AOS94 started on 24 July and ~~finished~~ was completed on 1 September, during which CFC-12 measurements were made at 39 stations. That section ~~starts~~ began in the Bering Strait, ~~enters~~ entered the Canada basin adjacent to Mendeleev ridge, ~~continues~~ continued to the Makarov basin, and ~~ends~~ ended at the boundary of the Nansen basin and the Barents Sea. The Beringia expedition started on 19 August and ended on 25 September 2005. It ~~started~~ began off the coast of Alaska, went through the Canada and Makarov basins, crossed the Lomonosov ridge, and its last CFC-12 station was taken on the Gakkel ridge. These two cruises were chosen ~~among other cruises because of their geographically similar placement and because~~ from among others because they cross large parts of the Arctic, including almost all four major basins.

10 2.8 Data-based estimates of anthropogenic carbon

Our simulated C_{ant} C_{ant} was compared to data-based estimates from Tanhua et al. (2009) for the year 2005 and from GLO-DAPv2 for the year 2002 (Lauvset et al., 2016), both based on the TTD approach.

3 Results

3.1 Physical Evaluation

15 3.1.1 Lateral water fluxes

The lateral water flux across each of the four Arctic boundaries is a fundamental reference for the simulated physical transport, especially ~~when the goal is~~ given our goal to construct a budget that includes lateral transport of passive tracers. Results for lateral water transport in the three model resolutions may be grouped into two classes: coarse resolution and higher resolutions. In ORCA2, water enters the Arctic Ocean from Barents Sea and the Bering Strait (2.1 Sv split evenly), with 86% of that total leaving the Arctic via the Fram Strait and the remaining 14% flowing out via the CAA (Table 5). Conversely, outflow through the CAA is seven times larger for ORCA05 and nine times larger for ORCA025, being fueled by 26 ~~%~~ to 46% more inflow via the Bering Strait and 110 ~~%~~ to 170% more inflow via the Barents Sea. Outflow via the Fram Strait is 1.76 Sv in ORCA2, 1.42–1.75 Sv in ORCA05, and 1.46–1.80 Sv in ORCA025, depending on the time period (Table 5).

Relative to the observed CAA outflow of 2.7 Sv (Curry et al., 2014; Straneo and Saucier, 2008), only ORCA05 and ORCA025 simulate similar results. In contrast, ORCA2's simulated CAA outflow is about one ninth of that observed. Likewise, its inflow via the Barents Sea is half of that observed, while the two higher resolution simulations have Barents Sea inflows that are 20% and 40% larger than observed. Yet for inflow through the Bering Strait, it is ORCA2 that is closest to the observed estimate, overestimating it by 30%, while ORCA05 and ORCA025 overestimate it by 60% and 90%. Thus too much Pacific water appears to be entering the Arctic Ocean. All resolutions underestimate the central observational estimate for the Fram Strait outflow by $\sim 12\%$ but still easily fall within the large associated uncertainty range.

Summing up, the net water transport across all four boundaries is not zero. A net Arctic outflow between 0.12 and 0.17 Sv is found for the three model resolutions owing to river inflow and precipitation as well as artifacts-artefacts caused by using monthly averages. In contrast, when the observed water transport estimates at all four boundaries are summed up, there is a net outflow of 1.9 Sv, more than ten times larger. This strong net outflow is also much larger than freshwater input from rivers (of 0.08 Sv) (McClelland et al., 2006) and precipitation (of 0.12 Sv) (Yang, 1999). It can only be explained by uncertainties in the data-based estimates of water transport, which are at least ± 2.7 Sv for the net transport based on the limited uncertainties available for transport across the individual boundaries (Table 5). The excessive central observational estimate for the net outflow might be explained by a data-based estimate for the Barents Sea inflow that is too weak combined with a data-based estimate for the Fram Strait outflow that is too strong, a possibility that is consistent with results from the higher resolution models ORCA05 and ORCA025.

3.1.2 Sea ice

Because sea-ice cover affects the air-sea CO₂ flux and hence anthropogenic carbon concentrations in the ocean, we compare the modeled sea-ice cover to that observed by the U.S. National Snow and Ice Data Center (Walsh et al., 2015). Yearly averages of sea-ice extent are quite similar-agree within 2% between the observations and models. Only in summer are simulated sea-ice concentrations slightly too high (by $0.25\text{--}0.5 \times 10^6 \text{ km}^3$, e.g. 5%). Despite this overall agreement in integrated sea-ice extent, regional-regionally differences are larger. During winter (Fig. 3), all three model configurations marginally-slightly overestimate the sea-ice extent northeast of Iceland and north of the Labrador Sea, while the simulated sea-ice extent in the Barents Sea and the Bering Strait are similar to observations. During summer, the simulated sea-ice extent resembles that observed in the eastern-western Arctic particularly near the Atlantic-Pacific, but all model resolutions slightly overestimate sea-ice extent in the Nordic Seas, north of the Kara-Barents Sea, the Laptev-Kara Sea, and the East-Siberian-Laptev Sea. This overestimation should reduce air-sea CO₂ fluxes locally in these regions. The-Overall, the close model-data agreement for sea-ice extent in terms of the total amount, its trend and seasonal coverage, as well as regional coverage in winter contrasts with the tendency of the models to overpredict sea-ice cover in summer in the highest latitudes of the eastern Arctic.

3.1.3 Atlantic water

In the Arctic Ocean, water temperature is used to help identify water masses, with values above 0°C typically coming from the Atlantic Ocean (Woodgate, 2013). The observed temperature along the 1994 and 2005 sections (Fig. 4) indicates that Atlantic Water (AW) is found between 200 and 1000 m, penetrating laterally below the strongly stratified Arctic Ocean surface waters. In ORCA025, this AW layer is deeper and more diffuse, lying between 500 and 1500 m, thus leading to a cold bias around 500 m and a warm bias around 1000 m. The Beringia station at the boundary between the Barents Sea and the Nansen basin indicates AW lies between 200 m (2.5°C) and the seafloor at 1000 m (0°C). Conversely in the same location in ORCA025, model temperatures remain above 1.5°C throughout the water column. That lower maximum temperature and weaker vertical gradient suggests that when ORCA025's Atlantic water enters the Arctic Ocean through the Barents Sea it is too diffuse, being well mixed throughout the water column. Weaker maxima in the-ORCA025's simulated temperature relative to observations

are also found further west in the Canada basin along both sections. There observed temperatures reach maxima of 1.1°C, while ~~ORCA025's~~ its simulated maxima reach only 0.5°C.

The ~~other two resolutions represent Atlantic water circulation more poorly~~ two lower resolutions represent lateral invasion of AW less successfully than does ORCA025. Both simulations show water with temperatures higher than 0°C only at the southern end of the Nansen basin. Vertically, these water masses are situated around 400 m for ORCA2 and between 200 and 1300 m for ORCA05.

3.2 CFC-12

Simulated CFC-12 was compared among the three resolutions and with observations, focusing first on basin-scale tendencies based on vertical profiles of the distance-weighted means along the Beringia 2005 section (Fig. 5). ~~That~~ This comparison reveals that among resolutions, simulated CFC-12 concentrations differ most between 400 and 1900 m; conversely, above and below that intermediate zone, simulated average profiles are nearly insensitive to resolution. In that intermediate zone and above, simulated concentrations are also generally lower than observed. The only exception is the top 100 m ~~of in~~ the Canada basin where all resolutions overestimate observed ~~values~~ concentrations by 10%. Between 200 and 400 m, all resolutions underestimate observations by ~50%. Below 400 m, the ORCA2 CFC-12 concentrations decline quickly to zero (~~by~~ ~1000 m), while the ORCA05 and ORCA025 concentrations continue to increase ~~both being by~~, both being 15% greater at 900 m than at 400 m. Below that depth, the ORCA05 concentrations decline quickly reaching zero at 1350 m, while ORCA025 concentrations remain above 1 pmol kg⁻¹ until 1400 m. Between 1100 and 1500 m, average CFC-12 concentrations along the Beringia section in ORCA025 are larger than observed by up to ~10% at 1300 m. This overestimation of CFC-12 by ORCA025 reaches up to 40% in the Canada and Makarov basins. Below 1900 m, the simulated concentrations are essentially zero, while the observations are slightly higher (0.12 pmol kg⁻¹). For comparison, the reported detection limit for CFC-12 for the Beringia 2005 expedition is 0.02 pmol kg⁻¹ (Anderson et al., 2011).

Given the closer overall agreement of the ORCA025 simulated CFC-12 to the observations, let us now focus on its evaluation along the 1994 and 2005 sections (Fig. 6). On the Atlantic end of the Beringia 2005 section, where water enters the Nansen basin from the Barents Sea, the water column in ORCA025 appears too well mixed, having CFC-12 concentrations that remain above 2.0 pmol kg⁻¹. Conversely, observed CFC-12 is less uniform, varying from 2.8 pmol kg⁻¹ at the surface to 1.3 pmol kg⁻¹ in bottom waters at 1000 m, thereby indicating greater stratification. ~~The same~~ Similar contrast in stratification was deduced from modeled and observed temperature profiles at the same location (Sect. 3.1.3). On the other side of the Arctic in the Canada basin, there are observed local chimneys of CFC-12 where concentrations remain at about 2.0 pmol kg⁻¹ from near the surface down to 1000 m, particularly along the 1994 section. These chimneys suggest localized mixing that is only barely apparent in ORCA025 (Fig. 6). Such localized features are absent at lower resolution.

~~Lastly, we calculate~~ The CFC-12 inventories were also calculated along the two sections, integrated over depth and distance (Table 7). Depending on the expedition, ORCA025 underestimates the observed CFC-12 section inventories by ~~13-18~~ 13-18%, ORCA05 by ~~31-38~~ 36-38%, and ORCA2 by ~~61-64~~ 47-61%.

3.3 Anthropogenic carbon inventories and concentrations

Simulated global ocean C_{ant} inventories are 152 Pg C in ORCA2, 146 Pg C in ORCA05, and 148 Pg C in ORCA025 in 2008, ~~all of which account after accounting~~ for corrections for ~~an the~~ earlier starting date ~~from of 1765 using~~ our perturbation simulations (P1765–P1870). The ~~corrections are correction is~~ similar for each resolution, e.g., 24–25 Pg C in 1995, and ~~are~~
5 ~~is~~ consistent with our biogeochemical model simulation strategy (all three resolutions initialized with the ORCA05 output in 1958). Furthermore, these model-based corrections ~~are much like the~~ 29 ± 5 ~~fall within the~~ 29 ± 5 Pg C correction calculated for the same ~~1765–1995–1765–1995~~ period with a data-based approach (Bronselauer et al., 2017). For the 1765–2008 period, the data-based global C_{ant} inventory estimate from (Khatiwala et al., 2009) is 140 ± 24 Pg C, the ~~uncertainty~~ range of which encompasses the results from all three model resolutions.

10 In the Arctic Ocean, the corrected modeled C_{ant} inventories range from 1.9 to 2.5 Pg C in 2002 and from 2.0 to 2.6 Pg C in 2005, ~~in each case with the low~~ from ORCA2 ~~to and the high from~~ ORCA025 (Table 6 and Fig. 2). These simulated ~~basin-wide Arctic Ocean~~ C_{ant} ~~basinwide Arctic Ocean~~ inventories were compared to the TTD-based estimates of anthropogenic carbon from (1) the GLODAPv2 assessment (Lauvset et al., 2016) normalized to the year 2002 and (2) the Tanhua et al. (2009) assessment normalized to 2005. The data-based assessment from GLODAPv2 suggests that 2.9 Pg C of
15 anthropogenic carbon was stored in the Arctic Ocean in 2002, while that from Tanhua et al. suggests that 2.5–3.3 Pg C was stored there in 2005. In 2002, the upper limit of the ~~of the modeled~~ C_{ant} ~~modeled~~ C_{ant} inventory range remains 0.4 Pg C lower than the GLODAPv2 data-based estimate. ~~In 2005, the upper limit of the model range falls just , but the ORCA025 result in~~
20 ~~2005 falls~~ within the data-based uncertainty range of Tanhua et al. (2009). As for the global estimates, the ~~Arctic Ocean~~ C_{ant} ~~modeled Arctic Ocean~~ inventories include corrections for the late starting date of the biogeochemical simulations. ~~These~~
~~This~~ correction is 0.4 Pg C in 2005 for each ~~resolution (Table of the three resolutions (Table 6))~~.

The differences in ~~basin-wide~~ ~~basinwide~~ inventory estimates were further studied by comparing vertical profiles of C_{ant}
 C_{ant} from the models to those from the GLODAPv2 data-based estimates (Fig. 7). Surface concentrations in ORCA05 and ORCA025 are up $\sim 35\%$ larger ($+12 \mu\text{mol kg}^{-1}$) than the data-based estimate, whereas the ORCA2 concentration is $\sim 22\%$
25 larger ($+7 \mu\text{mol kg}^{-1}$). ~~Moving downward, by~~ ~~By~~ 150 m, the simulated concentrations in all resolutions have dropped below the data-based estimates and remain so, except for ORCA025, down to the ocean bottom. Data-model differences are largest at 400 m, with all resolutions underestimating data-based C_{ant} estimates by up to $\sim 28\%$ ($9 \mu\text{mol kg}^{-1}$). Below that depth, results from the three resolutions differ more. The ~~ORCA2~~ C_{ant} ~~concentration decreases monotonically to~~ C_{ant} ~~concentration~~
30 ~~in ORCA2 decreases monotonically reaching~~ $11 \mu\text{mol kg}^{-1}$ at 1000 m and ~~to 0 mol kg⁻¹ at essentially zero concentration by~~
~~reaching zero concentration by 1400 m~~. In ORCA05, C_{ant} concentrations decrease slowly to $19 \mu\text{mol kg}^{-1}$ at 1000 m, below which they ~~decrease rapidly, essentially to 0 mol kg⁻¹~~ ~~decline rapidly, reaching zero~~ at 2300 m. Only ~~in~~ ORCA025 ~~increases~~
~~do~~ C_{ant} ~~concentrations increase~~ again below 400 m, reaching a local maximum ~~in~~ C_{ant} at 900 m, an increase that causes the ORCA025 results to exceed data-based estimates by up to $2 \mu\text{mol kg}^{-1}$ ($\sim 11\%$) at 1100 m. ~~A similar maximum and~~
~~excess are also seen in the CFC-12 profile for ORCA025 as is the minimum around 400 m (Fig. 5)~~. Below 1500 m, ~~the~~

ORCA025 C_{ant} concentrations decline quickly, essentially reaching zero at 2300 m. Conversely, data-based anthropogenic carbon concentrations remain at roughly a constant C_{ant} concentrations remain roughly constant at $6 \mu\text{mol kg}^{-1}$ all the way down to the seafloor. The main ~~Thus the largest vertically integrated~~ differences between ORCA025 and data-based estimates are ~~thus~~ found in the deep Arctic Ocean below 1600 m. ~~An analogous simulated local maximum and the underestimation near 400 m was also seen for CFC-12 (Fig. 5). Yet unlike for C_{ant} , CFC-12 results differ below 2000 m, where observed CFC-12 concentrations are proportionally much smaller than those above.~~

3.4 Anthropogenic carbon budget

~~We calculated the budget for C_{ant} from 1960 to 2012. For the budget of C_{ant} , we focused on the final decades over which the model resolutions differed~~ (Tables 5 ~~and~~ and 6). During ~~this period, the C_{ant} 1960 to 2012, the C_{ant}~~ inventory in ORCA025 increased by 1.98 Pg C, 80% of which is stored in the four major Arctic Ocean basins: the Nansen Basin (0.30 Pg C), the Amundsen Basin (0.34 Pg C), the Makarov Basin (0.33 Pg C) and the Canada Basin (0.61 Pg C). Although the Canada Basin C_{ant} C_{ant} inventory increased most, its volume is larger so that its average C_{ant} C_{ant} concentration increased less than in the other basins (Fig. 7). Out of the total inventory stored in the Arctic Ocean during ~~these five decades, that time, only~~ about one-fourth (0.48 Pg C) entered the Arctic Ocean via air-sea flux, most of which was transferred from atmosphere through the surface of the Barents Sea (Fig. 8). The remaining 75% (1.50 Pg C) entered the Arctic Ocean via lateral transport. This net lateral influx is the sum of the fluxes (1) from the Atlantic ~~through the Fram Strait (-0.06 to the Barents Sea (1.98 Pg C)~~, (2) from the ~~Atlantic to the Barents Sea (-1.98 Pacific through the Bering Strait (1.03 Pg C)~~, (3) ~~from the Pacific through the Bering Strait (-1.03 to the Atlantic via the Fram Strait (-0.06 Pg C)~~, and (4) to the Atlantic via the CAA (-1.50 Pg C). Summed up, the net lateral inflow of carbon across the four boundaries is 1.45 Pg C. This lateral flux computed from monthly mean C_{ant} C_{ant} concentrations and flow fields is 0.05 Pg C (~3%) ~~is~~ smaller than the lateral flux computed from the change in inventory minus the cumulative air-sea flux (Fig. 8). Within the Arctic, coastal regions typically exhibit net lateral losses, while the deep basins exhibit net lateral gain. The largest lateral loss occurs in the Barents Sea, where the cumulative air-sea flux of C_{ant} C_{ant} is also largest (Fig. 8).

The budget of C_{ant} C_{ant} changes notably with resolution. Higher resolution results in more simulated C_{ant} C_{ant} being stored in the Arctic region, with increases in both the cumulative air-sea flux and lateral transport. The C_{ant} C_{ant} inventory change from 1960 to 2012 nearly doubles with the resolution increase between ORCA2 and ORCA025 (from 1.08 to 1.98 Pg C). Out of that additional C_{ant} C_{ant} , 93% is found between 300 and 2200 m with the maximum being located at 1140 m. The remaining 7% is located ~~between the surface and in the upper~~ 300 m (Fig. 7). ~~Besides these differences in the vertical partitioning of stored C_{ant} , resolution~~ Resolution also affects regional partitioning of C_{ant} C_{ant} (Figs. 7 and 8). When refining resolution from ORCA2 to ORCA05, the Arctic Ocean C_{ant} C_{ant} inventory increases by 0.47 Pg C, 72% of which occurs in the ~~two~~ Eurasian basins: the Nansen basin (0.19 Pg C) and Amundsen basin (0.15 Pg C) basins. Another 23% of that increase occurs in the ~~two~~ Amerasian basins: the Makarov basin (0.06 Pg C) and Canada basin (0.05 Pg C) basins. Coastal regions account for only 5% of the total inventory increase. In contrast, the subsequent resolution enhancement between ORCA05 and ORCA025 results in little increase in inventory in the Eurasian basins (0.03 Pg C) but much more in the Amerasian basins (0.37 Pg C).

As resolution is refined between ORCA2 and ORCA025, the Arctic $C_{\text{ant}}C_{\text{ant}}$ inventory increases as a result of a 66% increase in the air-sea flux (+0.19 Pg C) and a 90% increase in the lateral flux (+0.71 Pg C). Thus the relative contribution of the lateral flux increases from 73% to 76%. Changing model resolution also affects the pathways by which $C_{\text{ant}}C_{\text{ant}}$ enters the Arctic Ocean (Table 5). The most prominent change occurs in the CAA. From ORCA2 to ORCA025, the net outflow of $C_{\text{ant}}C_{\text{ant}}$ through the CAA increases sevenfold (from -0.22 to -1.50 Pg C). Other notable changes include (1) the net ~~transport-outflow~~ through the Fram Strait declining ~~from a sizable (-0.59 Pg C) to a slight net outflow (12-fold from -0.74 to -0.06 Pg C)~~, (2) the inflow through the Barents Sea increasing by 150% (from 0.79 to 1.98 Pg C), and (3) the inflow of $C_{\text{ant}}C_{\text{ant}}$ through the Bering Strait increasing by 39% (from 0.74 to 1.03 Pg C).

4 Discussion

10 4.1 CFC-12

The simulated CFC-12 in ORCA025 underestimates observed concentrations between 100 and 1100 m, ~~overestimates them slightly overestimates them on average~~ between 1100 and 1500 m, and again underestimates the low observed concentrations below 1500 m. ~~Temperature~~The temperature sections suggest that excess simulated CFC-12 between 1100 and 1500 m is due to a vertical displacement of inflowing Atlantic water, which descends too deeply into the Arctic (Fig. 4). Such vertical displacement would indeed reduce simulated CFC-12 concentrations above 1000 m and enhance them between 1100 and 1500 m. Yet the underestimation of integrated CFC-12 mass above 1100 m is larger than the overestimation below 1100 m. Thus vertical displacement of Atlantic water cannot provide a full explanation. Simulated CFC-12 concentrations above 1100 m could also be too low because ventilation of subsurface waters is ~~probably~~ too weak, an hypothesis that is consistent with the simulated vertical gradients in both temperature and CFC-12 that are too strong between 100 and 1100 m.

20 4.2 Anthropogenic carbon

~~Relative to Vertical profiles of C_{ant} and CFC-12, simulated deep C_{ant} in ORCA025 underestimates observational estimates by proportionally much more. Both tracers have similarly shaped profiles, but the data-based C_{ant} profile differs from the observed CFC-12 profile below 1500 m are similar.~~ Above 1000 m, ORCA025 underestimates data-based estimates of $C_{\text{ant}}C_{\text{ant}}$ as well as observed CFC-12 owing to weak ventilation in the model. Between 1000 and 1500 m, simulated $C_{\text{ant}}C_{\text{ant}}$ and CFC-12 in ORCA025 ~~are exhibit local maxima, which make them on average slightly~~ higher than data-based and observed concentrations. ~~The local maxima for simulated CFC-12 and C_{ant} in that depth range~~These local maxima can be explained by the ~~excessively deep penetration of~~ simulated Atlantic water masses, ~~which are~~ rich in both tracers. ~~, being too deep. However, the slight overestimation between 1000 and 1500 m is much smaller than the underestimation between 200 and 1000 m.~~ Below 2000 m, simulated $C_{\text{ant}}C_{\text{ant}}$ largely underestimates data-based estimates. The ~~data-based C_{ant} concentrations remain higher than those simulated, with an offset of $\sim 6 \mu\text{mol kg}^{-1}$ from 2000 m to the ocean floor (18% of the surface concentration), while observed low simulated C_{ant} stems from too little deep-water formation in the model as~~

indicated by the absence of simulated CFC-12 concentrations are relatively closer to zero, dropping to 0.1 pmol kg^{-1} below 2000 m (3% of the surface concentration). Hence the data-based estimates of C_{ant} , which are not measured directly, appear to be too large below 2000 m in the Arctic Ocean.

There are reasons to suspect that the GLODAPv2 estimate using the TTD method may overestimate C_{ant} in the deep Arctic. First, the water mass mean ages below 2000 m are shown to be of the order of 300 to 400 years (Tanhua et al., 2009; Schlosser et al., 1994), older than the atmospheric CO_2 perturbation. Second, the TTD method estimates C_{ant} concentrations ($\sim 5 \text{ } \mu\text{mol kg}^{-1}$), even if the, an absence that contrasts with the observed CFC-12 concentrations approach zero (Vaugh et al., 2006), which demonstrates the large uncertainty of the method when dealing with old water masses. To assess the maximum error associated with these potentially excessive deep TTD C_{ant} estimates, we recalculated the C_{ant} budget after zeroing out the C_{ant} below 2000 m. Doing so reduces the data-based inventory of C_{ant} in the Arctic Ocean in 2002 by 10%. Applying the same 10% relative decrease to both the upper and lower limits of the data-based range from Tanhua et al. (2009) leads to a minimum C_{ant} inventory of 2.2–3.0 Pg C in 2005. Simulated inventories from both ORCA05 and ORCA025 are within this lower limit. concentrations that remain detectable all the way down to the ocean floor (Fig. 5).

Meanwhile, we also consider that the simulated Arctic C_{ant} . A second reason for the low simulated Arctic C_{ant} inventory in ORCA025 may well be too low because is that it was initialized with ORCA05 results in 1958. Had ORCA025 been initialed instead initialized in 1765, which was not computationally feasible, its simulated inventory would probably be larger. Although we cannot assess this affect directly, we can do so indirectly by running tests at lower resolution and noting how trends differ between model resolutions after 1958. First, let us estimate how that same 1958 initialization affects the ORCA2 results, by taking the difference in simulated C_{ant} inventory between (1) the ORCA2 biogeochemical simulation from 1958 to 2012 initialized with be larger, given that both C_{ant} and CFC-12 storage for ORCA025 are larger than those for ORCA05 in 1957 minus (over 1958–2012. That hypothesis is consistent with our finding that ORCA2* (complete simulation at 2°) without branching from the ORCA2 perturbation simulation from 1765 to 2012. That difference is -0.4 Pg C in 2005. Next let us assume that there is a symmetry during 1765-2005 about the ORCA05 result 0.5° configuration) takes up less C_{ant} than does ORCA2 (0.5° until 1958 then 2° afterwards), which is in line with ORCA2 being lower and ORCA025 being higher as seen for the simulated period after 1958 (Figure 2 and Table 6). We infer then that ORCA025 Arctic C_{ant} inventory in 2005 would be $\sim 0.4 \text{ Pg C}$ larger had it been run initialized in 1765 rather than with the taking up less C_{ant} and CFC-12 than ORCA05 (Figs. 2, 5, and 7). The initialization of ORCA2 with ORCA05 output in 1958. If so, the ORCA025 C_{ant} inventory in 2005 would increase from 2.6 to 3.0 Pg C, pushing it to closer to the center of the data-based range of 2.5–3.3 Pg C from Tanhua et al. (2009). After correcting both 1958 mainly affects C_{ant} storage between 1000 and 2000 m, the same depth range over which differences in simulated CFC-12 concentrations are largest between ORCA2 and ORCA025 for their ORCA05. Nevertheless that 1958 initialization with ORCA05 output, has little effect on subsequent changes in C_{ant} storage, cumulative lateral flux, and air-sea flux (Fig. 9). Rather it is the changes before 1958 that dominate the difference between ORCA2 and ORCA2*.

All our model configurations underestimate C_{ant} concentrations in the deep waters of the Arctic Ocean based on the CFC-12 model evaluation. The same conclusion is drawn from comparing simulated to data-based estimates of C_{ant} . However, results from different data-based approaches to estimate C_{ant} can differ substantially in the deep ocean (e.g. Vázquez-Rodríguez et al., 2009).

~~Furthermore, the model range for the Arctic C_{ant} inventory would then be 1.6–3.0 Pg C in 2005, emphasizing even more the need to be beyond coarse-resolution models in the Arctic~~ TTD approach typically produces the highest values in deep waters due to its assumption of constant air-sea disequilibrium (Khatiwala et al., 2013). Hence applying other data-based approaches to assess the Arctic Ocean inventory of C_{ant} inventory would eventually help to further constrain uncertainties.

5 4.3 Lateral flux

In our model, about three-fourths of the net total mass of C_{ant} that accumulates in the Arctic Ocean enters laterally from the Atlantic and Pacific Oceans, independent of model resolution. Our simulated lateral fluxes of C_{ant} in ORCA025 were compared to data-based estimates from studies that multiply C_{ant} data-based C_{ant} concentrations (TTD data-based estimates) along the Arctic boundaries by corresponding observation-based estimates of water transport.

10 The simulated lateral transport of C_{ant} in ORCA025 generally agrees with data-based estimates within their large uncertainties. These uncertainties result from uncertainties in data-based estimates of C_{ant} and from uncertainties in observational constraints on water flow, which also varies interannually (Jeansson et al., 2011). For the Fram Strait, Jeansson et al. (2011) estimated a net C_{ant} outflux (from the Arctic) of $1 \pm 17 \text{ Tg C yr}^{-1}$ in 2002, while for 2012 Stöven et al. (2016) estimate an outflux of 12 Tg C yr^{-1} but without without indicating uncertainties. For the same years, ORCA025 simulates a net outflux of
15 8 Tg C yr^{-1} in 2002 but a net inflow (to the Arctic) of 5 Tg C yr^{-1} in 2012. Both model and data-based estimates vary greatly between 2002 and 2012. Across the Barents Sea Opening, there is a consistent net inflow from the Atlantic to the Arctic Ocean, i.e., $41 \pm 8 \text{ Tg C yr}^{-1}$ in 2002 for the data-based estimate (Jeansson et al., 2011) and $(50 \text{ Tg C yr}^{-1})$ for ORCA025 in the same year.

More recently, Olsen et al. (2015) added data-based estimates of lateral fluxes at of C_{ant} across the two other major Arctic
20 Ocean boundaries, completing the set of four boundaries that define the perimeter. They estimate an inflow a C_{ant} influx of $\sim 18 \text{ Tg C yr}^{-1}$ from the Pacific through the Bering Strait and an outflow a C_{ant} outflux through the CAA of $\sim 29 \text{ Tg C yr}^{-1}$, both for the 2000s. For the same time period, ORCA025 simulates one-half 50% more inflow through the Bering Strait ($\sim 27 \text{ Tg C yr}^{-1}$) and 24% more outflow through the CAA ($\sim 36 \text{ Tg C yr}^{-1}$). The larger Bering-Strait C_{ant} inflow C_{ant} influx in ORCA025 is consistent with its overestimated Bering-Strait water inflow (Table 5, Section 3.1.1). Integrating over all four
25 lateral boundaries, Olsen et al. found a total net C_{ant} influx of $\sim 29 \text{ Tg C yr}^{-1}$, which is 24% less than that simulated in ORCA025 averaged over 2000–2010 ($\sim 38 \text{ Tg C yr}^{-1}$). Olsen et al. did not provide uncertainties, but the uncertainty of their net lateral flux estimate is at least $\pm 18 \text{ Tg C yr}^{-1}$ based on the data-based transport estimates at the two other Arctic boundary sections where uncertainties are available (Table 5).

Weighing in at about one-third one-quarter of the lateral flux is the simulated air-sea flux of C_{ant} in ORCA025 of
30 10 Tg C yr^{-1} in 2005. when both are averaged over 2000–2010. That simulated estimate is only about 40% of the data-based estimate of 26 Tg C yr^{-1} from Olsen et al. (2015). Although no uncertainty is provided with that data-based air-sea flux estimate, it too must be at least $\pm 18 \text{ Tg C yr}^{-1}$ given that it is calculated as the difference between the data-based storage estimate (Tanhua et al., 2009) and Olsen et al.'s data-based net lateral flux. The simulated air-sea flux of C_{ant} falls within that assigned uncertainty range for the data-based estimate. In any case, both the model and data-based estimates suggest that the air-sea flux plays a

~~minor role in the of C_{ant} is not the dominant contributor to the~~ anthropogenic carbon budget of the Arctic Ocean, respectively representing ~~24-21~~ and 47% of the total ~~C_{ant} input C_{ant} input averaged over 2000–2010~~. For both, the lateral flux dominates.

4.4 Model Resolution

Basin inventories of ~~simulated~~ anthropogenic carbon differ ~~between model configurations~~ because of how resolution affects their volume, bathymetry, circulation patterns, and source waters. Much of the water in the Nansen and Amundsen basins has entered laterally from the Atlantic Ocean through the Fram Strait and the Barents Sea (Jones et al., 1995). Water inflow through the Barents Sea increases by 150% when ~~changing moving~~ from ORCA2 to ORCA05 but only by 20% more between ORCA05 and ORCA025. Water inflow in those two higher resolution models is also closer to observational estimates. ~~With increasing~~ ~~Along with the increase in~~ water inflow, ~~the inflow of C_{ant} is also increased. Although more C_{ant} is entering the Arctic~~ ~~Ocean~~ ~~higher resolution also increases the lateral influx of C_{ant} . Yet despite this increase in the C_{ant} lateral influx,~~ the air-sea ~~C_{ant} C_{ant}~~ flux into the Arctic Ocean ~~also~~ increases with resolution. This ~~apparent contradiction finding~~ can be explained by two ~~mechanism mechanisms~~: (1) ~~Higher higher~~ resolution increases the ~~inflow of C_{ant} influx of C_{ant}~~ through the Fram Strait, which ~~is mainly occurring mainly occurs~~ in subsurface currents and ~~therefore does not substantially impact surface C_{ant} concentrations nor hence~~ ~~thus does not greatly affect surface C_{ant} concentrations nor~~ air-sea exchanges of ~~C_{ant} C_{ant}~~ , and (2) higher resolution enhances deep-water formation, mainly in the Barents Sea, which reduces surface ~~C_{ant} C_{ant}~~ and thus enhances the air-to-sea flux of ~~C_{ant} C_{ant}~~ . Although the air-sea flux ~~of C_{ant}~~ increases slightly, the larger lateral water fluxes in ORCA05 and ORCA025 mainly explain their higher ~~C_{ant} C_{ant}~~ concentrations in the Nansen and Amundsen basins. Some of this inflowing water continues to flow further along the slope, across the Lomonosov ridge into the Makarov basin, and then across the Mendeleev ridge into the Canada basin. Yet how well models simulate that flow path depends on lateral resolution. Between ORCA2 and ORCA05, basin ~~C_{ant} C_{ant}~~ inventories increase by 16% in the Canada basin (+0.05 Pg C) and by 40% the Makarov basin (+0.06 Pg C). But between ORCA05 to ORCA025, increases are two to ~~four five~~ times greater: +0.25 Pg C in the Canada basin and +0.12 Pg C in the Makarov basin (Sect. 3.4). The change from ORCA2 to ORCA05 ~~seems to mainly mainly seems to~~ improve lateral exchanges with adjacent oceans, while the change from ORCA05 to ORCA025 improves ~~inner-Arctic interior Arctic~~ Ocean circulation.

As the increase from ORCA05 to ORCA025 stems from finer, more realistic representation of lateral transport within the Arctic, it would appear that eddying ocean models may be needed to adequately simulate the interior circulation in terms of its effect on ~~C_{ant} C_{ant}~~ storage in the Arctic Ocean. In the Canada basin, such lateral inflow may not be the only source of ~~C_{ant} C_{ant}~~ . Another major source appears to come from density flows along the continental slope, driven by brine rejection from sea-ice formation over the continental shelves (Jones et al., 1995). A signature of this source in the observed sections may be the chimneys of constant CFC-12 concentration from the surface to about 1000 m in the Canada basin, features for which only ORCA025 exhibits any such ~~signature indication~~, albeit faint. To adequately model lateral exchanges of ~~C_{ant} C_{ant}~~ in the Arctic Ocean, at least a resolution comparable to that used in ORCA05 may be needed, while resolutions comparable to that in ORCA025 or above may well be required to begin to capture the effects from density flows along the slope. As a consequence

of the deficient representation of these density flows, we would expect to see an increase in $C_{\text{ant}}-C_{\text{ant}}$ when using even higher resolution.

~~Similar to our the results in the Arctic Ocean, improving circulation with~~ Improved modeled circulation from higher model resolution has also been shown to be ~~the key driver for an improved representation of~~ critical to improve simulated anthropogenic tracers in the Southern Ocean (Lachkar et al., 2007) ~~or and simulated~~ oxygen concentrations in the ~~tropics-tropical~~ Atlantic (Duteil et al., 2014).

4.5 CMIP5 comparison

For wider perspective, we compared ~~the results from our~~ forced NEMO-PISCES simulations to ~~those from~~ nine ocean biogeochemical models that were coupled within different earth system modeling frameworks as part of CMIP5 (Fig. 9). When the CMIP5 models are compared to the ~~corrected~~-data-based estimate of the $C_{\text{ant}}-C_{\text{ant}}$ inventory (Sect. 4.2) $C_{\text{ant}}-C_{\text{ant}}$ inventory, only the MIROC-ESM with its inventory of 2.7 Pg C ~~fall-falls~~ within the data-based uncertainty estimate (2.5 to 3.3 Pg C in 2005). ~~Nearby is the~~ The next closest CMIP5 models are NorESM1-ME and HadGEM2-ES, which fall below the lower limit of the data-based range by 0.1 and 0.5 Pg C, ~~receptively. Further off are~~. Then come the MPI-ESM and GFDL-ESM models with their $C_{\text{ant}}-C_{\text{ant}}$ inventories in 2005 that are 0.9 to 1.5 Pg C lower than the lower limit. The lowest CMIP5 estimates are from both versions of the IPSL model whose inventories reach only $\sim 20\%$ of the lower limit of our revised data-based range. Adjusting all the CMIP5-model Arctic inventories upward by ~ 0.4 Pg C to account for their late start date in 1850, as we did for our three simulations, would place two of them (MIROC-ESM and NorESM1-ME) above the lower boundary of our revised the data-based uncertainty estimate, and another (HadGEM2-ES) just 0.1 Pg C below this lower boundary. ~~For the cumulative air-sea flux between 1960 and 2012, for which there is no data-based constraint, all models fall between 0.3 and 1.2 Pg C. Lateral fluxes~~ Lateral fluxes over 1958–2012 also vary between CMIP5 models, from an outflow of 0.3 Pg C in the IPSL-CM5A-LR model and an inflow of 1.1 Pg C in the MIROC-ESM model. Only the first three CMIP5 models mentioned above exhibit large net inflow of $C_{\text{ant}}-C_{\text{ant}}$ into the Arctic basin (between 0.7 and 1.1 Pg C ~~from during~~ 1960–2012), a condition that appears necessary to allow a model to approach the estimated data-based inventory range. Indeed, the six other CMIP5 models have lower lateral fluxes (-0.5 to 0.5 Pg C) and simulate low $C_{\text{ant}}-C_{\text{ant}}$ storage in 2005.

What is perhaps most surprising are the large differences between our forced ORCA2* model and the IPSL-CM5A-LR and IPSL-CM5A-MR ESMs. All three of those models use ~~ORCA2~~ the same coarse-resolution ocean model, although both ESMs rely on an earlier version with a different vertical resolution (31 instead of 46 vertical levels). ~~That~~ The contrast in vertical resolution may explain part of the large differences in inventory (~~1.5 difference in inventories~~ 1.3 Pg C for our forced version that is not corrected for the late starting date vs. 0.3–0.6 Pg C for the two coupled versions) but the forcing and different model parameters could just as well be responsible. Thus lateral could also play a role. Lateral resolution is not the only factor when aiming to provide realistic simulations of $C_{\text{ant}}-C_{\text{ant}}$ storage and lateral transport in the Arctic. Sensitivity studies testing other potentially critical factors are clearly merited.

4.6 Effect on aragonite saturation state

Given that ~~C_{ant}~~ simulated C_{ant} is affected by lateral model resolution, so must be simulated ocean acidification. The aragonite saturation state (Ω_A) was computed for each resolution from the historical run's C_T , A_T , T , S , P_T , and Si_T , after correcting C_T and A_T for drift based on the control run. The higher concentrations of ~~C_{ant}~~ C_{ant} in the ORCA05 and ORCA025 simulations reduces Ω_A between 1960 and 2012 by more than twice as much as found with ~~the ORCA2 model~~ during the same period (Fig. 10). These differences translate into different rates of shoaling for the aragonite saturation horizon (ASH), i.e., the depth where $\Omega_A = 1$. During 1960–2012, the ASH shoals by ~ 50 m in ORCA2, while it shoals by ~ 150 m in ORCA05 and ~ 210 m in ORCA025. Thus model resolution also affects the time at which waters become undersaturated with respect to aragonite with higher resolution producing greater shoaling.

Although basin-wide mean surface Ω_A does not differ among resolutions, there are regional differences such as over the Siberian shelf (Fig. 11). ~~While the~~ The minimum Ω_A in that region reaches 0.9 in ORCA2, while it drops to 0.3 in ORCA05 and 0.1 in ORCA025. That lower value in ORCA025 is more like that observed, e.g., down to 0.01 in the Laptev Sea (Semiletov et al., 2016). As these ~~low extremes~~ lows in Ω_A are extremely local, they cannot be expected to be captured in coarse-resolution models (~~such as ORCA2~~). ~~Higher resolution~~. Higher resolution models are needed in the Arctic to assess local extremes not only in terms of ocean acidification but also other biogeochemical variables.

5 Conclusions

Global-ocean biogeochemical model simulations typically have coarse resolution and tend to underestimate the mass of ~~C_{ant}~~ C_{ant} stored in the Arctic Ocean. Our sensitivity tests suggest that more realistic results are offered by ~~higher resolution~~ higher resolution model configurations that begin to explicitly resolve ocean eddies. Our ~~highest resolution model falls~~ high resolution model simulates an Arctic C_{ant} inventory of 2.6 Pg C in 2005, falling within the uncertainty ~~range of Tanhua et al. (2009)'s~~ from the data-based ~~estimate for C_{ant} storage in the Arctic Ocean estimates~~ (2.5–3.3 Pg C in 2005). ~~Yet that data-based range may need to be adjusted downward. Data-based C_{ant} concentrations below 2000 m remain at about $6 \mu\text{mol kg}^{-1}$, while observed~~). That model estimate should be considered a lower bound because it generally underestimates CFC-12 concentrations ~~upon which they are based are close to negligible, being proportionally much smaller relative to near-surface concentrations. A lower limit is estimated by zeroing out the C_{ant} concentrations below 2000 m in the GLODAPv2 climatology (Lauvset et al., 2016). Thus, the lower limit of the~~. Thus it essentially confirms the lower bound from the data-based ~~estimates for Arctic Ocean C_{ant} storage would be reduced by 10% to 2.2 Pg C in 2005. That lower limit encompasses the adjusted simulated basin-wide inventories from the two higher resolutions, ORCA05 and ORCA025 (2.4 and 2.6 Pg C). At the same time, our highest resolution inventory is likely an underestimation as it was initialized~~ estimates, which are based on CFC-12 derived C_{ant} concentrations that are not without uncertainties, particularly in the deep Arctic Ocean where measured CFC-12 concentrations are small. The high-resolution model would have simulated a higher Arctic C_{ant} inventory had computational resources been available to use it throughout the entire industrial era rather than initializing it in 1958 with ~~ORCA05 results from 1765–1957. Details in~~ results from the intermediate resolution model (ORCA05), in which the penetration of CFC-12 and

C_{ant} into Arctic intermediate waters is weaker. The largest source of differences in C_{ant} inventory between resolutions is due to the increasing ventilation of intermediate waters as model resolution is refined, as revealed by CFC-12 and C_{ant} model-data based comparison differ, e. g., with the comparison. The highest resolution model, ORCA025 results underestimating C_{ant} , still underestimates the C_{ant} data-based estimates at around 400 m and overestimating slightly overestimates them at around 1300 m.

5 ~~That deeper model~~The deeper overestimate appears due to excessive penetration of C_{ant} -rich Atlantic water. The shallower model underestimates may be partly due to inadequate representation of ventilation of intermediate waters via down-slope flows that are driven by brine formation over the Arctic's enormous continental shelf, a transport process that is notoriously difficult to represent in z-coordinate models, especially at lower resolution.

10 Data-based estimates of C_{ant} in the Arctic Ocean might be improved by testing the TTD method in the model, using the same approach but with modeled CFC-12 and temperature and then comparing the resulting calculated C_{ant} to the directly simulated C_{ant} . With a series of those calculations, the parameters of the TTD approach (Δ/Γ) could be varied and the best ratio selected for the closest match between calculated and simulated C_{ant} . Then that chosen ratio could be applied to the observed CFC-12 rather than using the default ratio of $\Delta/\Gamma = 1$. We leave this effort for future work.

15 Our forced ocean simulations suggest that Arctic Ocean storage of C_{ant} is driven mostly by net lateral inflow, the total input of which is about three times that from the air-sea flux. That 3:1 ratio varies little with resolution because both the lateral flux and the air-sea flux both increase as resolution is refined. Lateral fluxes are generally less dominant in the CMIP5 models but are also highly inconsistent both in magnitude and in the lateral:but its magnitude varies greatly as does its ratio relative to the air-sea fluxratio. Some CMIP5 models even simulate net lateral outflow of C_{ant} and unrealistically low C_{ant} inventories. The only CMIP5
20 models that succeed in reaching the lower limit of the data-based C_{ant} inventory range are those that have a large net lateral input. Unfortunately, the causes of the CMIP5 model differences remain unclear as is often the case when comparing models having many differences. We expect that most Most of the CMIP5 models have not appear to not have been evaluated in terms of their ability to simulate realistic lateral water transport at the boundaries of the Arctic Ocean, which is fundamental to simulating realistic C_{ant} but may be problematic given their coarse resolution. The next phase of CMIP (CMIP6) plans
25 to include is ongoing and includes CFC-12 and related transient tracers, which will help weigh simulated results for C_{ant} .

As the mass of simulated anthropogenic carbon in the Arctic Ocean increases with resolution, so does the simulated acidification. For instance, during 1960–2012, the average ASH in the Arctic shoals four times faster in ORCA025 than in ORCA2. Higher resolution is also needed to capture local extremes. Although higher horizontal resolution appears necessary to improve
30 fine-scale future projections of Arctic Ocean acidification, the Despite these benefits, the higher computational costs of making centennial-scale, high-resolution, biogeochemical ocean simulations remain prohibitive. More practical in the short term would be to assess effects from less-costly model improvements, including heightened vertical resolution, subgrid-scale parameterizations, and adjustments to model parameters for viscosity and slip conditions. For such regional studies, nested models would offer the advantage of focused higher resolution while still avoiding adverse effects from imposed lateral boundary conditions.

These efforts along with including more coastal ocean processes in global models should eventually lead to greater prognostic skill and more reliable projections not only for the Arctic Ocean but for regional seas and the coastal ocean in general.

Code availability. The code for the NEMO ocean model version 3.2 is available under CeCILL license at <http://www.nemo-ocean.eu>.

Appendix A: Perturbation vs. full biogeochemical approach

To assess the reliability of the perturbation approach, we compared its results from the coarse-resolution ocean model over 1870–2012 (P1870-ORCA2*), i.e., without branching from ORCA05, to those from the analogous full biogeochemical simulation (B1870-ORCA2*). Globally, the simulated C_{ant} inventory with the perturbation approach in 2012 is 2% larger than that with the full biogeochemical approach (Table A1). These differences are mainly located in the top 200 m (Fig. A1) of the tropics and Southern Ocean (Fig. A2), where regional inventories of C_{ant} from the perturbation approach overestimate those from the full approach by up to 3%. Those two regions are also the ones that store most of the anthropogenic carbon. Conversely, in the Arctic, the perturbation approach underestimates the 2012 C_{ant} inventory of the full approach by 3% because of its deficit between 200 and 600 m, the depth zone that is directly affected by Atlantic inflow (Sect. 3.1.3). Overall, these differences are small, thus supporting our use of the more efficient perturbation approach to correct for the late start date of the full biogeochemical simulations.

These differences, although small, merit an explanation. The perturbation approach is regionally biased because its preindustrial reference state is assumed to be everywhere in equilibrium with the atmosphere (Sect. 2.3). Hence its results will differ from the full approach, which allows for disequilibrium between preindustrial atmospheric and oceanic $p\text{CO}_2$. For example, with the full approach, simulated surface-ocean $p\text{CO}_2$ in the tropics and Southern Ocean generally exceeds atmospheric $p\text{CO}_2$ under preindustrial conditions, a supersaturation that is also seen with ocean inversions for the same regions (Gruber et al., 2009). So by assuming equilibrium and not accounting for this supersaturation, the perturbation approach relies on a buffer capacity that is too high. That is, when its preindustrial surface-ocean $p\text{CO}_2$ reference is too low, its corresponding carbonate ion concentration is too high and thus so must be its buffer capacity, i.e., its chemical capacity to absorb C_{ant} .

In contrast, in the North Atlantic, surface-ocean $p\text{CO}_2$ is generally undersaturated in the full approach under preindustrial conditions (B1870-ORCA2* in 1870), as it is in ocean inversions (Gruber et al., 2009). By not accounting for this undersaturation, the perturbation approach overestimates the preindustrial surface ocean $p\text{CO}_2$ and thus underestimates the corresponding reference carbonate ion concentration, buffer capacity, and uptake of C_{ant} relative to the full approach. The growing influence of this underestimated uptake in the North Atlantic can be seen as its waters invade the Arctic during the course of the simulation (Fig. A3). That lateral invasion overwhelms the small but opposite tendency early in the perturbation simulation to overestimate Arctic C_{ant} uptake, an artefact of the perturbation approach's preindustrial reference state not accounting for local impacts from riverine inputs. Conversely, in the full approach with PISCES, riverine inputs typically lower the carbonate ion concentration and buffer capacity of shelf seas.

Despite its simplifications, the perturbation approach differs little from the full approach in terms of basinwide results. With it, we can envision garnering sufficient computational resources to soon make a global C_{ant} simulation at high resolution (ORCA025) over the full industrial era without branching it off from a lower resolution model along the way. That should in turn allow us to help further refine limits for C_{ant} uptake and storage in the Arctic as well as other regions. With the full biogeochemical approach, such would not be feasible for years to come.

Competing interests. The authors declare that there are no competing interests.

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Table 1. Grid size in the Arctic Ocean and volumes by basin as a function of model resolution.

Configuration	Horizontal grid (km)			Volume (10^6 km^3)				
	Mean	Min	Max	Basins				
				Arctic	Nansen	Amundsen	Makarov	Canada
ORCA2	120.8	63.3	180.5	14.3	2.8	3.2	2.2	4.7
ORCA05	29.0	9.4	41.3	13.3	2.6	2.7	1.9	4.9
ORCA025	14.4	3.2	20.5	13.3	2.3	2.9	1.8	5.0

Table 2. ~~Selected physical coefficients~~ Coefficients for lateral diffusivity, lateral viscosity, and parameters ~~eddy induced velocity~~ eddy induced velocity for ORCA2, ORCA05, and ORCA025.

Configuration	Lateral diffusivity	Lateral viscosity ^a	Eddy parameterization <u>induced velocity</u>
ORCA2 ^(a) ^b	2000 m²s⁻¹ <u>m²s⁻¹</u>	$4 \times 10^4 \text{ m}^2 \text{ s}^{-1}$ ^(b) ^c	$2000 \text{ m}^2 \text{ s}^{-1}$
ORCA05	600 m²s⁻¹ <u>m²s⁻¹</u>	$-4 \times 10^{11} \text{ m}^2 \text{ s}^{-1}$	$1000 \text{ m}^2 \text{ s}^{-1}$
ORCA025	300 m²s⁻¹ <u>m²s⁻¹</u>	$-1.5 \times 10^{11} \text{ m}^2 \text{ s}^{-1}$	none

^a In ORCA2, a Laplacian viscosity operator was used, whereas a bi-Laplacian operator was used in ORCA05 and ORCA025.

^b Lateral diffusivity and viscosity coefficients decrease towards the poles proportional to the grid size.

^c Reduced to $2100 \text{ m}^2 \text{ s}^{-1}$ in the tropics (except along western boundaries)

Table 3. Fitted parameters for the perturbation Set of simulations P1765 and P1870.

<u>Name</u>	<u>Resolution</u>	
	<u>Before 1958</u>	<u>After 1958</u>
Biogeochemical		
<u>B1870-ORCA2</u>	<u>ORCA05</u>	<u>ORCA2</u>
<u>-ORCA05</u>	<u>ORCA05</u>	<u>ORCA05</u>
<u>-ORCA025</u>	<u>ORCA05</u>	<u>ORCA025</u>
<u>-ORCA2*</u>	<u>ORCA2</u>	<u>ORCA2</u>
Perturbation		
<u>P1870-ORCA2</u>	<u>ORCA05</u>	<u>ORCA2</u>
<u>-ORCA05</u>	<u>ORCA05</u>	<u>ORCA05</u>
<u>-ORCA025</u>	<u>ORCA05</u>	<u>ORCA025</u>
<u>-ORCA2*</u>	<u>ORCA2</u>	<u>ORCA2</u>
<u>P1765-ORCA2</u>	<u>ORCA05</u>	<u>ORCA2</u>
<u>-ORCA05</u>	<u>ORCA05</u>	<u>ORCA05</u>
<u>-ORCA025</u>	<u>ORCA05</u>	<u>ORCA025</u>
<u>-ORCA2*</u>	<u>ORCA2</u>	<u>ORCA2</u>

Table 4. Fitted parameters in Eqs. 2 and 3 for the perturbation simulations P1765 and P1870.

Parameter	P1765	P1870
a_0	1.7481	1.8302
a_1	-3.2813×10^{-2}	-3.4631×10^{-2}
a_2	4.1855×10^{-4}	4.3614×10^{-4}
b_0	3.9615×10^{-3}	4.0105×10^{-3}
b_1	-7.3733×10^{-5}	-7.3386×10^{-5}
b_2	$5.4759 \times 10^{-5} \times 10^{-7}$	$5.1199 \times 10^{-5} \times 10^{-7}$

Table 5. Lateral transport of water and $C_{\text{amt}} - C_{\text{out}}$ across Arctic Ocean boundaries - **Simulated** with **average simulated** values **are** calculated for the same time period as observations.

	Model configuration			Observations	Year	Sources
	ORCA2	ORCA05	ORCA025			
Lateral water transport (Sv)						
Fram Strait	-1.76	-1.75	-1.80	-2.0 ± 2.7	1997–2006	Schauer et al. (2008)
	-1.76	-1.42	-1.46	-1.7	1980–2005	Rudels et al. (2008)
Barents Sea	1.20	2.50	2.77	2.0	2003–2005	Skagseth et al. (2008)
	1.04	2.42	2.78	2.0	1997–2007	Smedsrud et al. (2010)
Bering Strait	1.02	1.29	1.49	0.8 ± 0.2	1991–2007	Woodgate et al. (2010)
CAA	-0.29	-2.00	-2.59	-2.7 ± 0.2	2004–2013	Curry et al. (2014)
Sum	-0.12	-0.16	-0.18			
Lateral $C_{\text{amt}} - C_{\text{out}}$ fluxes (Tg C yr^{-1})						
	Model configuration			Observations	Year	Sources
	ORCA2	ORCA05	ORCA025			
Fram Strait	-17	-12	-8	-1 ± 17	2002	Jeansson et al. (2011)
	-17	-7	5	-12	2012	Stöven et al. (2016)
Barents Sea	16	43	50	41 ± 8	2002	Jeansson et al. (2011)
Bering Strait	18	22	27	18	2000 – 2010 ^a	Olsen et al. (2015)
CAA	-5	-28	-36	-29	2000 – 2010 ^a	Olsen et al. (2015)
Sum	18	29	38	29	2000 – 2010 ^a	Olsen et al. (2015)

^aObservational year or period impossible to identify exactly as C_{amt} and velocity measurements are not from the same year

Table 6. Total inventory, its change during 1960–2012, the cumulative air-sea flux, and the lateral flux of C_{ant} in Pg C

	Model configuration		
	ORCA2	ORCA05	ORCA025
C_{ant} inventory ^a			
C_{ant} in 2002 ^b	1.90 (1.47)	2.25 (1.81)	2.49 (2.06)
C_{ant} in 2005 ^c	1.99 (1.56)	2.37 (1.96)	2.64 (2.21)
Inventory change (1960-2012)			
Total Arctic	1.08	1.55	1.98
Nansen Basin	0.14	0.33	0.30
Amundsen Basin	0.13	0.28	0.34
Makarov Basin	0.15	0.21	0.33
Canada Basin	0.31	0.36	0.61
Cumulative fluxes (1960-2012)			
Air-Sea flux	0.29	0.43	0.48
Lateral flux of C_{ant} ^d	0.79	1.13	1.50
Fram Strait	-0.74	-0.40	-0.06
Barents Sea	0.79	1.75	1.98
Bering Strait	0.74	0.89	1.03
CAA	-0.22	-1.20	-1.50
Summed lateral flux	0.57	1.05	1.45

^a Numbers in parenthesis show the uncorrected value (starting date 1870)

^b Data-based inventory in 2002: 2.95 Pg C (GLODAPv2)

^c Data-based inventory in 2005: 3.03 Pg C (2.5-3.3) (Tanhua et al., 2009)

^d Computed as inventory change minus cumulative air-sea flux

Table 7. Along-section CFC-12 inventories [$\mu\text{mol m}^{-1}$] integrated over depth and distance along the AOS94 and Beringia 2005 ~~expedition~~ ~~compared to sections vs. colocated~~ results ~~from in~~ ORCA2, ORCA05, and ORCA025 ~~along the same sections~~.

	AOS94	Beringia 2005
Observation	5.5	9.4
ORCA2	4.8 <u>2.9</u>	7.7 <u>3.7</u>
ORCA05	3.5	5.8
ORCA025	2.9 <u>4.8</u>	3.7 <u>7.7</u>

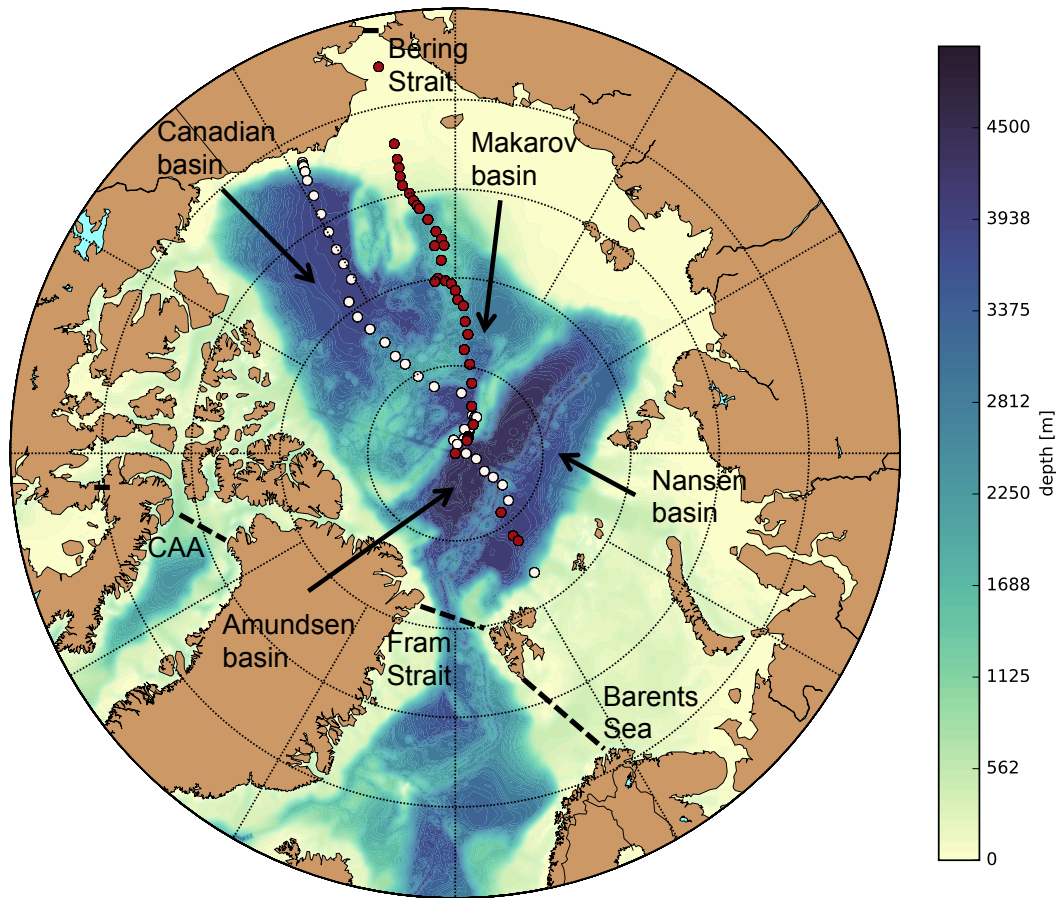


Figure 1. CFC-12 stations occupied during along the AOS94 (red) and Beringia 2005 expedition-expeditions (white). The filled-color scheme indicates Other colors indicate the bathymetry of the Arctic Ocean, while the four dashed black lines show the boundaries of the Arctic Ocean domain used in this study.

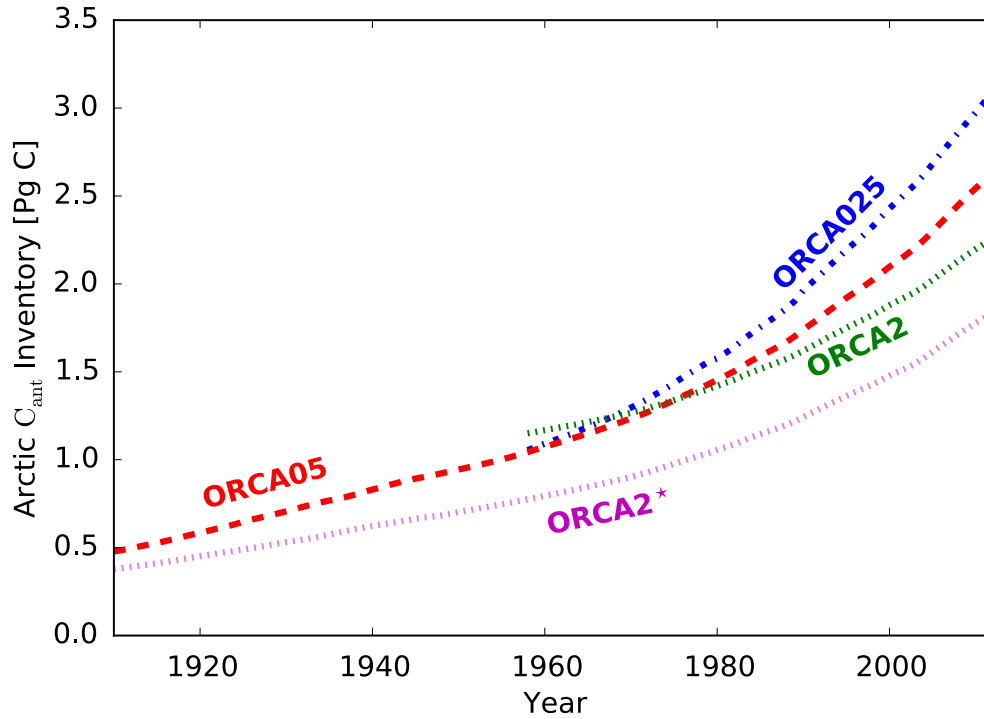


Figure 2. Arctic Ocean C_{ant} inventory for the three biogeochemical simulation resolutions corrected for the late starting date. The intermediate-resolution model (ORCA2, ORCA05—red dashed) was integrated from 1870 to 2012. The high-resolution model (ORCA025—blue dash-dot) and the low-resolution model (ORCA2—dotted green) were initialized with the ORCA05 output at the end of 1957 and integrated from 1958 to 2012. The discontinuity for ORCA2 in 1958 is due to its larger total volume of water when integrated across the Arctic domain (Table 1).

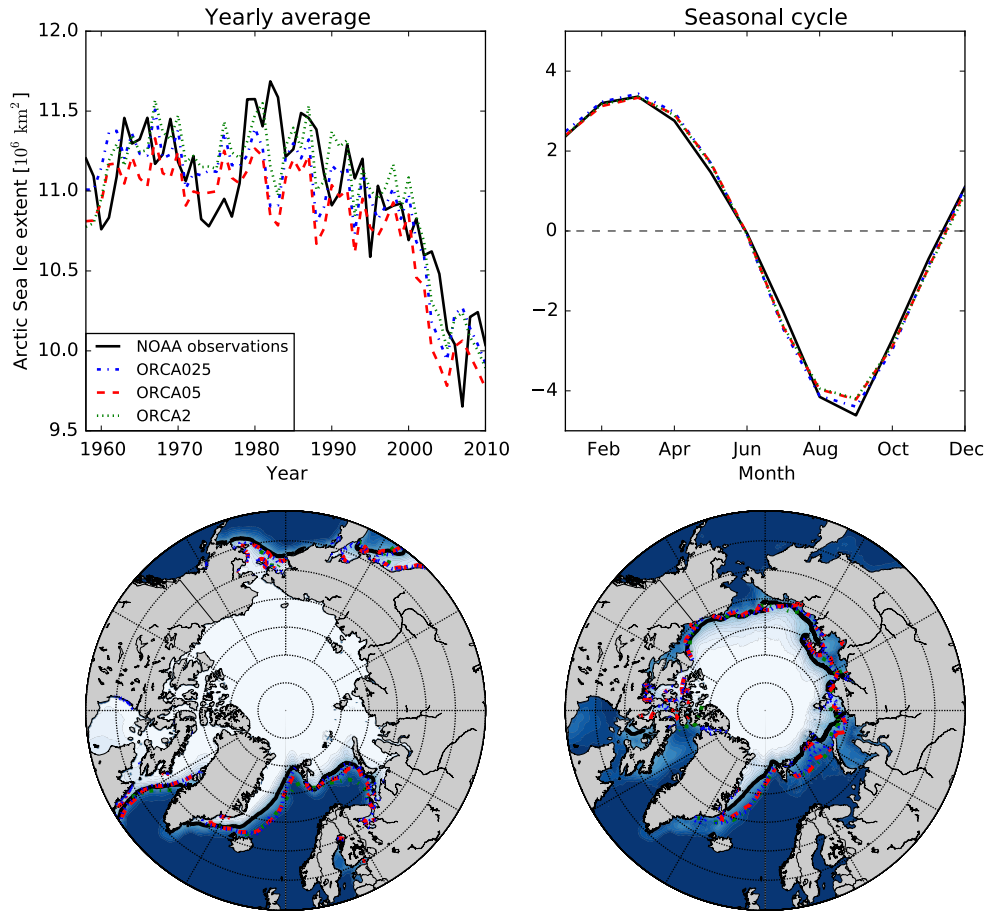


Figure 3. Sea-ice extent (top) and sea-ice concentration in (bottom) over the Arctic from 1960 to 2012 comparing microwave-based observations from NOAA (black) to simulated results from ORCA2 (green dots), ORCA05 (red dashes), and ORCA025 (blue dot-dash). Shown are the yearly average-averages (top left), and after detrending, the average (climatological) seasonal cycle over 1958–2010 (top right), and the average sea-ice extent in winter (December, January, February) (bottom left) and summer (July, August, September) (bottom right). The lines on the maps show the 50% sea-ice cover for the three model resolutions and the observations. The white, while the color indicates the observed sea-ice concentration.

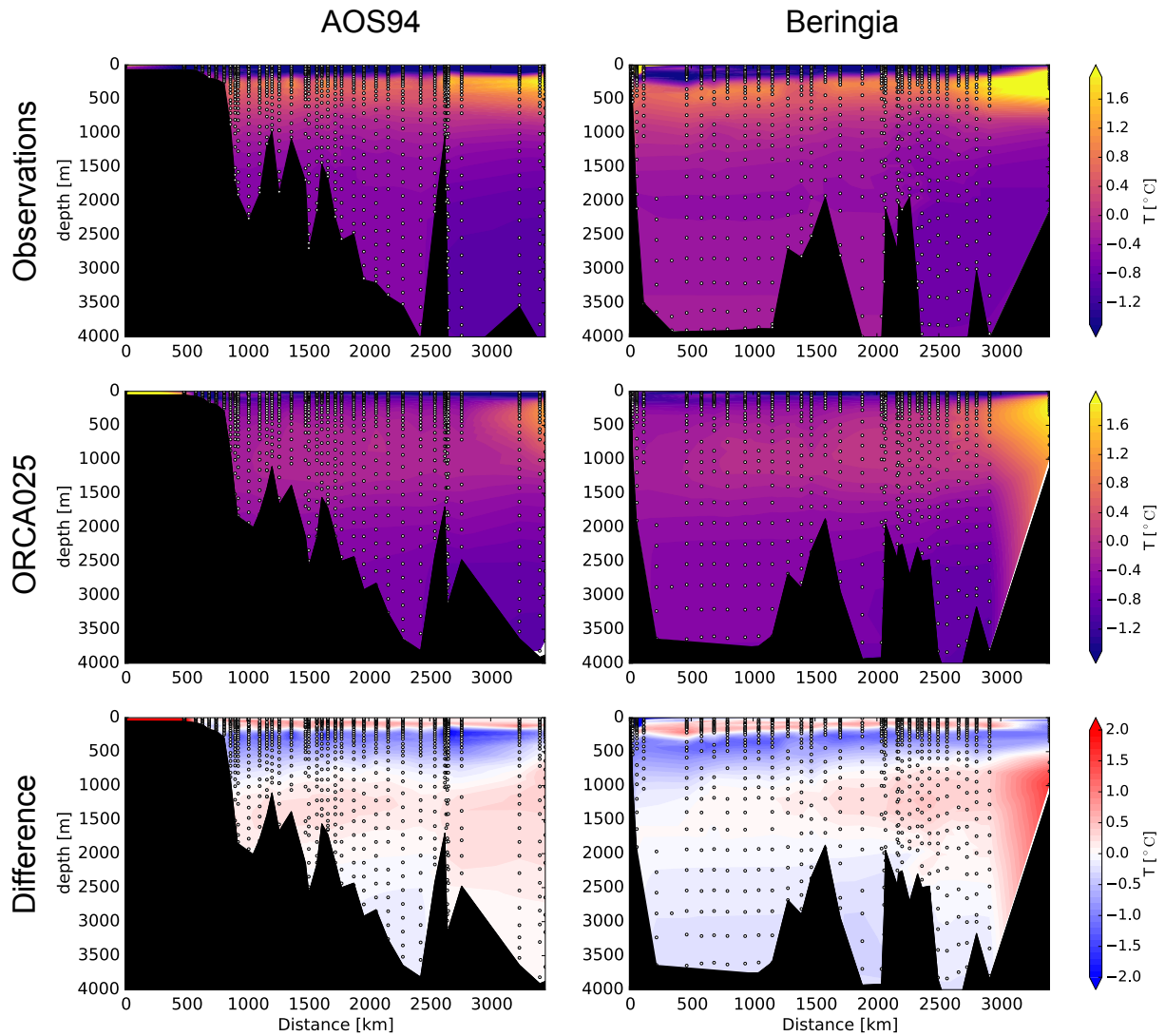


Figure 4. Temperature along the 1994 Arctic Ocean Section (AOS94) cruise (left) and the Beringia/HOTRAX 2005 [expedition section](#) (right), both trans-Arctic transects (Fig. 1). The observations (top) are compared to simulated results from ORCA025 averaged over summer of the respective year (middle). The [model-data](#) difference ($\text{model} - \text{observations}$) is shown at the bottom. [The location of the sections is shown in Fig. 1.](#)

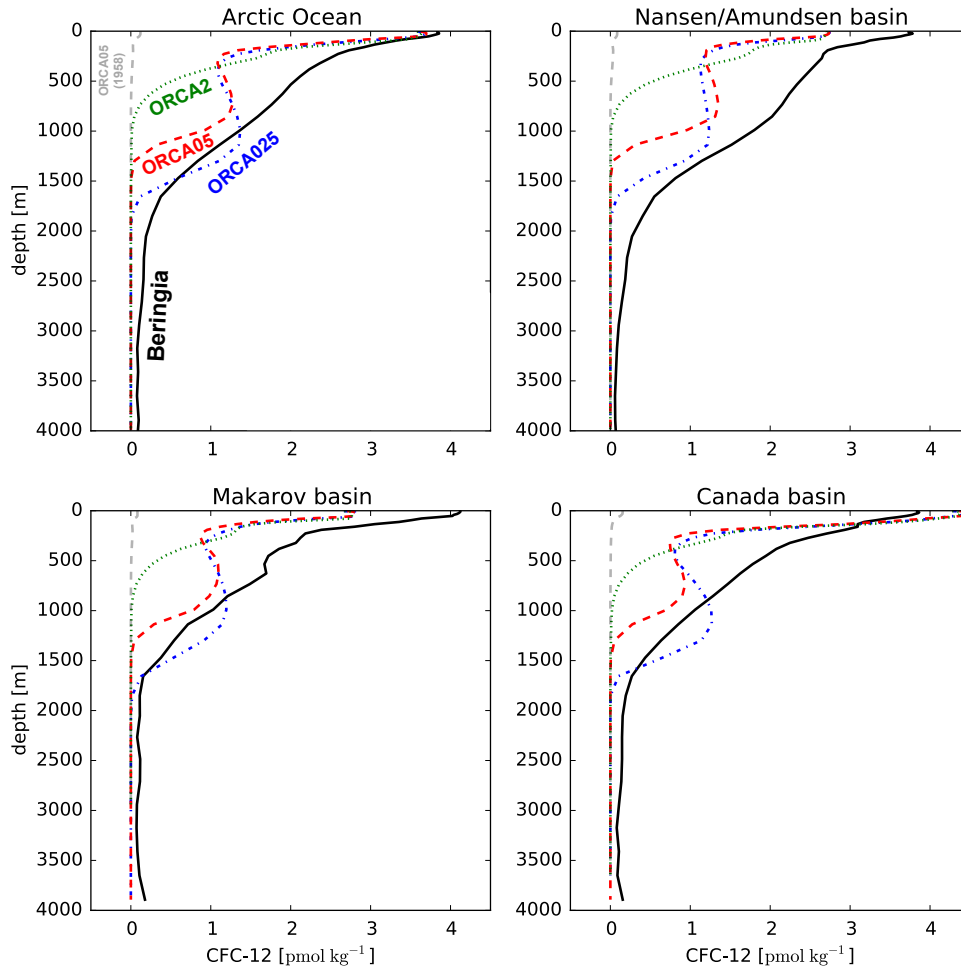


Figure 5. Profiles of observed CFC-12 for observations (black solid) and the ORCA025 simulated CFC-12 in ORCA2 (blue green dots), ORCA05 (red dashes), and ORCA2-ORCA025 (green blue dot-dash) model along the Beringia 2005 sections section. Shown are distance-weighted mean means across each that entire section (top left) as well as over that section covering the Nansen and Amundsen basins (top right), the Canada-Makarov basin (bottom left), and the Makarov-Canada basin (bottom right). Shown in light grey is the vertical profile in 1958, the branching point for the three resolutions.

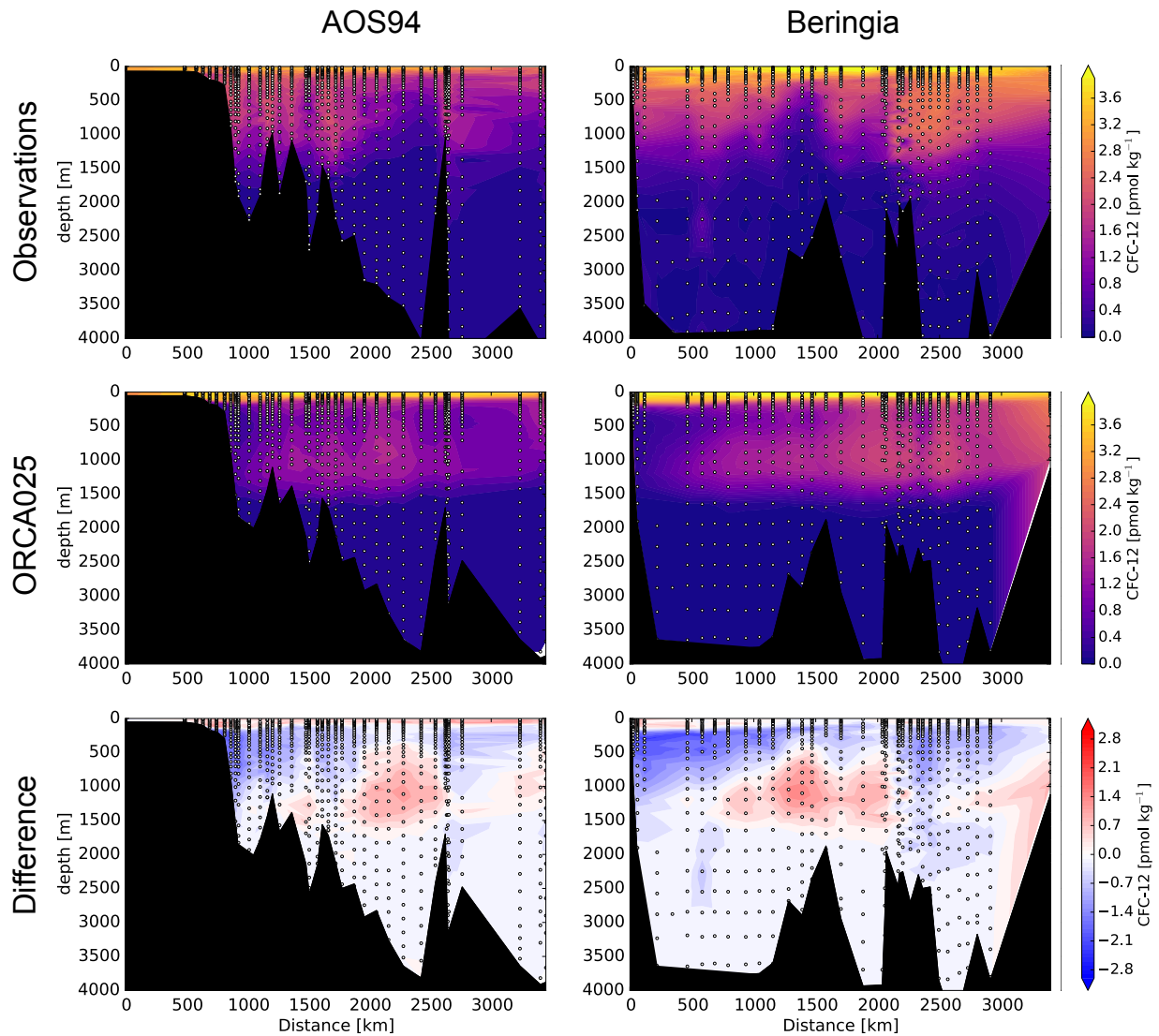


Figure 6. CFC-12 sections along the AOS94 section (left) and the Beringia section (right). The observations (top) are compared to the simulated summer means in ORCA025 (middle), and the model-data difference is shown at the (bottom).

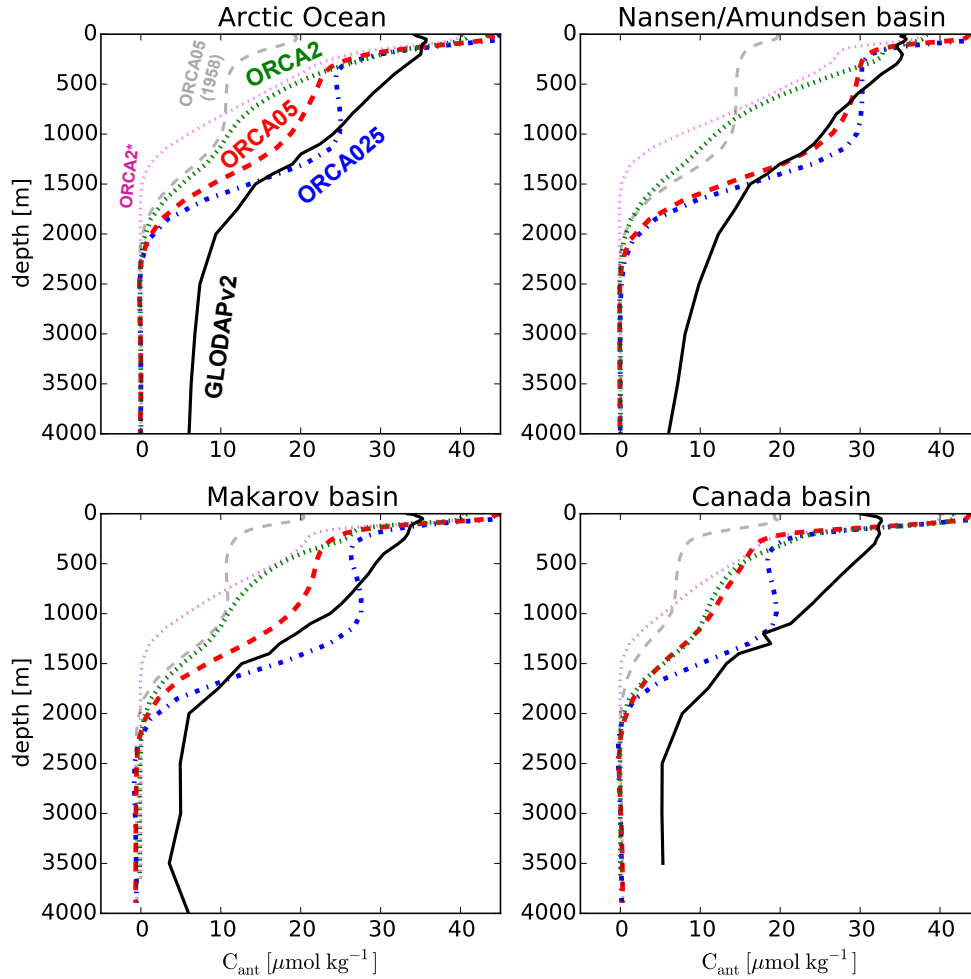


Figure 7. Area-weighted basinwide average vertical profiles of C_{ant} concentration in 2002 for GLODAPv2 data-based estimates (black solid), ORCA2 (green dots), ORCA05 (red dashes) and ORCA025 (blue dot-dash) over the entire Arctic (top left) as well as over the Nansen and Amundsen basins (top right), the Makarov basin (bottom left), and the Canada basin (bottom right). Ocean corrected for the starting year by the perturbation approach simulations. The shown are the vertical profile in 1958 (dashed, when light grey), the simulation is divided branching point for the three resolutions, is shown in light grey and results from ORCA2* for 2002 (magenta dots).

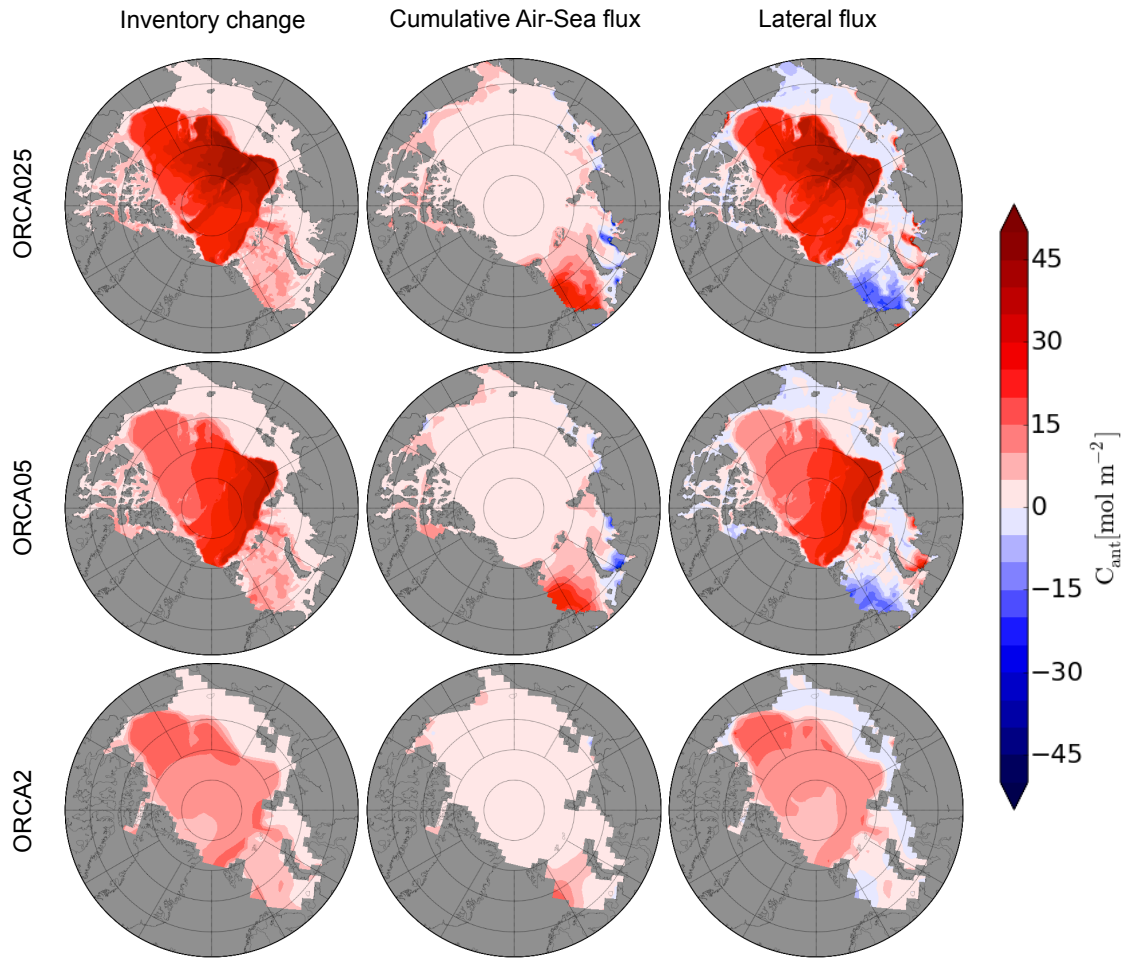


Figure 8. Inventory change (left), cumulative air-sea flux (middle), and the lateral flux calculated as the **difference of inventory change and minus the cumulative** air-sea flux (right) of C_{ant} over 1960–2012 for the period from 1960 to 2012 for the ORCA025 (top), ORCA05 (center), and ORCA2 (bottom) model configurations.

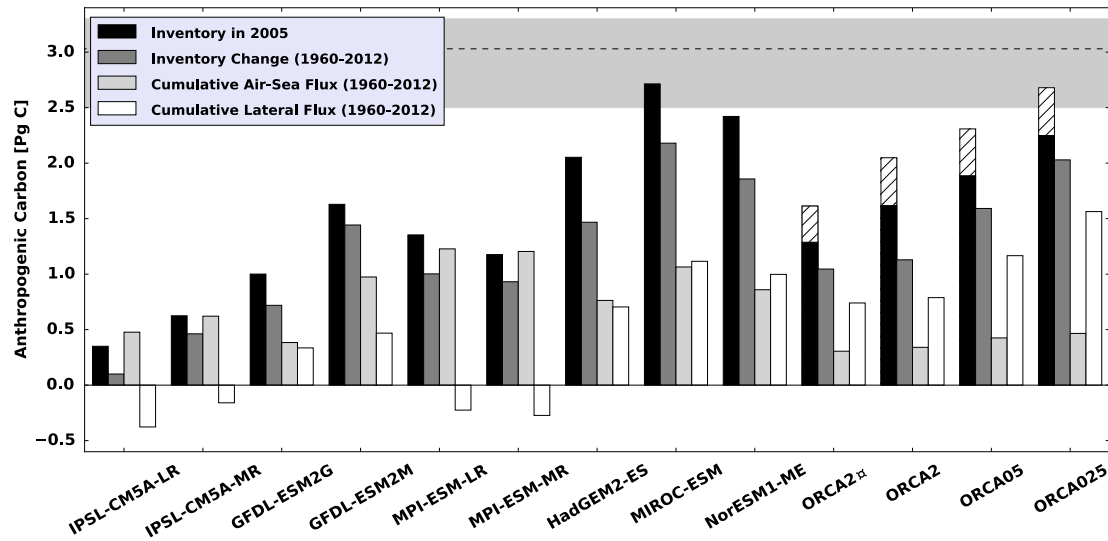


Figure 9. Comparison of results for the Arctic Ocean from the three resolutions of NEMO-PISCES (ORCA2, ORCA05, ORCA025, and ORCA2^{*}) and the nine Earth System Models that participated in CMIP5. Shown are the C_{ant} inventory in 2005 (black), the inventory change of C_{ant} (dark grey) between 1960 and 2012, the corresponding cumulative air-sea flux of C_{ant} (light grey), and the cumulative lateral flux of C_{ant} (white). Also indicated are the data-based estimate by Tanhua et al. (2009) (dashed black line) and the associated uncertainty estimates range (grey background). The inventory correction for the biogeochemical late starting date for our forced simulations using the perturbation approach is added to the results of the biogeochemical simulations indicated as striped bars.

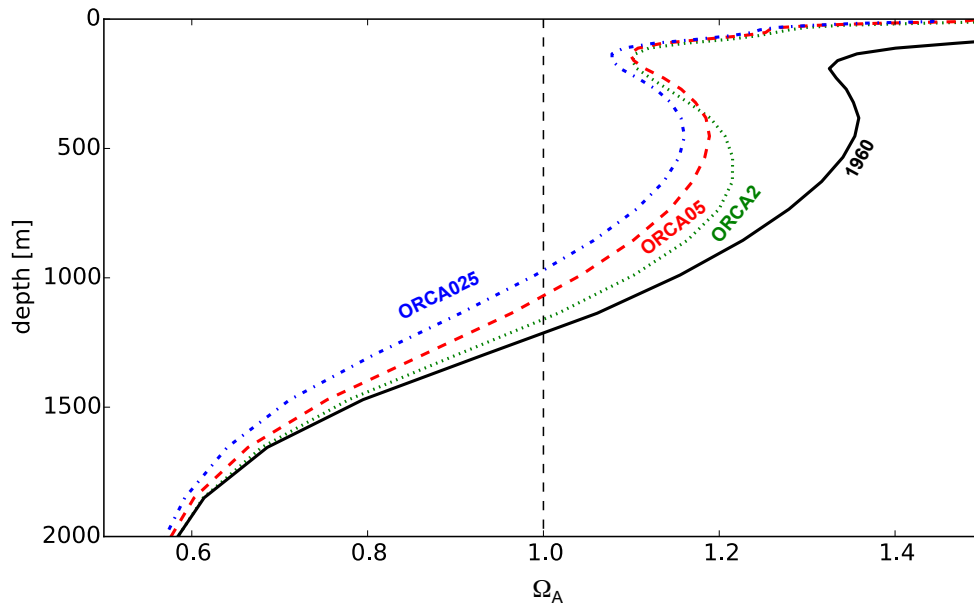


Figure 10. Profiles of Ω_A for ORCA05 in 1960 (black solid) as well as ORCA2 (green ~~dot-dash~~dots), ORCA05 (red dashes), and ORCA025 (blue ~~dots~~dot-dash) in 2012. The vertical black dashed line indicates the chemical threshold where $\Omega_A = 1$. Where that vertical line intersects the other curves indicates the depth of the ASH ~~in each case~~.

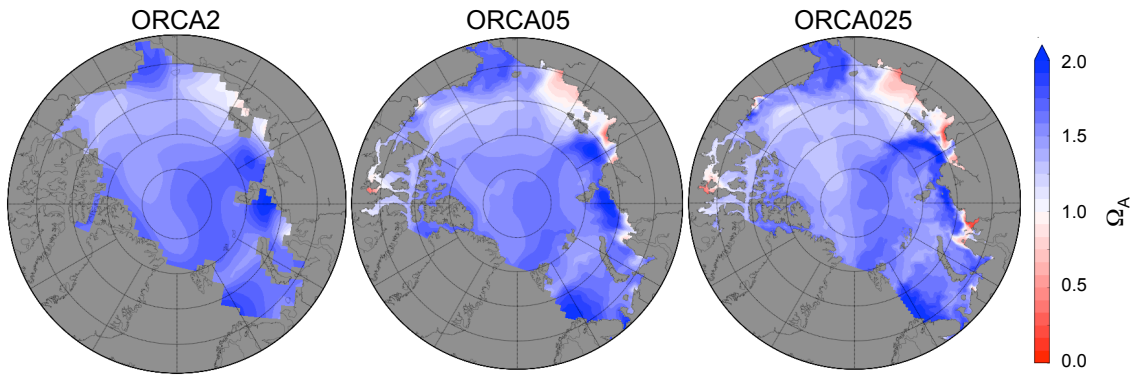


Figure 11. Surface Ω_A for ORCA2, ORCA05 and ORCA025 (from left to right) in August 2012.

Table A1. C_{ant} inventories in 2012 in Pg C.

	<u>Global Ocean</u>	<u>Arctic Ocean</u>
<u>B1870-ORCA2*</u>	<u>137.3</u>	<u>1.48</u>
<u>P1870-ORCA2*</u>	<u>139.6</u>	<u>1.43</u>

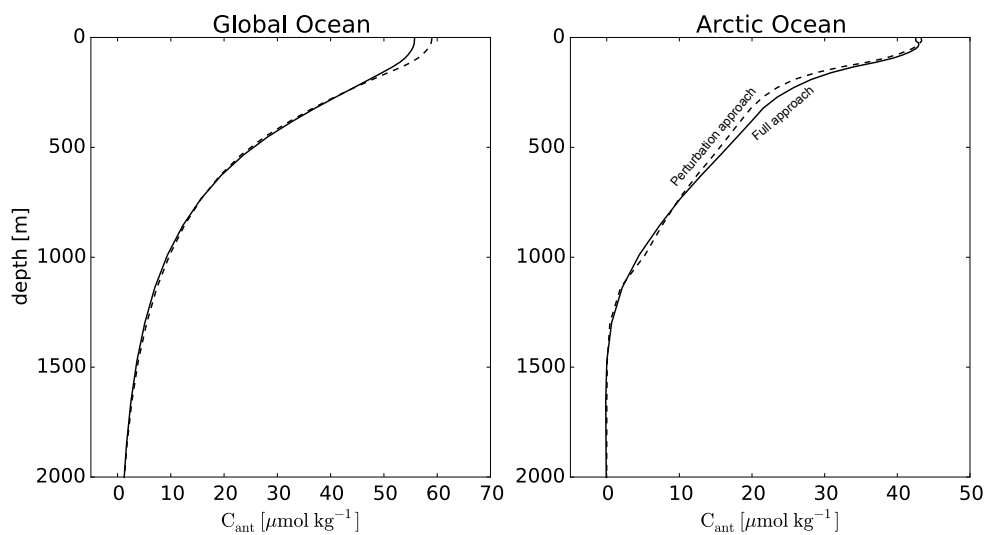


Figure A1. Mean vertical profiles of C_{ant} for the global ocean (left) and the Arctic Ocean (right) in 2012 for the full biogeochemical approach (B1870-ORCA2*) (solid) and the perturbation approach (P1870-ORCA2*) (dashed).

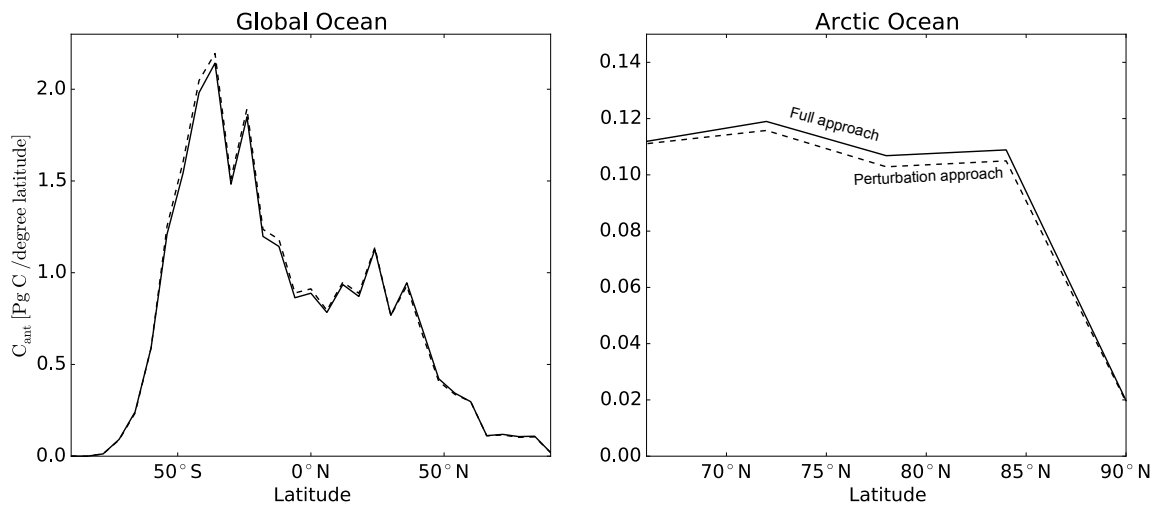


Figure A2. Zonal integral of vertically integrated C_{ant} per degree of latitude in 2012 for the global ocean (left) and the Arctic Ocean (right) using the full biogeochemical approach (B1870-ORCA2*) (solid) and the perturbation approach (P1870-ORCA2*) (dashed)

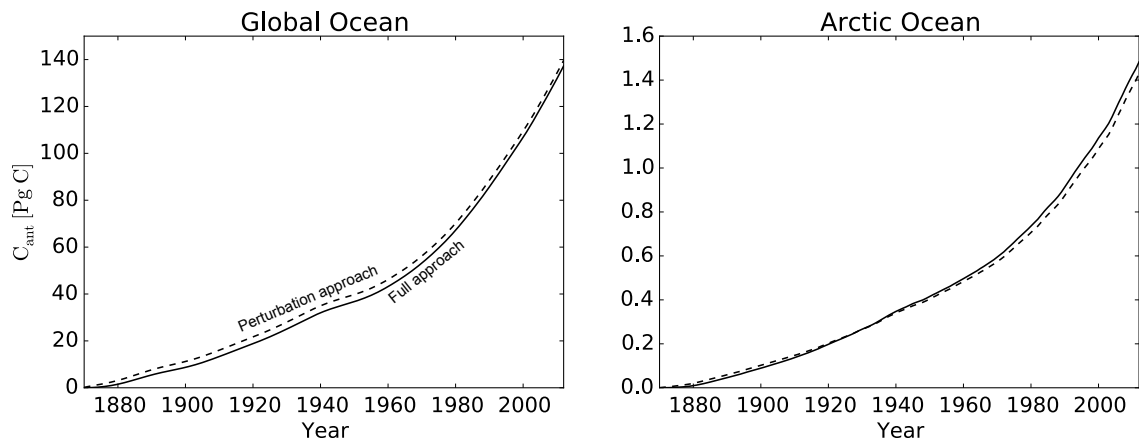


Figure A3. Arctic Ocean C_{ant} inventory in the global ocean (left) and the Arctic Ocean (right) for the full biogeochemical approach (B1870-ORCA2*) (solid) and the perturbation approach (P1870-ORCA2*) (dashed).