Adective Controls on the North Atlantic Anthropogenic Carbon Sink

S. M. Ridge and G. A. McKinley

1Lamont-Doherty Earth Observatory, Columbia University, Palisades, NY, USA

Abstract Though it is clear that the North Atlantic is the site of the highest storage of anthropogenic carbon (C_{ant}) per area, it is uncertain whether the air-sea C_{ant} fluxes contributing to North Atlantic C_{ant} storage occur in the subpolar gyre or upstream in the subtropical gyre. Using data and models, we show that air-sea C_{ant} uptake capacity is advected into the subpolar gyre along the same subsurface pathway as nutrients. This pathway is known as the nutrient stream. On the A22 section between Woods Hole and Bermuda, nutrient stream waters are the oldest in the upper 2,000 m and contain low C_{ant}. These northward moving waters are sufficiently depleted in C_{ant} such that they could sustain a subpolar air-sea C_{ant} flux of \(-0.19\) Pg C_{ant} year\(^{-1}\). The ocean hindcast model used here indicates that despite some subtropical re-circulation, uptake capacity is transported into the subpolar gyre where it sustains subpolar air-sea C_{ant} uptake. With this model, we show that high- and low-end estimates of subpolar air-sea C_{ant} flux are reconciled by accounting for a factor of two difference in their respective study areas. If half of the observed air-sea C_{ant} uptake capacity transport at A22 is ventilated in the subpolar region, it can fully support high-end estimates of the subpolar air-sea C_{ant} sink (\(\sim0.09 \pm 0.01\) Pg C_{ant} year\(^{-1}\)).

Plain Language Summary The ocean has absorbed approximately 40% of the CO\(_2\) from fossil fuel burning and cement production, lowering atmospheric CO\(_2\) and limiting climate change. Ocean storage of this additional carbon from human activities (anthropogenic carbon, C_{ant}) is most intense in the North Atlantic. In the subpolar North Atlantic, C_{ant} is injected into the deep ocean and thus sequestered from the atmosphere for thousands of years. It is currently uncertain whether any of this sequestered C_{ant} was absorbed from the atmosphere in the subpolar North Atlantic. Here we present evidence that the upper limb of the ocean’s overturning circulation supplies the subpolar North Atlantic with the capacity to absorb C_{ant} from the atmosphere. Subpolar uptake capacity travels along the same subsurface pathways as the nutrients that support subpolar phytoplankton growth. Waters following this pathway likely started their northward journey decades ago, in the Southern Ocean and thus have not been in contact with the high atmospheric carbon concentrations that exist today. When these waters are exposed to the atmosphere in the subpolar North Atlantic, they can absorb a large amount of C_{ant}. We show that this is the mechanism by which the subpolar North Atlantic absorbs a substantial amount of C_{ant}.

1. Introduction

The ocean is a sink for approximately 40% of anthropogenic carbon (C_{ant}) emissions resulting from fossil fuel combustion and cement production (Ciais et al., 2013; Khatiwala et al., 2009). Before the Industrial Revolution, it is believed that there was a steady state natural carbon (C_{nat}) cycle. C_{nat} is brought to the surface by ocean circulation and is consumed by phytoplankton, whose remains and waste products ultimately sink and are remineralized, returning the carbon to depth. This process contributes to more C_{nat} being stored at depth. Regional surface heating and cooling tendencies also play an important role in the C_{nat} cycle. The cooling of northward moving subtropical waters favors absorption of atmospheric CO\(_2\) and eventually results in the transformation of subtropical waters into deep waters, thus also enhancing deep water C_{nat} storage.

C_{ant} has been added to the atmosphere and is absorbed because the surface ocean is out of equilibrium with the rapidly growing C_{ant} perturbation. To date, there is no evidence of significant direct interaction between the C_{nat} and C_{ant} components of the carbon cycle; therefore we assume that the C_{ant} behaves as a passive tracer, unaffected by biology. Contemporary carbon (C_{con}) is the sum of the C_{nat} and C_{ant} components, and C_{con} is what is directly measured. Estimates of the relatively small C_{ant} signal contained in C_{con} must be inferred from other biogeochemical tracers.
$C_{\text{ant}}$ is stored primarily in the upper ocean, mostly in mode and intermediate waters (Gruber et al., 2019; Sabine et al., 2004). While approximately 35–50% of ocean $C_{\text{ant}}$ is stored in subpolar mode waters and intermediate waters ($\sigma_t = 26.5–27.5$ kg m$^{-3}$), these water masses account for only 20% of ocean volume (Iudicone et al., 2016). Mode and intermediate waters dominate air-sea $C_{\text{ant}}$ uptake, a result of winds and buoyancy fluxes shaping a water mass with high outcrop area to volume ratio (Iudicone et al., 2016). The Southern Ocean and North Atlantic are primary ventilation regions for mode and intermediate waters, and accordingly, air-sea $C_{\text{ant}}$ uptake occurs primarily in those regions (Mikaloff Fletcher et al., 2006). These regions sustain high $C_{\text{ant}}$ fluxes due to the upwelling of waters with a partial pressure of CO$_2$ (pCO$_2$) that is close to preindustrial values, and therefore these waters are low in $C_{\text{ant}}$.

The North Atlantic basin is the most intense air-sea $C_{\text{ant}}$ sink (Mikaloff Fletcher et al., 2006) and the site of rapid injection of $C_{\text{ant}}$ into the deep ocean facilitated by Labrador Sea deep convection (Yashayaev & Loder, 2016). However, the spatial distribution of air-sea $C_{\text{ant}}$ uptake between the subtropical and subpolar North Atlantic is under debate (Mikaloff Fletcher et al., 2006; Pérez et al., 2013). Ocean inversions suggest subpolar air-sea $C_{\text{ant}}$ uptake is approximately half ($-0.09 \pm 0.01$ Pg $C_{\text{ant}}$ year$^{-1}$) of North Atlantic uptake ($-0.22 \pm 0.02$ Pg $C_{\text{ant}}$ year$^{-1}$ for 36–76°N) (Mikaloff Fletcher et al., 2006). However, a recent observational analysis suggests that most air-sea $C_{\text{ant}}$ uptake occurs in the subtropics, and the subpolar air-sea $C_{\text{ant}}$ sink is minimal (Pérez et al., 2013). Differing estimates of subpolar air-sea $C_{\text{ant}}$ flux may be attributable simply to substantially different regional boundaries used to define the subpolar North Atlantic. Mikaloff Fletcher et al. (2006) utilize latitudinal boundaries of 49°N to 76°N to define the subpolar North Atlantic, whereas Pérez et al. (2013) set the southern boundary of the subpolar North Atlantic along the Portugal-Greenland A25 (~40–60°N) section and northern boundary at 78°N. The latter definition leaves out the Labrador Sea and a significant portion of the Irminger Sea from the subpolar region. Here we explore the effects of various study regions on the magnitude of subpolar air-sea $C_{\text{ant}}$ flux estimates.

Mechanisms controlling the interannual variability of the North Atlantic $C_{\text{con}}$ and $C_{\text{ant}}$ sink have been investigated in the context of the changes in the NAO state from the early 1990s to the early 2000s (Pérez et al., 2013; Thomas et al., 2008; Ullman et al., 2009). We will expand upon this work in our effort to determine what controls the magnitude of the subpolar $C_{\text{ant}}$ sink. Pérez et al. (2013) investigate the role of the North Atlantic Oscillation (NAO) and the Atlantic Meridional Overturning Circulation (AMOC) in subpolar North Atlantic $C_{\text{ant}}$ sink variability from 1997 to 2004. Using hydrographic data, Pérez et al. (2013) find that lateral transport and storage rates of $C_{\text{ant}}$ were higher in 1997 than in 2004. From 1989 to 1995, there was an extended period of high wintertime NAO (Hurrell & National Center for Atmospheric Research Staff (eds), 2018). Under the assumption that circulation in 1997 is representative of the high NAO period (1989–1995) and that 2004 is representative of the ensuing low NAO period (2002–2006), Pérez et al. (2013) conclude that subpolar North Atlantic air-sea $C_{\text{ant}}$ flux was more intense in 1997 than in 2004 due to higher NAO and AMOC in 1997. However, the changes in the air-sea $C_{\text{ant}}$ flux are not statistically significant given uncertainty bounds. Using two different hindcast models, Thomas et al. (2008) and Ullman et al. (2009) find that as the NAO index declines from the 1990s to the 2000s, there is a reduction of air-sea $C_{\text{con}}$ uptake in the eastern subpolar gyre and increases in air-sea $C_{\text{con}}$ uptake in the western subpolar region. Because the available hydrographic cruise data require the southern boundary for the subpolar gyre to be at A25, the analysis of Pérez et al. (2013) likely misses a compensating region of increased air-sea $C_{\text{con}}$ uptake in the west that was identified in these two modeling studies.

To improve mechanistic understanding of subpolar air-sea $C_{\text{ant}}$ uptake, we investigate the lateral supply of intermediate waters into the subpolar North Atlantic. Lagrangian drifters indicate very limited net flow of surface subtropical waters into the subpolar gyre (Burkholder & Lozier, 2011, 2014). Instead, parcels follow subsurface pathways to the surface of the subpolar gyre, meaning that mode and intermediate waters in the subtropics eventually reach the surface in the subpolar gyre.

The pathways of mode and intermediate waters have also been investigated in the context of the nutrient stream (Palter & Lozier, 2008; Pelegrí et al., 1996; Sarmiento et al., 2004; Williams et al., 2006). We expand upon these nutrient stream studies by considering how tracer distributions in nutrient stream waters can influence air-sea $C_{\text{ant}}$ uptake. The nutrient stream is a subsurface region of high nutrients embedded within the western boundary current that supplies the subpolar North Atlantic with nitrate (NO$_3$) and supports its intense subpolar primary production (Pelegrí et al., 1996; Williams et al., 2011). NO$_3$ increases with depth due to biological removal at the surface and remineralization at depth. In contrast, the surface $C_{\text{ant}}$ source
means that $C_{\text{ant}}$ decreases with depth. Steeply sloping isopycnals result in isopycnal transport bringing high NO$_3$ concentrations, and possibly low $C_{\text{ant}}$ concentrations, toward the surface where they then intersect with high velocity currents. Deep winter mixing at higher latitudes allows for these isopycnals to outcrop and for the nutrients to ultimately surface in the subpolar North Atlantic (Williams et al., 2011). Nutrient stream transport occurs primarily in intermediate waters ($\sigma_\theta = 27.0–27.5$ kg m$^{-3}$) that dominate net volume transport into the subpolar gyre (Burkholder & Lozier, 2011). This interpretation is supported by model experiments (Williams et al., 2006) where idealized tracer released in dense waters of the subtropical North Atlantic outcrops in the subpolar gyre, while tracer released in lighter subtropical surface waters tends to recirculate in the subtropical gyre. Lateral supply of nutrients in the nutrient stream is much greater than supply by Ekman suction within the subpolar gyre (Whitt, 2019; Williams et al., 2006). Thus, studies of both simulated tracer release and observed drifter trajectories indicate that the surface of the subpolar gyre is irrigated from the south with nutrients originating in mode and intermediate waters. Here we investigate whether the nutrient stream also irrigates the subpolar gyre with low $C_{\text{ant}}$ waters.

Nutrient stream waters are mostly composed of the remnants of Antarctic Intermediate Water (AAIW) (Palter & Lozier, 2008; Sarmiento et al., 2004; Sen Gupta & England, 2007), and their slow northward transit beneath the euphotic layer (Sen Gupta & England, 2007) allows for high remineralized nutrient concentrations (Williams et al., 2011). Respiration of organic matter also drives down O$_2$ concentration and results in a large difference between measured and saturation O$_2$ (Apparent Oxygen Utilization, AOU). The high AOU found in nutrient stream waters (Williams et al., 2011) indicates that they are relatively old, that is, have not been in contact with the atmosphere for many decades. The above evidence suggesting high nutrient stream water mass age is consistent with the nutrient stream containing low $C_{\text{ant}}$ waters.

The supply of waters with the potential to take up anthropogenic CO$_2$, low in $C_{\text{ant}}$, is necessary to sustain a subpolar $C_{\text{ant}}$ sink. In the Southern Ocean, the supply of low $C_{\text{ant}}$ comes from Circumpolar Deep Water that is upwelled to the surface, accumulates anthropogenic carbon, and is then subducted in mode and intermediate waters (Sabine et al., 2004). Relative to the rate of increase in atmospheric CO$_2$, circulation timescales (Sen Gupta & England, 2007) are long enough that these waters have the potential to absorb more anthropogenic carbon when they eventually resurface in subpolar North Atlantic. If nutrient stream waters contain sufficiently low $C_{\text{ant}}$, their ventilation in the subpolar North Atlantic may contribute significantly to air-sea $C_{\text{ant}}$ uptake in the subpolar North Atlantic.

Our objective is to quantify the northward transport of nutrient stream waters with a substantially lower $C_{\text{ant}}$ content. We will determine the degree to which transport in the nutrient stream density band, which has been shown to outcrop in the subpolar North Atlantic, is sufficient to sustain subpolar North Atlantic air-sea $C_{\text{ant}}$ uptake. With our analysis of $C_{\text{ant}}$ transport, we improve our understanding of the mechanisms driving subpolar-subtropical partitioning of North Atlantic air-sea $C_{\text{ant}}$ uptake. We will test the following hypothesis: Sufficient $C_{\text{ant}}$ uptake capacity to sustain the subpolar North Atlantic $C_{\text{ant}}$ sink is advected northward in the nutrient stream.

In order to determine the pathways of low $C_{\text{ant}}$ waters in the North Atlantic, we use a gridded biogeochemical data product (Lauvset et al., 2016) and ocean circulation from the best estimate of the physical state (Forget et al., 2015). Though there are interpolation issues with the gridded data product (section 2.7), we can qualitatively understand these effects by comparing the gridded data product to a hindcast simulation (Yeager et al., 2018) with internally consistent circulation and biogeochemistry. We estimate transport of air-sea $C_{\text{ant}}$ uptake capacity along these pathways using biogeochemical and hydrographic measurements from a quality controlled bottle data product (Olsen et al., 2016). Using the hindcast simulation, we can also look at how subpolar air-sea $C_{\text{ant}}$ flux changes with time. After validating the hindcast simulation against transport estimates from this study and others, we investigate variability in subpolar ventilation and relate it to variability in subpolar air-sea $C_{\text{ant}}$ flux.

2. Data and Methods

We leverage a suite of observational products, model simulations, and direct observations to investigate the link between the nutrient stream and the North Atlantic $C_{\text{ant}}$ sink.

2.1. Gridded Observations

We use the GLODAPv2.2016b data product (GLODAPv2) (Lauvset et al., 2016) for climatological spatial distributions of biogeochemical variables. This product is an aggregation of high-quality observations of
primary biogeochemical variables, collected from 1972 to 2013, and gridded to 1° × 1°. Most of these observations were collected as part of the World Ocean Circulation Experiment (WOCE) in the 1990s and the subsequent and ongoing CLIVAR repeat hydrography and GO-SHIP programs. For example, while GLODAPv2 incorporates observations over 42 years, half of the stations with DIC measurements used to make GLODAPv2 were sampled between 1997 and 2007. \( C_{ant} \) and \( C_{nat} \) are estimated using the Transit Time Distribution (TTD) method of Waugh et al. (2006) and are normalized to 2002. GLODAPv2 is available online at NOAA's Ocean Carbon Data System (OCADS, https://www.nodc.noaa.gov/ocads/).

2.2. Bottle Data

The GLODAPv2 bottle data product (Olsen et al., 2016) is used to resolve tracer gradients within the Gulf Stream. Improved representation of tracer gradients, relative to the gridded GLODAPv2 product, is necessary for more accurate calculation of \( C_{ant} \) transport because it depends critically on the intersection of velocity (section 2.5) and tracer gradients. The GLODAPv2 bottle data product features additional bias corrections and quality control on the original cruise data. Measurements of key biogeochemical variables (oxygen, DIC, and alkalinity) and transient tracers are from the October to November 2003 occupation of the WOCE A22 section (Bermuda-Woods Hole segment, Figure 1). These measurements and all derived variables are interpolated between stations along isopycnals, using Ocean Data View 4.7 (ODV), to be consistent with the physics of interior transport. The Bermuda-Woods Hole segment, centered at approximately 38°N (Figure 1), is chosen because it is the northernmost WOCE section that is also south of the nutrient stream outcrop region at approximately 45°N. These data are available online from NOAA's OCADS.

2.3. \( C_{ant} \) Estimate

The \( \phi C_{T} \) method (Pérez et al., 2008; Vázquez-Rodríguez et al., 2009) is used to estimate \( C_{ant} \) along the Bermuda-Woods Hole segment. The \( \phi C_{T} \) method is an upgraded version of the \( \Delta C^* \) back-calculation method (Gruber et al., 1996) that includes improved parameterization of preformed alkalinity and the time evolution of air-sea disequilibrium.

While the TTD method, used in GLODAPv2, and the \( \phi C_{T} \) method provide similar estimates, they obtain their estimates in very different ways. The \( \phi C_{T} \) method removes background biological remineralization and \( C_{nat} \) from measured DIC to obtain a \( C_{ant} \) estimate, while the TTD method derives \( C_{ant} \) based on a distribution of transit times that is informed by transient tracers and assumptions about the importance of advection versus mixing. A detailed intercomparison of \( C_{ant} \) estimation methods, including the \( \phi C_{T} \) method, can be found in Vázquez-Rodríguez et al. (2009). Like the TrOCA method (Touratier & Goyet, 2004), local CFC measurements are not needed because relationships between the air-sea disequilibrium component and the transient tracers are predetermined for each basin. This allows the \( \phi C_{T} \) method to be applied to sections lacking CFC observations. In the case of our analysis of A22, CFC data provide independent evidence that the waters of the nutrient stream are old. The uncertainty of \( C_{ant} \) estimates made with the \( \phi C_{T} \) method is
We also employ a hindcast model from which the GLODAPv2 product are normalized to 2002. We focus on a 1997–2007 period, centered on 2002, because carbonate system measurements in the gridded Monthly output, provided by NASA’s Jet Propulsion Laboratory, is available online for the period 1992–2015. These observations include profiling floats (ARGO), satellite altimetry, and mass distribution derived from satellite (GRACE). These observations are consistent both with physical laws and sparse data (Wunsch, 2016). These observations can be used to calculate internally consistent global integrals and trends. These are best-fit estimates that can be tied to the climate system and used to calculate internal and consistent global integrals and trends. These are best-fit estimates that are consistent both with physical laws and sparse data (Wunsch, 2016). These observations include profiling floats (ARGO), satellite altimetry, and mass distribution derived from satellite (GRACE). Monthly output, provided by NASA’s Jet Propulsion Laboratory, is available online for the period 1992–2015.

We use Bermuda-Woods Hole A22 hydrographic data from the GLODAPv2 bottle data product to estimate volume transport and combine this with the C_\text{ant,def} estimate of the transport of uptake capacity. Geostrophic velocity is calculated from baroclinic shear, assuming a level of no motion at 3,000 m. Geostrophic transport is calculated from the product of geostrophic velocity normal to the section and cross-sectional area. We choose 3,000 m because there is transport below the 2,000 m level of no motion (Pelegri & Csanady, 1991). A deeper reference level decreases carbon transport by less than 10% but increases our Gulf Stream transport estimate to 74 Sv, bringing it closer to the 97 Sv estimate of Rossby et al. (2014). We neglect any barotropic transport; therefore our transport estimate is a low-end estimate for 2003.

\[ C_{\text{ant,def}} = C_{\text{ant}} - \langle C_{\text{ant}} \rangle_{\sigma_\theta<26.5} \]  
\[ \langle C_{\text{ant}} \rangle_{\sigma_\theta<26.5}^{2003} = 45.0 \mu \text{mol kg}^{-1} \]  

The left hand side, C_{\text{ant,def}} is the difference between the saturation concentration of C_{\text{ant}} the surface layer \((\langle C_{\text{ant}} \rangle_{\sigma_\theta<26.5})\) and the local estimate of C_{\text{ant}} \((\langle C_{\text{ant}} \rangle_{\sigma_\theta<26.5}^{2003})\) is the mean concentration of C_{\text{ant}} above the \(\sigma_\theta=26.5 \text{ kg m}^{-3}\) surface along the Bermuda-Woods Hole A22 segment in 2003.

When this calculation is made for the hindcast model, it is made for a timeseries of years. Due the continued increase in atmospheric CO2, \(\langle C_{\text{ant}} \rangle_{\sigma_\theta<26.5}\) increases over time. This means that in our model analysis C_{\text{ant,def}} becomes increasingly negative at depth over time. This is simply due to the fact atmospheric CO2 is continually increasing at a rate much faster than C_{\text{ant}} can be transferred to depth by mixing and advection.

2.5. Geostrophic Transport Estimation
We use Bermuda-Woods Hole A22 hydrographic data from the GLODAPv2 bottle data product to estimate volume transport and combine this with the C_{\text{ant,def}} estimate of the transport of uptake capacity. Geostrophic velocity is calculated from baroclinic shear, assuming a level of no motion at 3,000 m. Geostrophic transport is calculated from the product of geostrophic velocity normal to the section and cross-sectional area. We choose 3,000 m because there is transport below the 2,000 m level of no motion (Pelegri & Csanady, 1991). A deeper reference level decreases carbon transport by less than 10% but increases our Gulf Stream transport estimate to 74 Sv, bringing it closer to the 97 Sv estimate of Rossby et al. (2014). We neglect any barotropic transport; therefore our transport estimate is a low-end estimate for 2003.

2.6. State Estimate
A gridded estimate of the physical state is provided by ECCOv4, (Forget et al., 2015), a nominal 1° global ocean state estimate. ECCOv4 is based on MITgcm and is used as a best estimate of the ocean’s physical state. ECCOv4 uses adjoint data assimilation to bias correct the model with observations. This creates a state estimate that can be used to calculate internally consistent global integrals and trends. These are best-fit estimates that are consistent both with physical laws and sparse data (Wunsch, 2016). These observations include profiling floats (ARGO), satellite altimetry, and mass distribution derived from satellite (GRACE). Monthly output, provided by NASA’s Jet Propulsion Laboratory, is available online for the period 1992–2015.

We focus on a 1997–2007 period, centered on 2002, because carbonate system measurements in the gridded GLODAPv2 product are normalized to 2002.

2.7. Hindcast Model
We also employ a hindcast model from which C_\text{ant} can be precisely determined and where biogeochemistry and circulation are internally consistent. For GLODAPv2 to provide a climatological three-dimensional fields of C_\text{ant} and nutrients, significant smoothing in space and time are required. A natural consequence of this processing is to damp spatial gradients in the resulting fields. The hindcast model avoids these limitations but has significant biases (Tagklik et al., 2017). We therefore validate that the model suitably represents
Figure 2. Intermediate water \( (\sigma_\theta = 27.0–27.5 \text{ kg m}^{-3}) \) concentration of \( C_{\text{ant}}, \text{NO}_3, \) and \( \text{AOU} \) from (a, c, e) the GLODAPv2 mapped climatology and (b, d, f) a historical model simulation (CESM hindcast). Streamlines are from the velocity field of (a, c, e) the ECCOv4 state estimate and (b, d, f) the CESM hindcast simulation. Hindcast concentrations and currents sampled from 2002, and (a–c) ECCOv4 currents are the 1997–2007 mean.

3. Results

Here we evaluate the distribution of \( C_{\text{ant}} \) in the nutrient stream and calculate transport of air-sea \( C_{\text{ant}} \) uptake capacity \( (C_{\text{ant,den}}) \) into the subpolar North Atlantic. We also examine interannual variability of ventilation of nutrient stream \( C_{\text{ant}} \).

3.1. Observations and Simulation of Low \( C_{\text{ant}} \) Pathways

The Gulf Stream flows northward along the east coast of North America, separating and then flowing into the subpolar North Atlantic (Figure 2). Currents, which are visualized as streamlines (Figure 2), are averaged over the densities of nutrient stream core \( (\sigma_\theta = 27.0–27.5 \text{ kg m}^{-3}) \). The Gulf Stream separates from the coast of North America and tracks zonally at Flemish Cap off of Newfoundland (approximately 45°N, 60°W) at which point it is known as the North Atlantic Current. The western boundary current in the hindcast simulation (Figure 2, bottom row) is broader than in the more-realistic state estimate. In the hindcast, the North Atlantic Current segment is biased southward and more zonal (Figure 2). These biases are common in coarse resolution models (Tagklis et al., 2017).

In the observations/state estimate, North of the Grand Banks, elevated \( \text{NO}_3 \) is embedded in the western boundary current (Figure 2c). The narrow streak of low \( \text{NO}_3 \), and also \( \text{AOU} \), extending from Cape Hatteras to the Grand Banks is likely an interpolation artifact; other interpolations (Palter & Lozier, 2008) lack this feature. The elevated region of nutrients in the western boundary current is bounded by \( \sigma_\theta = 27.0–27.5 \text{ kg m}^{-3} \).
is known as the nutrient stream (Palter & Lozier, 2008; Pelegri et al., 1996; Williams et al., 2006). In the hindcast model, the southward bias in the North Atlantic Current shunts the simulated $\text{NO}_3$ maximum southward (Figure 2d). The fact that biases in the circulation of the hindcast simulation yields biases in $\text{NO}_3$ is consistent with circulation being the primary determinant of nutrient gradients. In both the direct observations of nutrients combined with the best estimate of the observed circulation, and in the hindcast model, the western boundary current transports high concentrations of $\text{NO}_3$ and AOU into the subpolar gyre.

Correspondence between high AOU, high $\text{NO}_3$, low $C_{\text{ant}}$, and high velocities are present in both observations and the hindcast simulation (Figure 2). AOU and $\text{NO}_3$ concentration decreases, and $C_{\text{ant}}$ concentration increases, along the streamlines leading into the subpolar gyre. South of the subpolar gyre, the GLODAPv2 $C_{\text{ant}}$ estimate is higher than the CESM hindcast. Ventilation timescales in the CESM hindcast simulation that are longer than observed (Long et al., 2013) contribute to differences between the $C_{\text{ant}}$ field in the CESM hindcast simulation and the $C_{\text{ant}}$ field from the GLODAPv2 gridded data product. Also, the GLODAPv2 TTD based $C_{\text{ant}}$ estimate is biased high by approximately 30% in intermediate waters in the subtropics due to assuming a uniform ratio of advective/diffusive transport (Steinfeldt et al., 2009). The assumption of a constant air-sea disequilibrium in TTD calculations can also contribute to a high bias in observations. The discrepancy between $C_{\text{ant}}$ concentrations in the hindcast simulation and data-based estimates is due to biases in both products.

In the observations/state estimate, high nutrients and AOU (Figures 2c and 2e) are found in layers that have not recently encountered the surface, suggesting nutrient stream waters have not had significant contact with the anthropogenic atmospheric $\text{CO}_2$ transient. The simulation also features high AOU and nutrients in the nutrient stream (Figures 2d and 2f). Consistent with an older water mass, there is a $C_{\text{ant}}$, minimum embedded within the nutrient stream in both observations (Figure 2a) and the hindcast simulation (Figure 2b).

### 3.2. Observed Vertical Structure of $C_{\text{ant}}$ at A22 (38°N)

The lower isopycnals of the nutrient stream are on density surfaces that outcrop in the subpolar North Atlantic (Pelegri et al., 1996; Sarmiento et al., 2004; Williams et al., 2006). The outcrop location is where these waters can restore the surface ocean $p\text{CO}_2$ toward its preindustrial value and support significant air-sea $C_{\text{ant}}$ fluxes. Using A22 observations, we investigate the structure and transport of $C_{\text{ant,def}}$ in the nutrient stream.

There are three water masses in the Gulf Stream region at 38°N. Distribution of all water mass properties generally follows isopycnals (Figures 3a–3d); thus we use isopycnal boundaries in our analysis. Waters down to the $\sigma_\theta = 26.5$ kg m$^{-3}$ surface are subtropical waters (Palter et al., 2005; Williams et al., 2011). These waters are low in nutrients and AOU and high in $C_{\text{ant}}$ (Figures 3a, 3b, and 3d), consistent with relatively recent ventilation. At $\sigma_\theta = 26.5–27.0$ kg m$^{-3}$ are thermocline waters that form the upper portion of the nutrient stream (Williams et al., 2011). Below this layer, between $\sigma_\theta = 27.0–27.7$ kg m$^{-3}$, lie the intermediate waters, that contain the highest nutrient concentrations. Waters in this density range outcrop in the subpolar gyre (Williams et al., 2011). Intermediate waters have a high nutrient concentration and high AOU, consistent with a large contribution from an AAIW end member (Palter & Lozier, 2008). Our analysis illustrates that these nutrient stream waters ($\sigma_\theta = 27.0–27.7$ kg m$^{-3}$) also contain very little $C_{\text{ant}}$ (Figure 3d).

As noted in section 2.3, local CFC-11 data are not used in our $C_{\text{ant}}$ estimate and thus can be used for qualitative comparison to the $C_{\text{ant}}$ distribution. The minimum of CFC-11 (Figure 3c) in the nutrient stream is in accordance with older waters with low $C_{\text{ant}}$. Waters below the nutrient stream ($\sigma_\theta > 27.7$ kg m$^{-3}$) are elevated in $C_{\text{ant}}$ and CFC-11. These waters are primarily composed of North Atlantic Deep Water, formed in the Labrador Sea, and have been ventilated more recently than the nutrient stream source waters (Hinrichsen & Tomczak, 1993; Smethie et al., 2000). Both AOU and CFC-11, independent tracers with age-dependent concentrations, indicate nutrient stream waters at this location are the oldest waters in the upper 2,000 m (Figure 3b and 3c).

### 3.3. Observed Lateral Transport of $C_{\text{ant}}$ at A22 (38°N)

Here we investigate lateral transport of $C_{\text{ant}}$ through the A22 (Bermuda-Woods Hole segment) section at approximately 38°N. We also estimate the degree to which lateral transport of $C_{\text{ant,def}}$ can sustain the subpolar North Atlantic air-sea $C_{\text{ant}}$ sink. Values given here are synoptic estimates for 2003.
Figure 3. Gulf Stream concentration of (a) NO₃, (b) AOU, (c) CFC-11, (d) $C_{\text{ant}}$. The φC₆ estimates of $C_{\text{ant}}$ are independent of local CFC-11 measurements. Gulf Stream (e) geostrophic velocity and (f) geostrophic $C_{\text{ant, def}}$ transport. A constant value of 45.0 μmol kg⁻¹ is removed from (d) to calculate $C_{\text{ant, def}}$. Negative values indicate northward transport of $C_{\text{ant}}$ uptake capacity. Observed at 38°N from the 2003 occupation of the WOCE A22 section (Figure 1). Contour lines are $\sigma_\theta$ (kg m⁻³) surfaces. The section origin is at 40.01°N, 70.01°W.

The highest estimate of $C_{\text{ant}}$ concentration is in subtropical surface waters ($\sigma_\theta < 26.5$ kg m⁻³, Table 1). This layer intersects with the Gulf Stream (Figures 3d and 3e), where we estimate velocities normal to the Bermuda-Woods Hole A22 section of over 1 m s⁻¹. Our observational estimate of volume transport normal to the Bermuda-Woods Hole section is 46 Sv. Associated with the total volume transport, 0.80 Pg C year⁻¹ of anthropogenic carbon are transported northward across the Bermuda-Woods Hole section. Due to the strong vertical gradient of $C_{\text{ant}}$, lateral $C_{\text{ant}}$ transport is maximized in the upper layers (Table 1).

| $\sigma_\theta$ (kg m⁻³) | $\theta$ (°C) | Salinity (PSU) | $C_{\text{ant}}$ (μmol kg⁻¹) | $C_{\text{ant}}$ transport (Pg C$_{\text{ant}}$ year⁻¹) | $C_{\text{ant, def}}$ transport (Pg C$_{\text{ant, def}}$ year⁻¹) | Volume transport (Sv) |
|-------------------------|--------------|----------------|-----------------------------|--------------------------------|--------------------------------|---------------------|
| <26.5                   | 18.8         | 36.3           | 45.0                        | 0.54                          | 0.14                          | 23                  |
| 26.5–27.0               | 15.9         | 36.0           | 33.0                        | 0.13                          | –0.05                         | 10                  |
| 27.0–27.5               | 11.2         | 35.4           | 17.7                        | 0.05                          | –0.11                         | 9                   |
| 27.5–27.7               | 6.9          | 35.0           | 17.2                        | 0.02                          | –0.04                         | 4                   |

Note. Negative transport of $C_{\text{ant, def}}$ indicates northward supply of uptake capacity.
Table 2

|                      | Volume transport (Sv)\(^a\) at A22 (34°N–40°N) | Overturning (Sv)\(^c\) at AR19 (48°N) |
|----------------------|-----------------------------------------------|--------------------------------------|
| Observed\(^b\)       | 46                                            |                                      |
| Hindcast             | 43                                            |                                      |
| Observed\(^d\)       | 19                                            |                                      |
| Hindcast             | 17                                            |                                      |

**Note.** Because the A22 segment does not extend zonally across the basin, calculation of meridional overturning is not possible. See Figure 1 for A22 and AR19 locations.

\(^a\)Calculated for the year 2003. \(^b\)This study. \(^c\)Calculated in density coordinates for the 1993–2000 period of Lumpkin et al. (2003). Max at \(\sigma_\theta = 27.7 \text{ kg m}^{-3}\). \(^d\)Estimate at AR19 from Lumpkin et al. (2003), the closest section to the southern boundary (49°N) of the subpolar region of Mikaloff Fletcher et al. (2006).

Observational estimates of air-sea \(C_{\text{ant,def}}\) uptake capacity (\(C_{\text{ant,def}}\), see section 2.4) are most negative between the \(\sigma_\theta = 26.5 \text{ kg m}^{-3}\) and \(\sigma_\theta = 27.7 \text{ kg m}^{-3}\) isopycnal surfaces. Waters in the nutrient stream are depleted in \(C_{\text{ant}}\) relative to surface waters and have an enhanced \(C_{\text{ant,def}}\). Our sign convention is such that a negative \(C_{\text{ant,def}}\) transport corresponds with potential downstream air-sea \(C_{\text{ant,def}}\) uptake. \(C_{\text{ant,def}}\) transport through this section within the nutrient stream (\(\sigma_\theta = 26.5–27.7 \text{ kg m}^{-3}\)) is \(-0.19 \text{ Pg } C_{\text{ant}} \text{ year}^{-1}\) (Table 1), indicating a northward transport of potential air-sea \(C_{\text{ant,def}}\) uptake. The most intense negative \(C_{\text{ant,def}}\) transport occurs on the \(\sigma_\theta = 27.0 \text{ kg m}^{-3}\) surface between 275 and 400 km (Figure 3f). There is anomalously high \(C_{\text{ant}}\) above the nutrient stream resulting in northward transport of positive \(C_{\text{ant,def}}\), indicating transport of waters that have little capacity to absorb additional \(C_{\text{ant}}\). Adjacent vertical streaks of less intense positive (440 km) and negative (500 km) \(C_{\text{ant,def}}\) transport are indicative of eddies along the section and minimally affect the large-scale transport.

3.4. Simulated Transport at A22 and the Subpolar Gyre Boundary

Here we validate simulated transport of \(C_{\text{ant,def}}\) against observations in order to determine if we can use the hindcast simulation to investigate basin-wide air-sea \(C_{\text{ant}}\) fluxes and interannual variability. We also estimate the fraction of \(C_{\text{ant,def}}\) transport at A22 that reaches the subpolar gyre. The subpolar gyre is defined as the area between 49°N and 65°N, consistent with the winter outcrop region of the nutrient stream core (Figure 1). At A22 and AR19, overturning and volume transport are well simulated in the CESM hindcast (Table 2). Simulated \(C_{\text{ant,def}}\) transport is higher than observed (Table 3). This is due to a higher surface \(C_{\text{ant}}\) concentration (Equation 1) in the hindcast simulation (observed: 45.0 μmol kg\(^{-1}\), simulated: 51.7 μmol kg\(^{-1}\)).

In observations, there is a cross stream surface \(C_{\text{ant}}\) gradient with slightly lower \(C_{\text{ant}}\) slope waters along the coast, high \(C_{\text{ant}}\) waters in the Gulf Stream, and slightly lower subtropical waters further offshore (Figure 3d). This gradient is greatly reduced in the hindcast simulation at A22, because there is no slope water simulated

Table 3

| Transport (Pg \(C_{\text{ant,def}}\) year\(^{-1}\)) |
|-----------------------------------------------|
| A22 (34°N–40°N) | 0.5×A22\(^a\) | 49°N |
| Observed          | \(-0.19\)       | \(-0.10\)       | N/A   |
| Hindcast          | \(-0.33\)       | \(-0.17\)       | \(-0.11\) |
| Transport (Pg \(C_{\text{ant,def}}\) year\(^{-1}\)), displaced A22 to match simulated Gulf Stream position |
| 60°W (~A22)\(^b\) | 0.5 × 60° W\(^b\) | 49°N |
| Hindcast          | \(-0.27\)       | \(-0.14\)       | \(-0.11\) |

\(^a\)Approximation of transport at 49°N, assuming ~50% is recirculated in the subtropical gyre (Pelegri et al., 1996; Williams et al., 2006). \(^b\)This section is comparable to the downstream distance of A22, ~800 km from where the simulated Gulf Stream separates from the coast.
at this location. This gradient does exist, however, further downstream. This downstream displacement is due to a simulated Gulf Stream that separates further downstream than is observed (Figure 2).

The A22 section is approximately 800 km northeast of Cape Hatteras, where the Gulf Stream is observed to separate from the coast. A north-south section along 60°W in the hindcast is at the observed downstream distance of A22 and is potentially more consistent with observed conditions at A22. If we sample the model at this longitude, with the same section length as A22, then volume transport is the same as at 60°W (43 Sv), but the $C_{ant,def}$ transport is closer to observations (Table 3), consistent with a Gulf Stream surface concentration (48.7 μmol kg$^{-1}$) that is closer to observed.

The observed fraction of $C_{ant,def}$ reaching the subpolar gyre, relative to the portion that is recirculated in the subtropical gyre, was calculated by others from observations of nutrient transport (Pelegri et al., 1996; Williams et al., 2006). The observed fraction is approximately 50% for layers between $\sigma_t = 26.5$–27.7 kg m$^{-3}$.

In the hindcast simulation, we can calculate this fraction directly from $C_{ant,def}$ transport. To calculate the fraction reaching the subpolar region in the hindcast, we take the $C_{ant,def}$ transport at A22 and 60°W and divide by the $C_{ant,def}$ transport at subpolar boundary (49°N). In our hindcast simulation this fraction is lower than this previous observation of 50%. We find 33% of the transport at A22 is transferred to the subpolar gyre but closer to observed if transport at 60°W is used (41%) (Table 3).

### 3.5. Air-Sea $C_{ant}$ Fluxes and the Subpolar $C_{ant,def}$ Budget

Here we compare the air-sea $C_{ant}$ flux of the CESM hindcast to observational estimates. We also calculate $C_{ant,def}$ transports into and out of the subpolar North Atlantic, between 49°N and 65°N. We analyze air-sea $C_{ant}$ fluxes and mean $C_{ant,def}$ transports for the 16 year time period 1992–2007, centered on the year 2003.

Previous studies have estimated air-sea $C_{ant}$ fluxes over quite different areas (Figure 4). Using simulated air-sea $C_{ant}$ flux from the CESM hindcast we estimate the effect varying areas has on subpolar air-sea $C_{ant}$ flux estimates. The simulated air-sea $C_{ant}$ flux is approximately twice as high in our study region as compared to the study region of Pérez et al. (2013) that is substantially smaller (Figure 4). The difference between the two prior observational estimates of the air-sea $C_{ant}$ flux (Mikaloff Fletcher et al., 2006; Pérez et al., 2013) is consistent with the difference in the simulated air-sea $C_{ant}$ flux integrated over the respective study regions. The smallest study region excludes the Labrador Sea and much of the nutrient stream outcrop region (A25–65°N; Figure 1). The inclusion of the Labrador Sea is necessary to encompass the outcrop region of the nutrient stream (Figure 5d). For our subpolar study region, we select a northern boundary of 65°N. This marks the boundary between the subpolar gyre and the Nordic Seas, which are separated physically by the Iceland-Scotland Ridge.

Using the hindcast, we estimate transport of $C_{ant,def}$ into the study region, to determine how the air-sea $C_{ant}$ flux is sustained. Northward simulated $C_{ant,def}$ transport is maximized above 830 m; therefore we select this model vertical level as the bottom boundary of our subpolar box. Deep mixing in the subpolar gyre reaches these depths every year and ventilates these waters.
Figure 5. (a–c) Budget from 0 to 830 m of $C_{ant,def}$ for the 1992–2007 period in the CESM hindcast. Orange arrow indicates the sign of fluxes supplying $C_{ant,def}$, and thus air-sea $C_{ant}$ uptake capacity, into the region. Lateral advective fluxes are density binned in (a) and (c). Black bar outline in (a) indicates the total $C_{ant,def}$ transport ($-0.11 \text{ Pg } C_{ant,def} \text{ year}^{-1}$) required to sustain simulated subpolar air-sea $C_{ant}$ flux ($-0.11 \text{ Pg } C_{ant} \text{ year}^{-1}$). Simulated fluxes (b) at the bottom boundary are vertical advection and diapycnal diffusion. All fluxes are in units of $\text{Pg } C_{ant,def} \text{ year}^{-1}$, hence the positive sign for the air-sea flux (bottom left below (c)). Red lines in (c) mark the study region, and shading is simulated mean DJFM sea surface density ($\text{kg m}^{-3}$). White density contours enclose the nutrient stream outcrop region.

Supply of low $C_{ant}$ waters, as indicated by negative $C_{ant,def}$, supports the air-sea $C_{ant}$ flux by lowering the carbon concentration of surface waters. The largest negative source of $C_{ant,def}$ to the upper 830 m of the subpolar box is from lateral advection in the nutrient stream (Figure 5). The mean nutrient stream lateral advective transport of $-0.12 \text{ Pg } C_{ant,def} \text{ year}^{-1}$ is sufficient to support the mean air-sea $C_{ant}$ flux, $-0.11 \text{ Pg } C_{ant} \text{ year}^{-1}$ (Figure 5). These waters enter the region in the upper (northward moving) limb of the overturning circulation, moving along isopycnals (Burkholder & Lozier, 2011) toward the surface. Because the circulation is rapid in the upper limb of the overturning circulation, there is little growth in atmospheric $CO_2$ during the transit, and thus there is little time for $C_{ant,def}$ concentration to change (Figure 6). The $C_{ant,def}$ that is supplied to the mixed layer in the nutrient stream is eliminated as $C_{ant}$ is added to the mixed layer by the air-sea $C_{ant}$ flux. By the time a parcel leaves the surface, $C_{ant,def}$ is $\sim 0$ (Figures 2 and 6). In the lower (southward moving) limb of the overturning, below $\sigma_g = 27.7 \text{ kg m}^{-3}$, waters move southward and $C_{ant,def}$ is removed from the subpolar region. The transport is positive below $\sigma_g = 27.7 \text{ kg m}^{-3}$ because $C_{ant,def}$ is negative and the velocity is southward, also negative, therefore advective transport, which is the product of velocity and $C_{ant,def}$, is positive. Waters below $\sigma_g = 27.7 \text{ kg m}^{-3}$ have not been in contact with the atmosphere for several years, and over time atmospheric $CO_2$ has increased. Thus the deeper waters, which have maintained relatively constant $C_{ant}$ after leaving the mixed layer in the subpolar gyre, have more negative $C_{ant,def}$. Diapycnal diffusion across the bottom boundary acts to make $C_{ant,def}$ more negative by removing carbon from the upper 830 m, while vertical advection acts to make it more positive via the downwelling of low carbon waters. The downwelling occurs primarily in the southeast corner of the study region, where the subtropical gyre extends slightly into the study region. The diapycnal diffusion $C_{ant,def}$ source is canceled by a vertical
advection $C_{\text{ant,def}}$ sink that is twice as large as the diapycnal diffusion source (Figure 5). Fluxes at the northern boundary are small. Integrating the fluxes over the region, there is a net positive growth of $C_{\text{ant,def}}$ (Figure 5, Storage), that is, a loss of uptake capacity within the subpolar box as $C_{\text{ant}}$ is absorbed from the atmosphere. Despite the loss of $C_{\text{ant}}$ uptake capacity, air-sea $C_{\text{ant}}$ uptake continues, because the surface $C_{\text{ant}}$ concentration is following the increase in atmospheric CO$_2$. As long as atmospheric CO$_2$ continues to increase, subsequent years will see more $C_{\text{ant,def}}$ supplied to the upper 830 m. In summary, the northward flux of $C_{\text{ant,def}}$ is satisfied by the air-sea $C_{\text{ant}}$ flux at the surface and the export of $C_{\text{ant,def}}$ below $\sigma_\theta = 27.7$ kg m$^{-3}$ can be attributed to increasing atmospheric CO$_2$ (Figure 6).

### 3.6. Variability of Lateral $C_{\text{ant,def}}$ Transport

We also use the CESM hindcast to examine variability of the $C_{\text{ant,def}}$ transport and air-sea $C_{\text{ant}}$ flux. We focus on the southern boundary at 49°N because we have shown above that the dominant source of $C_{\text{ant,def}}$ is provided through the southern boundary. We compare the high NAO period of the 1990s to the low NAO period of the 2000s, focusing on sustained periods of high (1992–1995) and low NAO (2003–2006) (Yashayaev & Loder, 2016).

For the two periods of interest, the simulated air-sea flux is within $\sim$0.01 Pg $C_{\text{ant,def}}$ year$^{-1}$ of the $C_{\text{ant,def}}$ transport at 49°N and 0.04 Pg $C_{\text{ant,def}}$ year$^{-1}$ at 60°W (Figure 7b, Table 3). Lateral $C_{\text{ant,def}}$ transport in the upper limb of the overturning circulation exceeds the air-sea flux for both periods. In the lower limb (southward moving), $C_{\text{ant,def}}$ is exported southward (Figures 5 and 6).

Air-sea $C_{\text{ant}}$ uptake increased (more negative) from the high NAO period (1992–1995) to the low NAO period (2003–2006) (Figure 4). An increase in ocean uptake is the expected response to the observed increase in atmospheric CO$_2$. Assuming a transient steady state, we estimate this expected increase as in Mikaloff Fletcher et al. (2006) and Gruber et al. (2019).

Anomalies shown in Figure 7a are the difference between expected and simulated variables and are the result of interannual climate variability. The air-sea $C_{\text{ant,def}}$ flux shown in Figures 5 and 6 is the same as the air-sea $C_{\text{ant}}$ flux, except that we have assigned a negative sign to the air-sea $C_{\text{ant}}$ flux, indicating atmospheric $C_{\text{ant}}$ removal. Air-sea $C_{\text{ant}}$ uptake increased less than the increase expected due to increased atmospheric...
Thus air-sea $C_{\text{ant}}$ uptake was anomalously high (more negative) in the 1992–1995 period and anomalously low (more positive) for the 2003–2006 period. At the same time, $C_{\text{ant,def}}$ transport was more negative in the early period compared to the late period, consistent with the simulated enhancement of air-sea $C_{\text{ant}}$ uptake anomalies. However, the simulated variability in $C_{\text{ant,def}}$ transport is greater than that for the simulated air-sea $C_{\text{ant}}$ flux.

Changes $C_{\text{ant,def}}$ transport are due either to changes in $C_{\text{ant,def}}$ or changes in volume transport. Anomalously negative $C_{\text{ant,def}}$ transport during the high NAO period is due to increased volume transport in the nutrient stream core (Figures 7a and 7d). The ocean response to high NAO periods is increased formation of dense Labrador Sea Water (Yashayaev & Loder, 2016). In the high NAO period of 1992–1995, simulated northward volume flux was higher in dense waters ($\sigma_\theta = 27.0–27.7 \text{ kg m}^{-3}$) that form the core of the nutrient stream (Figure 7d). During positive NAO events, formation of denser waters in the subpolar region acts to enhance the east-west density gradient and increase northward transport at depth (Lozier et al., 2010). In the low NAO period, there was reduced deep water formation and reduced volume transport in dense waters. Reduced northward transport in the nutrient stream core results in anomalously positive $C_{\text{ant,def}}$ transport (Figures 7a and 7d). This is consistent with the transition seen in air-sea $C_{\text{ant}}$ flux. During periods of positive NAO, more uptake capacity is advected northward relative to what would be expected due to atmospheric CO$_2$ increase, which allows for anomalously negative air-sea $C_{\text{ant}}$ uptake.

Denser surface waters are not necessarily limited to regions of deep convection during high NAO events. Wherever the surface is denser relative NAO neutral periods, the deeper mixed layers there will penetrate further into the nutrient stream and therefore access more negative $C_{\text{ant,def}}$. Likewise, when the state of the NAO is lower, mixed layers are shallower, and less $C_{\text{ant,def}}$ is accessed. Enhanced ventilation of the nutrient stream layer that occurs during positive NAO periods also contributes to anomalously negative air-sea $C_{\text{ant}}$ uptake.

Although there is decreased northward volume flux in the dense waters, total $C_{\text{ant,def}}$ transport becomes more negative as the NAO index transitions from high to low. The more negative northward $C_{\text{ant,def}}$ transport...
is consistent with the increase in atmospheric CO$_2$ (Figure 6b). Uptake capacity grows along with atmospheric CO$_2$ increase, permitting the continued growth of the sink over time.

4. Discussion

The nutrient stream has been demonstrated to be an important source of nutrients to the subpolar North Atlantic Ocean (Palter & Lozier, 2008; Pelegri et al., 1996; Williams et al., 2011). We show that low $C_{ant}$ waters are also transported through the subtropics and into the subpolar gyre. Our results indicate the transfer of strongly negative $C_{ant,def}$ (air-sea $C_{ant}$ uptake capacity) in intermediate waters from the subtropical to subpolar gyre sustains subpolar air-sea $C_{ant}$ uptake. The magnitude of subpolar air-sea $C_{ant}$ fluxes are only $\sim$10% relative to the advective fluxes of $C_{ant}$ at A22 0.8 Pg $C_{ant}$ year$^{-1}$. This is consistent with the $C_{ant}$ in subpolar mode water being primarily sourced from upstream air-sea $C_{ant}$ uptake in subtropical waters (Iudicone et al., 2016; Levine et al., 2011). In the absence of a supply of low $C_{ant}$ waters, surface cooling would result in air-sea $C_{ant}$ uptake and lower the DIC to alkalinity ratio, which would progressively limit $C_{ant}$ absorption (Vlker et al., 2002). Here we show that the subsurface supply of low $C_{ant}$ waters and the subpolar exposure of these waters supports significant air-sea $C_{ant}$ uptake. While most uptake occurs upstream, downstream air-sea exchange of $C_{ant}$ in denser waters establishes the subpolar region as the site of approximately half ($-0.09 \pm 0.01$ Pg $C_{ant}$ year$^{-1}$) of North Atlantic air-sea $C_{ant}$ flux ($-0.22 \pm 0.02$ Pg $C_{ant}$ year$^{-1}$)(Mikaloff Fletcher et al., 2006), and as one of the most intense regions of air-sea $C_{ant}$ flux per area.

The magnitude of the subpolar North Atlantic air-sea $C_{ant}$ sink is still an important question. Forward models and inverse estimates agree that this region is an air-sea $C_{ant}$ sink (Long et al., 2013; Mikaloff Fletcher et al., 2006). Pérez et al. (2013) suggest the region is a minor air-sea sink for $C_{ant}$, because northward flowing intermediate waters attain $C_{ant}$ saturation via air-sea exchange in the subtropics; however, a southern boundary at A25 (Pérez et al., 2013) excludes much of the western outcrop region of the nutrient stream, including the Labrador Sea, and reduces subpolar area by 50% ($7.3 \times 10^6$ km$^2$ to $3.8 \times 10^6$ km$^2$) relative to the subpolar area enclosed by the southern boundary of Mikaloff Fletcher et al. (2006). A larger subpolar region is supported by the results of Ullman et al. (2009) and global biome mapping efforts (Fay & McKinley, 2014). Our results illustrate that a 49°N southern boundary (Figure 6, Mikaloff Fletcher et al. 2006) for the subpolar North Atlantic is more fully representative of the region because it includes the important outcrop area for nutrient stream waters that carry a large air-sea $C_{ant}$ uptake capacity.

Low $C_{ant}$ intermediate waters ultimately outcrop in the subpolar region (49°N–65°N). Northward transport of $C_{ant,def}$ at A22 provides a potential air-sea $C_{ant}$ uptake capacity of $\sim$0.19 Pg C year$^{-1}$, but only some of this reaches the surface of the subpolar region. If half of the $C_{ant,def}$ transport reaches the subpolar region (Pelegri et al., 1996; Williams et al., 2006), we estimate a subpolar North Atlantic air-sea $C_{ant}$ sink of $\sim$0.10 Pg C year$^{-1}$. Our air-sea $C_{ant}$ sink estimate is comparable to the $-0.09 \pm 0.01$ Pg C year$^{-1}$ ocean inverse estimate of Mikaloff Fletcher et al. (2006) and the CESM hindcast model used here.

North Atlantic carbon cycle variability occurs in response to the NAO (Corbière et al., 2007; Pérez et al., 2013; Schuster et al., 2013; Thomas et al., 2008; Ullman et al., 2009; Watson et al., 2009). We find enhanced nutrient stream $C_{ant,def}$ transport during high NAO periods, consistent with an enhanced subpolar air-sea $C_{ant}$ sink during these periods (Fröb et al., 2016; Pérez et al., 2013). A recent observational analysis indicates that after the high NAO period of the early 1990s, subpolar $C_{ant}$ accumulation was anomalously low, with the greatest North Atlantic reductions occurring in the outcrop region of the nutrient stream (Gruber et al., 2019). The results of Gruber et al. (2019) are compatible with nutrient stream ventilation variability, illustrated here, as a driver of long-term subpolar air-sea $C_{ant}$ flux variability. Decreased transport in dense waters in the CESM hindcast is consistent with reduced deep water formation and reduced nutrient stream ventilation. Reductions to nutrient stream ventilation are consistent with reduced vertical supply of $C_{ant}$ (Ullman et al., 2009) and reduced air-sea fluxes during neutral NAO periods in hindcast simulations of the carbon cycle (Thomas et al., 2008; Ullman et al., 2009). Over a similar period, observations indicate that the air-sea $pCO_2$ gradient decreased in the vicinity of the North Atlantic Current (McKinley et al., 2011; Schuster et al., 2009).

In summary, our proposed mechanism is consistent with historical subpolar carbon cycle variability and enhanced subpolar air-sea $C_{ant}$ uptake when the wintertime NAO index is high.

Here we approach this problem with a traditional Eulerian framework in a coarse resolution model. The CESM hindcast suggests more $C_{ant,def}$ transport than observed at A22, and a greater fraction recirculates in the subtropical gyre than suggested by observations (Pelegri et al., 1996; Williams et al., 2006). Lagrangian
methods could be used to resolve where the nutrient stream waters eventually outcrop (Burkholder & Lozier, 2014). These methods could yield more accurate estimates of the fraction of nutrient stream waters reaching the subpolar gyre.

5. Conclusion

The nutrient stream has been defined as the region of dense waters within the western boundary current that is elevated in nutrients. As indicated by high AOU and CFC-11, source waters of the nutrient stream (\(\sigma_T = 26.5–27.7 \text{ kg m}^{-3}\)) are some of the oldest waters in the upper 2,000 m. They have been sequestered from the atmosphere for long enough that they are highly depleted in \(C_{\text{ant}}\) relative to subtropical waters. These waters are from the tropical Atlantic (Palter & Lozier, 2008) and ultimately may have an AAIW source (Sarmiento et al., 2004; Sen Gupta & England, 2007).

Global ocean \(C_{\text{ant}}\) uptake occurs primarily in the North Atlantic and Southern Ocean (Khatiwala et al., 2013; Mikaloff Fletcher et al., 2006; Sabine et al., 2004). Here we focus on the North Atlantic which features high intensity air-sea fluxes (Landschützer et al., 2014; Takahashi et al., 2009) and rapid sequestration of \(C_{\text{ant}}\) into the deep ocean (Fröb et al., 2016; Pérez et al., 2013). We find that the low \(C_{\text{ant}}\) waters of the nutrient stream provide significant air-sea \(C_{\text{ant}}\) uptake capacity to the surface of the subpolar North Atlantic.

Upward tilt of isopycnals in the Gulf Stream region allows for the low \(C_{\text{ant}}\) waters to be laterally transported into the Gulf Stream along isopycnals and thus preserving their properties. \(C_{\text{ant}}\) uptake capacity of \(-0.19 \text{ Pg} \ C_{\text{ant}} \text{ year}^{-1}\) is transported northward at A22. The hindcast simulation used here indicates that the simulated fraction of A22 \(C_{\text{ant}}\) uptake capacity transport that reaches the subpolar North Atlantic is sufficient to support the simulated air-sea \(C_{\text{ant}}\) flux.

Observations of nutrient transport (Pelegri et al., 1996; Williams et al., 2006) suggest that half of the transport at A22 reaches the subpolar North Atlantic, while the hindcast simulation indicates 33–41% of the air-sea \(C_{\text{ant}}\) uptake capacity reaches the subpolar North Atlantic. If half of the \(C_{\text{ant,def}}\) transport at A22 reaches the surface in the subpolar North Atlantic, it would suggest a subpolar North Atlantic uptake capacity of \(-0.10 \text{ Pg} \ C_{\text{ant}} \text{ year}^{-1}\) in 2003. This is comparable to the total subpolar \(C_{\text{ant}}\) uptake, \(-0.09 \pm 0.01 \text{ Pg} \ C_{\text{ant}} \text{ year}^{-1}\) (Mikaloff Fletcher et al., 2006).

The hindcast simulation indicates that differences between the Mikaloff Fletcher et al. (2006) estimate and that of Pérez et al. (2013) is that the smaller study region of the latter excludes regions of the nutrient stream outcrop. Ventilation of low \(C_{\text{ant}}\) in nutrient stream waters fully sustains a significant subpolar North Atlantic air-sea \(C_{\text{ant}}\) sink.

In the future, circulation and biogeochemistry in the subpolar North Atlantic are projected to undergo significant changes (Goris et al., 2018; Halloran et al., 2015; Tagkli et al., 2017; Whitt, 2019). \(C_{\text{ant}}\) uptake is projected to decline mid-century after an initial period of growth (Halloran et al., 2015). CMIP5 models project that \(O_2\) inventory is likely to decline, but patchy regions of \(O_2\) increase may also occur, driven by declines in AOU transport in the nutrient stream (Tagkli et al., 2017). Analyzing the historical response to anthropogenic forcing of the subpolar air-sea \(C_{\text{ant}}\) sink and nutrient stream, as well as the future response (e.g., Whitt, 2019) can improve our understanding of the mechanisms driving subpolar \(C_{\text{ant}}\) uptake.

Earth-system models and observations, utilized in complementary fashion, provide a potent framework for the study of regional air-sea \(C_{\text{ant}}\) uptake in the present and future.

Data Availability Statement

All biogeochemical and hydrographic data (GLODAPv2) are available online via NOAA OCADS (https://www.nodc.noaa.gov/ocads/). ECCOv4 output is available online via NASA JPL (https://ecco.jpl.nasa.gov), and the CESM hindcast output is available the NCAR Earth System Grid (https://www.earthsystemgrid.org). Colorbars used for plotting are from the cmocean package for MATLAB and Python (https://matplotlib.org/cmocean/). This package has colorbars that consider both accurate presentation and accessibility (Thyng et al., 2016).

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Acknowledgments

We would like to thank the editor and the four anonymous reviewers for the time they took to provide thoughtful comments. Their comments have greatly improved this manuscript.
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