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The ^3He flux gauge in the Sargasso Sea: a determination of physical nutrient fluxes to the euphotic zone at the Bermuda Atlantic time series site

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Abstract

We provide a new determination of the annual mean physical supply of nitrate to the euphotic zone in the western subtropical North Atlantic based on a three year time-series of measurements of tritiogenic ^3He from 2003 to 2006 in the surface ocean at the Bermuda Atlantic Time-series Study (BATS) site. We combine the ^3He data with a sophisticated noble gas calibrated air–sea gas exchange model to constrain the ^3He flux across the sea–air interface, which must closely balance the upward ^3He flux into the euphotic zone. The product of the ^3He flux and the observed subsurface nitrate- ^3He relationship provides an estimate of the minimum rate of new production in the BATS region. We also applied the gas model to an earlier time series of ^3He measurements at BATS in order to recalculate new production fluxes for the 1985 to 1988 time period. The observations, despite an almost three-fold difference in the nitrate- ^3He relationship, yield a roughly consistent estimate of nitrate flux. In particular, the nitrate flux from 2003–2006 is estimated to be $0.65 \pm 0.3 \text{ mol m}^{-2} \text{ y}^{-1}$, which is $\sim 40\%$ smaller than the calculated flux for the period from 1985 to 1988. The difference between the time periods, which is barely significant, may be due to a real difference in new production resulting from changes in subtropical mode water formation. Overall, the nitrate flux is larger than most estimates of export fluxes or net community production fluxes made locally for BATS site, which is likely a reflection of the larger spatial scale covered by the ^3He technique and potentially also by decoupling of ^3He and nitrate during obduction of water masses from the main thermocline into the upper ocean.

1 Introduction

Primary production in the subtropical oligotrophic gyres has been an active area of study for decades. In particular, scientists have long puzzled over the seemingly paradoxical drawdown of summertime dissolved inorganic carbon despite no visible source of nutrients (Michaels et al., 1994). Numerous studies using geochemical

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tracers, sediment traps, and bottle incubations have been performed at the Bermuda Atlantic Time-Series Study (BATS) site over the past several decades (e.g. Brew et al., 2009; Jenkins and Doney, 2003; Jenkins and Goldman, 1985; Spitzer and Jenkins, 1989; Gruber et al., 1998; Stanley et al., 2012; Stewart et al., 2011; Buesseler et al., 2008; Maiti et al., 2009, 2012; Owens et al., 2013; Lomas et al., 2010), in order to quantify various aspects of biological production and to shed light on this enigma. Floating sediment traps give a direct measure of export production but may be biased by collection efficiency due to hydrodynamic biases and swimmers (Buesseler, 1991), as well as by the limited amount of time they are in the water. Bottle incubations though primarily used to determine net primary production (Marra, 2002, 2009), can also be conducted to give determinations of new production when conducted with ^{15}N (Dugdale et al., 1992). Bottle incubations give useful information but may be limited by so-called bottle effects of constraining organisms to a bottle (Peterson, 1980; Harrison and Harris, 1986; Scarratt et al., 2006). Geochemical tracers give large scale averages of rates of new production, net community production, or export production. These rates, however, can be difficult to interpret since quantitative interpretation of the tracer data often depends on estimates of physical transport. Thus it is useful to calculate rates of production using numerous approaches and to compare them.

One approach that has been used before in the Sargasso Sea is to estimate a lower bound of new production by calculating the upward physical nutrient flux (Jenkins and Doney, 2003; Jenkins, 1988a). The global inventory of natural tritium has been dwarfed by the production of so-called “bomb tritium” that was created during the atmospheric nuclear weapons tests in the 1950s and 1960s (Weiss and Roether, 1980). This tritium was deposited in large part in the Northern Hemisphere (Doney et al., 1992; Stark et al., 2004) and has subsequently entered the oceanic thermocline and abyss by subduction, water mass formation, mixing, and advection (e.g., Rooth and Ostlund, 1972; Ostlund et al., 1974; Broecker and Peng, 1980). Tritium, which has a half-life of 12.31 years (MacMahon, 2006), decays to ^3He , a stable, inert, and rare isotope of helium. Over the decades since the bomb-transient, a significant inventory of this isotope has accrued

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within the main thermocline of the North Atlantic. There is evidence of an efflux of this isotope via gas exchange from the surface ocean (Jenkins, 1988b, c). Inasmuch as this tritiogenic excess ^3He has a nutrient-like distribution in the thermocline – it is small in the surface ocean due to gas exchange loss and reaches a maximum within the thermocline due to in situ tritium decay – it is tempting to argue that the physical return of this isotope to the shallow ocean can be used as a “flux gauge” to determine the rate of physical nutrient supply to the euphotic zone (Jenkins, 1988c; Jenkins and Doney, 2003). Here we report the results of a three year time-series of helium isotope measurements taken approximately monthly between 2003 and 2006 in the surface ocean near Bermuda that allow yet another determination of this nutrient flux. We compare the calculated nutrient flux to the nutrient flux determined at the same location using the same method for the period of 1985–1998, as well as to export production fluxes calculated in the Sargasso Sea for the time period of 2003–2006.

2 Methods

2.1 Data collection

Samples for ^3He , a suite of noble gases, and tritium were collected at the BATS site (31.7° N, 64.2° W) on core BATS cruises at approximately monthly resolution between April 2003 and April 2006. The BATS site, located in the subtropical North Atlantic, is representative of a typical oligotrophic gyre. Much biogeochemical research has occurred at that site, because of the long-standing time-series located there (Lomas et al., 2013). In particular, as part of the regular time-series, export fluxes are estimated monthly from surface-tethered floating, upper-ocean sediment traps (Lomas et al., 2010) and rates of net primary production are estimated monthly from radiocarbon bottle incubations (Steinberg et al., 2001). In addition, other researchers have measured export using ^{234}Th (Maiti et al., 2009), neutrally buoyant sediment traps (Owens et al., 2013), or apparent oxygen utilization rates (Stanley et al., 2012; Jenkins,

1980). Net community production has been estimated from the seasonal accumulation of O_2/Ar (Spitzer and Jenkins, 1989) and the drawdown of dissolved inorganic carbon (Gruber et al., 1998; Brix et al., 2006; Fernandez-Castro et al., 2012). New production has been estimated from bottle incubations (Lipschultz, 2001; Lipschultz et al., 2002) and has also been studied using nitrogen isotopes (Fawcett et al., 2014; Knapp et al., 2008).

The 3He and noble gas samples for this study were collected from Niskin bottles by gravity feeding through tygon tubing into valved 90 mL stainless steel cylinders. Typically 22 samples were collected within the upper 400 m, and thus depending on mixed layer depth, there were usually several samples collected within the mixed layer. Within 24 h of sampling, the gas was extracted from the water stored in the cylinders into ~ 30 mL aluminosilicate glass bulbs. The bulbs were then brought to the Isotope Geochemistry Facility at WHOI where they were analyzed for 3He , 4He , Ne, Ar, Kr, and Xe using a dual mass spectrometric system with the 3He being analyzed by a magnetic sector mass spectrometer and the other noble gases being analyzed by a quadrupole mass spectrometer (Stanley et al., 2009a). In particular, the magnetic sector mass spectrometer for 3He measurements was a purposefully constructed, branch tube, statically operated, dual collector instrument equipped with a Faraday cup and a pulse counting secondary electron multiplier. Precision of the 3He measurements, based on duplicates, was 0.15%. The focus of this paper is on the 3He measurements, but the other noble gases were used to calculate gas exchange fluxes (Stanley et al., 2009b), which is an important term in the calculation of 3He flux from the 3He data.

Samples for tritium were collected from the same Niskins by gravity feeding through tygon tubing into 500 mL argon-filled flint glass bottles, as described in Stanley et al. (2012). The tritium samples were degassed at the Isotope Geochemistry Facility at WHOI (Lott and Jenkins, 1998), and then the resulting 3He ingrowth was measured on a purposefully constructed, branch tube dual collector magnetic sector mass spectrometer (a different one than the one used above for 3He samples). The resulting

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tritium concentrations were used to correct for tritium ingrowth in the ^3He samples between time of collection and time of measurement (see Sect. 3.3).

2.2 Calculation of fluxes

The nitrate flux was calculated in a similar way as is described in Jenkins and Doney (2003). The most notable difference was that in this study the dynamic solubility equilibrium value of ^3He was modeled taking both solubility and bubble injection into account, as described in more detail below. To calculate the nitrate flux, first a ^3He flux was calculated and then the slope of the nitrate : ^3He ratio was applied. The ^3He flux was calculated from the gas exchange parameterization of Stanley et al. (2009b), which had been devised specifically from the noble gas samples collected at the same time as the ^3He samples and thus is well suited to the study site and sampling conditions. In particular, the ^3He flux (F_{He3}) was calculated as the product of a gas transfer velocity k , as determined in Stanley et al. (2009b), and the difference in concentration between the measured ^3He concentration (C) and the dynamic solubility equilibrium value (C_{eq}):

$$F_{\text{He3}} = k \cdot (C - C_{\text{eq}}) \quad (1)$$

Laboratory experiments have determined the isotope effect in solution for helium in water as a function of temperature (Benson and Krause, 1980). Given that the helium isotope ratio may be further affected by isotopic fractionation in molecular diffusion (Bourg and Sposito, 2008) associated with the balance between wave-induced bubble trapping and air–sea exchange (Fuchs et al., 1987; Jenkins, 1988a) we have used our observations of the full suite of noble gases on these samples to develop a much more complete model of this equilibrium isotope effect. Thus the dynamic solubility equilibrium value for ^3He , C_{eq} , was determined by adding ^3He isotopes to a one dimensional Price–Weller–Pinkel (PWP) model (Price et al., 1986) forced by 6 hourly NCEP reanalysis forcing (Kalnay et al., 1996) and QuikSCAT winds from the BATS site (Stanley et al., 2006, 2009b). The model used the temperature dependent solubility of

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^3He from Benson and Krause (1980) and the molecular diffusivity value from Bourg and Sposito (2008). The calculated dynamic solubility equilibrium is sensitive to the amount of air injection, and thus the other noble gases were used to constrain the air injection (Stanley et al., 2009b). Calculations of the dynamic solubility equilibrium and the flux of ^3He were also made using NCEP reanalysis winds instead of QuikSCAT winds. Note that when the NCEP reanalysis winds were used in the model, the gas exchange parameterization of Stanley et al. (2009b) was modified to a parameterization that was calculated using NCEP winds. For example, the gas exchange scaling factor is 0.97 when using QuikSCAT winds (as reported in Stanley et al., 2009b) but is only 0.7 using NCEP winds.

The ^3He flux is then corrected for the flux due to in situ tritium decay ($F_{\text{HeFromTrit}}$):

$$F_{\text{HeCorr}} = F_{\text{He}} - F_{\text{HeFromTrit}} \quad (2)$$

The flux of helium from tritium decay is calculated by using the radioactive decay equation, the half-life of tritium (12.31 years), and the mixed layer tritium concentrations measured in this study.

The nitrate flux (F_{NO_3}) was then calculated as the product between the corrected ^3He flux and the nitrate : ^3He ratio (R):

$$F_{\text{NO}_3} = F_{\text{HeCorr}} \times R \quad (3)$$

The ratio R was calculated by determining the slope of a type II regression of NO_3 vs. ^3He for samples measured in the upper 400 m of water during the three year time series ($N = 218$). Only data with $[\text{NO}_3] > 2 \text{ } \mu\text{mol kg}^{-1}$ were used in the regression since water with NO_3 concentration below this threshold represents water in the euphotic zone where ^3He and NO_3 are decoupled.

3 Results and discussion

3.1 The fluxes of helium-3 and nitrate

The ^3He and tritium data collected in this study between 2003 and 2006 are presented in Fig. 1. The gradient of ^3He with depth is clearly visible. In contrast, tritium has a more uniform distribution with depth in the upper 300 m. The lack of excess ^3He in the mixed layer (mixed layer is demarcated by thick black line) is because of air–sea gas exchange, which results in a flux of excess ^3He out of the ocean into the atmosphere. This sustained air–sea gas exchange results in a decreasing inventory of tritiogenic ^3He in the ocean through time. Multiple measurements within the mixed layer were averaged in order to calculate the mixed layer concentrations of ^3He (Fig. 2a). The dynamic solubility equilibrium (blue curve on Fig. 2a) is significantly smaller than the ^3He concentrations, resulting in a sea to air flux of ^3He (Fig. 2c).

Additionally, since we now have a better understanding of the dynamic solubility equilibrium, both because of the extensive information on gas exchange garnered by the noble gases and because of more accurate estimates of molecular diffusivity of ^3He , we also have recalculated the ^3He and nitrate fluxes for the data from 1985–1988 that was originally presented in Jenkins and Doney (2003). Thus the ^3He concentrations from 1985–1988 as well as the dynamic solubility equilibrium for that time-period are presented in Fig. 2b. Note the difference of scales in Fig. 2a and b. There is much less ^3He in 2003–2006 than in the 1980s because of a decreased ^3He source in the thermocline due to tritium decay over time and decades of outgassing of ^3He .

The average ^3He flux, corrected for tritium ingrowth, over the 2003–2006 time period is calculated to be $7.9 \pm 1 \text{ pmol m}^{-2} \text{ y}^{-1}$ (Table 1). The flux due to tritium ingrowth in the mixed layer during this period was $1.2 \pm 0.1 \text{ pmol m}^{-2} \text{ y}^{-1}$. The integrated ^3He flux is multiplied by a $\text{NO}_3 : ^3\text{He}$ ratio of $82.9 \times 10^9 \pm 2 \times 10^9 \text{ mol NO}_3 \text{ mol}^{-1} ^3\text{He}$ (Fig. 3) in order to calculate a NO_3 flux of $0.66 \pm 0.2 \text{ mol N m}^{-2} \text{ y}^{-1}$. The nitrate flux calculated by

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the flux gauge method used here represents the lower bound of new production in the northern half of the subtropical gyre. It represents a lower bound estimate because it only includes the new production based on the upward physical transport of nutrients. It does not include any new production due to nitrogen fixation, zooplankton migration, or atmospheric deposition of nitrate. At BATS, nitrogen fixation has been estimated to be 0.03 to 0.08 molN m⁻² y⁻¹ (Singh et al., 2013; Knapp et al., 2008), which is equivalent to 5 to 12 % of the new production we report from the flux gauge method. Zooplankton migration from 2003 to 2006 has been estimated to support a new production of 2 gC m⁻² y⁻¹ (Steinberg et al., 2012), which is equivalent to 0.025 molN m⁻² y⁻¹ using the revised Redfield ratios of Anderson and Sarmiento (1994), and thus is only 4 % of the new production rate estimated by the flux gauge technique. Estimates of the nitrate supply due to atmospheric deposition range from 0.006 to 0.026 molN m⁻² y⁻¹ (Singh et al., 2013; Knapp et al., 2010), thus being at most 4 % of the new production flux estimated here from the flux gauge method. Thus in total, the sources of new nitrate that are not accounted by the flux gauge method may mean that the new production estimate given here is only about 80 to 85 % of the total new production rate. The flux estimate represents the northern half of the gyre – rather than just the BATS site – because the water in the thermocline that is vertically transported at the BATS site sources from the Northern half of the gyre (Talley, 2003).

The nitrate fluxes calculated with the NCEP wind-derived ^3He fluxes are very similar to those calculated by QuikSCAT winds (Table 1). This is because the gas exchange parameterizations we used to calculate the flux from the ^3He concentration data and to calculate the dynamic solubility equilibrium were separately tuned to observed noble gas data for QuikSCAT and NCEP. We were able to do this since we had the wealth of noble gas data collected concurrently allowing for a good model of air–sea gas exchange with two different wind products.

3.2 Comparison to 1980s fluxes

The estimated nitrate flux for the period between 1985 to 1988 is 50 % larger than the nitrate flux for the 2003–2006 period, though over half of this difference can be accounted for by uncertainties in the flux estimates. For 1985–1988, our recomputed nitrate flux estimate is $1.05 \pm 0.2 \text{ mol N m}^{-2} \text{ y}^{-1}$ (Table 1), which is 25 % larger than the nitrate flux calculated for the same time period in Jenkins and Doney (2003). This difference between the 1985–1988 fluxes calculated here vs. those calculated in Jenkins and Doney (2003) stems from this calculation using a well-modeled dynamic solubility equilibrium. In the earlier study, we did not have the other noble gas data nor updated estimates of molecular diffusivity (Bourg and Sposito, 2008) and thus employed a simpler and likely less accurate estimate of the dynamic solubility equilibrium.

It is interesting to note that although the nitrate flux in 1985–1988 is only 50 % larger than the nitrate flux in 2003–2006, the ^3He flux in 1985–1988 is 300 % larger than the ^3He flux in 2003–2006. This is because in the 1980s, there was a much larger tritium inventory and consequently larger concentrations of ^3He in the main thermocline (Fig. 4). However, the slope of the $\text{NO}_3 : ^3\text{He}$ relationship also changes with time. The distribution of nutrients in the main thermocline are in an approximate steady state established by a balance between nutrient release by in situ remineralization of organic material and removal by physical processes related to ventilation, advection, and mixing. The corresponding thermocline distribution of tritiogenic ^3He is evolving as a transient tracer. Over time, as the bomb-tritium pulse penetrates the thermocline, the resultant ^3He maximum deepens and broadens (Jenkins, 1998). Consequently the relationship between ^3He and nutrients is changing with time. Figure 3 is a plot of the $\text{NO}_3 : ^3\text{He}$ relationship for the upper 500 m of the water column near Bermuda at four points in time. Notably, the slope of the $\text{NO}_3 : ^3\text{He}$ relationship has increased by over a factor of greater than two in the approximately 25 years spanned by this data. The fact that despite almost a factor of 3 difference in $\text{NO}_3 : ^3\text{He}$ ratio, the estimated nitrate

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flux from this method is almost the same in 2003–2006 as in 1985–1988 illustrates the robustness of this technique.

While the nitrate flux is broadly similar between the two time periods, there is still a 50 % difference with the flux being larger in 1985–1988 than in 2003–2006. What can account for this difference? It is not because of NCEP winds being used in the 1985–1988 calculation and QuikSCAT winds being used in the 2003–2006 calculation, because even if we do the 2003–2006 calculation with NCEP winds, we still get a 40 % difference between the flux in the two different decades (Table 1). It also is not likely due to the 1985–1988 data being from Hydrostation S whereas the 2003–2006 data is from BATS. Those two sites are only 28 km apart and since the ^3He flux gauge estimate is reflection of a much broader region, the relatively small difference in locations of samples likely does not play a role. It could be, in part, due to a time lag between the evolving subsurface NO_3 - ^3He ratio and surface fluxes. Most likely, however, it is due to a real elevation in new production in the late 1980s compared to the 2003–2006 period. Winter mixed layers in the two time periods are similar, with the exception of a shallower than typical winter mixed layer depth in 1986, and thus are likely not an explanation for the difference in production between the periods.

Lomas et al. (2010) observed significant changes in export production at BATS over time, with the period between 1988 and 1995 having lower export fluxes than the period from 1995 to 2008. They attributed these changes to a shift in the North Atlantic Oscillation (NAO) from positive in the 1988 to 1995 period to neutral in the 1996 to 2008 period. Our older data is from 1985 to 1988 and was not included in the Lomas et al. (2010) study. The winter NAO index (JFM) which has been shown to be most sensitive to changes in subtropical mode water formation (Billheimer and Talley, 2013) and primary production (Lomas et al., 2010) was -1.2 , 0.2 , and -1.1 for 1985, 1986 and 1987 respectively. It was -0.3 , -0.5 , and -0.6 for 2004, 2005, and 2006 respectively. A more negative winter NAO, as was mostly seen in 1985–1988 period, according to Lomas et al. (2010) would be associated with higher production, which is indeed what we found in this study.

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A more negative NAO is usually correlated with a greater production of subtropical mode waters (STMW) via enhanced surface buoyancy loss and vertical convection (Billheimer and Talley, 2013). Indeed, estimates of Kelly and Dong (2013) suggest that there was increased formation of STMW in 1985–1988 compared to 2003–2006. We thus find higher rates of new production are associated with time periods of higher generation of STMW. This is in contrast to the hypothesis of Palter et al. (2005) who suggested that increased STMW production would lead to a reduction in primary production due to decreased nutrients below the mixed layer in the vertically-homogenized mode water region since the decreased nutrients would lead to a smaller nutrient supply from the main thermocline below the mode water region and thus to smaller rates of primary production.

The highest annual flux in the 1985–1988 period comes from 1987 (Fig. 2d). Interestingly, while the NAO index of 1987 was similar to that of 1985 and 2003–2006, the NAO index of 1986 was positive. It has been shown that chlorophyll correlates better with NAO index at BATS using a one year time lag (Cianca et al., 2012). Thus potentially the higher fluxes we see in 1987 are a result of the higher NAO index in 1986. However, this would be counter to the general trend suggested by Lomas et al. (2010) and seen in the rest of our data of higher rates of production with more negative NAO indices.

3.3 Seasonal cycle

A seasonal cycle in ^3He flux is observed in both the 1985–1988 time period and the 2003–2006 time period (Fig. 5). The ^3He fluxes are highest in winter-time when the deep winter mixed layers at BATS mine water from the seasonal thermocline, bringing up higher amounts of ^3He and nitrate. But even in the summer, there is an upward flux of ^3He suggesting an upward flux of nitrate. There is no observable nitrate in the summer mixed layer at BATS (Michaels et al., 1994; Steinberg et al., 2001) likely because the organisms consume all the nitrate as soon as it enters the euphotic zone. Thus the lack of observable nitrate, long known at BATS, does not mean that nitrate was never there. Hence the “paradox” of how summertime production can be supported

at BATS without observable nutrients is in some sense answered by this clear sign that there is an upward nutrient flux, even in the summer. This supports the recent finding of Fawcett et al. (2014) showing evidence of nitrate supply to the mixed layer at BATS even in the summer.

3.4 Comparison to other rates of biological productivity at BATS

The rate of new production estimated by the helium flux gauge technique presented in this study is larger than most of the rates of new production, net community production or export production at BATS derived from other geochemical tracer approaches. Over long periods of time and long spatial scales, new production, net community production and export production should be equal (Dugdale and Goering, 1967). In carbon units, using the revised Redfield ratio of Anderson and Sarmiento (1994) of 106 : 16, new production estimated in this study was $4.6 \pm 1.2 \text{ mol C m}^{-2} \text{ y}^{-1}$ in 2003–2006 and $6.96 \pm 1.3 \text{ mol C m}^{-2} \text{ y}^{-1}$ in 1985–1988. As noted above, these rates represent new production over the Northern half of the subtropical gyre.

Export fluxes as estimated by apparent oxygen utilization rates (AOUR) also represent fluxes over a similar northern region (Jenkins, 1980). Tritium samples were collected and used in conjunction with ^3He and O_2 data from the same cruises in 2003–2006 that the ^3He data in this paper come from to estimate apparent oxygen utilization rates (Stanley et al., 2012). The AOUR values were integrated to 500 m to yield a lower bound on annual export from the remineralization and oxygen consumption between 200 and 500 m of $2.1 \pm 0.5 \text{ mol C m}^{-2} \text{ y}^{-1}$. Thus the fluxes estimated by the helium flux gauge technique are nearly a factor of two greater than the fluxes by AOUR, even though both represent a large geographical region.

A more local estimate of production comes from seasonal drawdown of DIC at BATS or by seasonal accumulation of O_2 with respect to Ar. Both techniques rely on the fact that photosynthesis produces O_2 and consumes CO_2 whereas respiration produces CO_2 and consumes O_2 . Thus the seasonal changes in O_2 or CO_2 constrain the net balance between photosynthesis and respiration. On the same cruises as data for

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^3He flux gauge technique were collected, the seasonal accumulation of O_2 and Ar was measured and used to estimate rates of net community production of 1.2 to 2.4 molC m $^{-2}$ y $^{-1}$ (Stanley, 2007). Notably, this rate is similar to that of the AOUR estimate and a factor of 2 smaller than the ^3He flux gauge estimate. The seasonal accumulation of oxygen and argon has been used at other time periods to estimate the rate of net community production at BATS to be 2.2 to 3 molC m $^{-2}$ y $^{-1}$ (Spitzer and Jenkins, 1989; Luz and Barkan, 2009). Seasonal drawdown of DIC directly as well as the change in isotopic composition of ^{13}C of DIC have been used to estimate annual net community production fluxes of 1.7 to 4.9 molC m $^{-2}$ y $^{-1}$ (Gruber et al., 1998; Brix et al., 2006; Fernandez-Castro et al., 2012). The upper end of this range approximates the rate of new production we find here using the flux gauge technique. Interestingly, the DIC drawdown and O_2/Ar approaches reflect a smaller spatial scale than the AOUR estimates, but at least in some cases agree better with the ^3He flux gauge approach.

On even smaller spatial and temporal scales, ^{234}Th has been used to estimate export fluxes at BATS, resulting in rates of export production calculated to be 0.3 to 0.8 molC m $^{-2}$ y $^{-1}$ (Maiti et al., 2009) These fluxes are much smaller than the fluxes estimated by other geochemical tracers, which may in part be due to the fact that the ^{234}Th technique does not include the contribution of export due to DOC whereas the other geochemical techniques do. DOC export in the Sargasso Sea has been estimated to be up to 1 molC m $^{-2}$ y $^{-1}$ (Hansell et al., 2012).

Why is the helium flux gauge technique yielding rates of new production at the high end of the range of rates from other geochemical tracers? In part this may be due to the broader spatial coverage of the flux gauge technique, but that is not enough to explain fully the discrepancy since the AOUR technique has similar spatial area but smaller fluxes. One reason may be that ^3He and NO_3 are decoupled during obduction in the northern part of the gyre. The northwest Sargasso Sea, where the warm waters of the Gulf Stream leave the North American continent, is characterized by large latent heat fluxes and substantial downstream winter mixed layer deepening (Worthington, 1972). In effect, upper thermocline isopycnals outcrop, a process referred to as obduction (Qiu

and Huang, 1995). This outcropping brings remineralized nutrients and tritiogenic ^3He back to the seasonal layer. Whereas the time constant associated with nutrient removal by biological processes is a matter of days, the exchange time-scale for tritiogenic ^3He loss to the atmosphere from a deep mixed layer may be several weeks. In this respect the nutrients may have been removed while the ^3He “signal” may persist, so the ^3He flux gauge may measure not only local new production, but may also hold a more “regional” memory of the upstream, previous winter’s production.

3.5 Uncertainties and sensitivity studies

There are a number of sources of uncertainty in the estimate of nitrate fluxes from the helium flux gauge technique. Here we describe these uncertainties and the results of sensitivity studies examining the effect of the sources of error. Table 2 lists the main sources of uncertainty in the calculations. One of the largest sources of uncertainty is uncertainty in the gas transfer velocity k (Eq. 1). Stanley et al. (2009b) illustrates how the time-series of noble gases collected concurrently with this data results in uncertainties of 14 % in the gas transfer velocity k . Since k is directly used to calculate the ^3He air–sea flux from the difference between measured ^3He concentration and dynamic solubility equilibrium, this uncertainty directly translates to a 14 % uncertainty in ^3He flux and ultimately in nitrate flux.

The second largest source of uncertainty in the nitrate flux is the uncertainty in the determination of dynamic solubility equilibrium. The largest uncertainty in the dynamic solubility equilibrium is caused by uncertainties in the molecular diffusivity of ^3He with respect to ^4He . The dynamic solubility equilibrium is sensitive to the molecular diffusivity due to the relative diffusive gas exchange of ^3He vs. ^4He (i.e. Schmidt number dependence) and due to the effect of air injection of partially trapped bubbles – during air injection, ^3He diffuses more quickly out of the bubbles than ^4He . We ran sensitivity studies with the range of molecular diffusivities estimated by Bourg and Sposito (2008)

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and found that the ^3He flux changed by $\pm 10\%$ depending on the molecular diffusivities used.

We also ran sensitivity studies to calculate the dynamic solubility equilibrium and the resulting ^3He and nitrate fluxes using a range of gas exchange parameters. Specifically, we use many of the parameter sets determined in Table 1 of Stanley et al. (2009b) including the sets of parameters determined for different physical parameters in the model and different weightings of the cost function. We found that the dynamic solubility equilibrium changed by only a small amount in these scenarios so that the overall SD of the ^3He flux for all the different scenarios was only 2%. We add 2 and 10% in quadrature to find that the overall uncertainty in ^3He or nitrate flux associated with the dynamic solubility equilibrium is 10.2%.

The effect of measurement error of ^3He is a smaller uncertainty than the systematic uncertainties listed above but does lead to an error of 5% when propagated through all the calculations. Interestingly, for the 1985–1988 period, the absolute ^3He concentrations were much higher but the measurement uncertainty at that time was much worse, resulting in a similar 5% contribution of measurement uncertainty during that period as well.

Uncertainties in the slope of $\text{NO}_3 : ^3\text{He}$ feed directly into uncertainty in the nitrate flux, resulting in a 2.5% uncertainty in the nitrate flux. The uncertainties were derived from the uncertainty associated with the calculation of slope using a type II regression and appropriate measurement uncertainties for the individual data points. Additional error in the ^3He flux – and thus propagated to the nitrate flux – comes from the correction for tritium ingrowth in the water column. However, since the ^3He flux due to in situ tritium production is relatively small (12% of the total ^3He flux), the uncertainty on that number only contributes to a small fraction of the total uncertainty in the helium and nitrate fluxes (1%).

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In summary, we have used the approach of Jenkins and Doney (Jenkins and Doney, 2003) to calculate the *physical* supply of subsurface nitrate to the euphotic zone at BATS to be $0.65 \pm 0.3 \text{ mol m}^{-2} \text{ y}^{-1}$. This flux may support the new production of approximately $4.8 \pm 2 \text{ mol C m}^{-2} \text{ y}^{-1}$ due to the upward flux of nutrients over a broad region of the subtropical Northwestern Atlantic. We show that the rates are consistent with, but lower than, rates of new production recalculated from similar data from 1985–1988 and that this difference may be related to subtropical mode water formation. We also show that the rates estimated by this technique are higher than most other rates of new production estimated at the BATS site. This work thus shows that upward flux of nutrients – even if not directly observed at BATS by traditional techniques – is more than sufficient to support the observed rates of net community production and export production calculated at BATS.

Acknowledgements. We would like to thank Mike Lomas, Rod Johnson, and the BATS research team for the opportunity to collect samples. We are grateful for the assistance of the captain and crew of the R/V *Weatherbird II* and the R/V *Atlantic Explorer*. This research was funded by the National Science Foundation (OCE-1434000 and OCE-221247).

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Table 1. Fluxes calculated from the flux gauge technique for two different time periods. 1σ uncertainty estimates for each flux is given in parentheses underneath the reported value for each quantity.

Time Period	$\text{NO}_3 : ^3\text{He} \times 10^{-3}$ ($\mu\text{mol pmol}^{-1}$)	QuikSCAT Winds		NCEP Winds	
		^3He Flux ($\text{pmol m}^{-2} \text{y}^{-1}$)	NO_3 Flux ($\text{mol m}^{-2} \text{y}^{-1}$)	^3He Flux ($\text{pmol m}^{-2} \text{y}^{-1}$)	NO_3 Flux ($\text{mol m}^{-2} \text{y}^{-1}$)
2003–2006	82.9	7.9	0.66	8.3	0.69
1985–1988	(2.1)	(1.4)	(0.18)	(1.4)	(0.18)
	34.5	–	–	30.4	1.05
	(1.1)	–	–	(5.4)	(0.2)

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Table 2. The fractional uncertainty caused by different sources in the calculations of nitrate flux (numbers given are for 2003–2006 time period but results are similar if use 1985–1988 time period).

Source of Error	% uncertainty	Reference or method
Air–sea gas exchange	14 %	Stanley et al. (2009)
Dynamic solubility equilibrium	10 %	Dynamic solubility equilibrium calculated with range from Bourg and Sposito (2008) and range of gas exchange from Stanley et al. (2009)
Measurement error	5 %	Integration of error at each time point
$\text{NO}_3\text{:}^3\text{He}$ Slope	2.5 %	Type 2 regression
Tritium correction	1 %	Tritium measurement uncertainty propagated to ^3He flux

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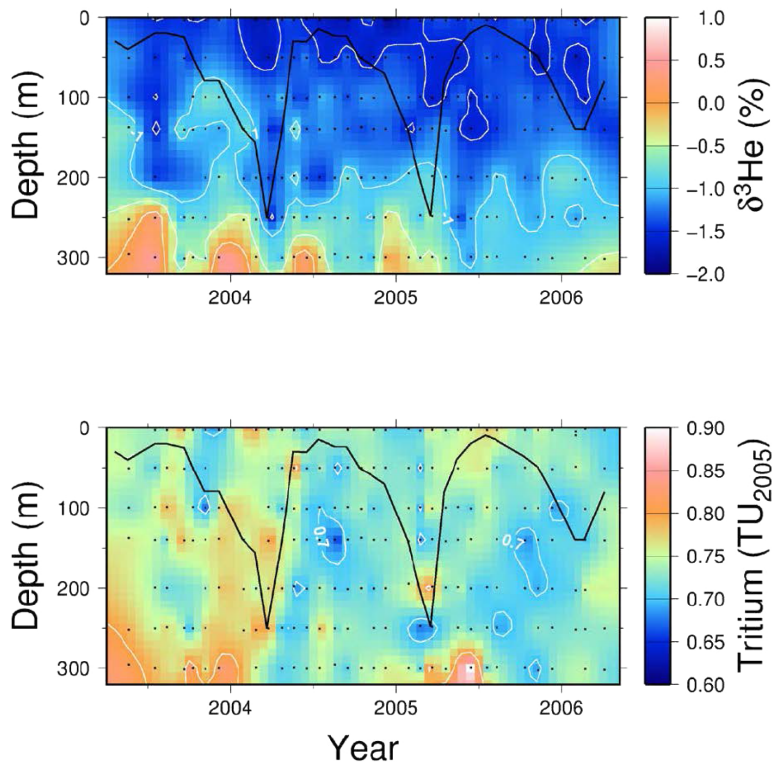



Figure 1. A time series of helium isotope ratio anomaly (in percent) relative to the atmospheric $^3\text{He}/^4\text{He}$ ratio (upper panel) and tritium (in Tritium Units) decay corrected to January 2005 (lower panel) at the Bermuda Atlantic Time Series Site in the North Atlantic near Bermuda. Sampling, designated by black dots, was approximately monthly over the ~ 3 year period. The black line is the mixed layer depth estimated from the CTD data.

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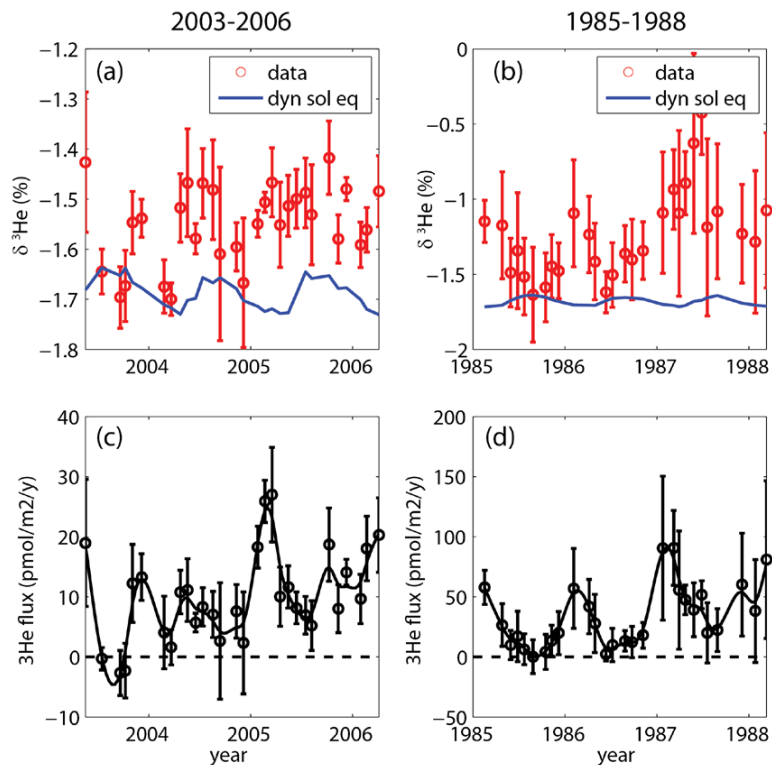


Figure 2. Mixed layer $\delta^3\text{He}$ data from (a) 2003–2006 and (b) 1985–1988 as well as the dynamic solubility equilibrium for d^3He . Error bars represent standard error of multiple measurements within the mixed layer. Fluxes of ^3He calculated from the data for (c) 2003–2006 and (d) 1985–1988. Note the difference in scales on the y axes for the two time periods.

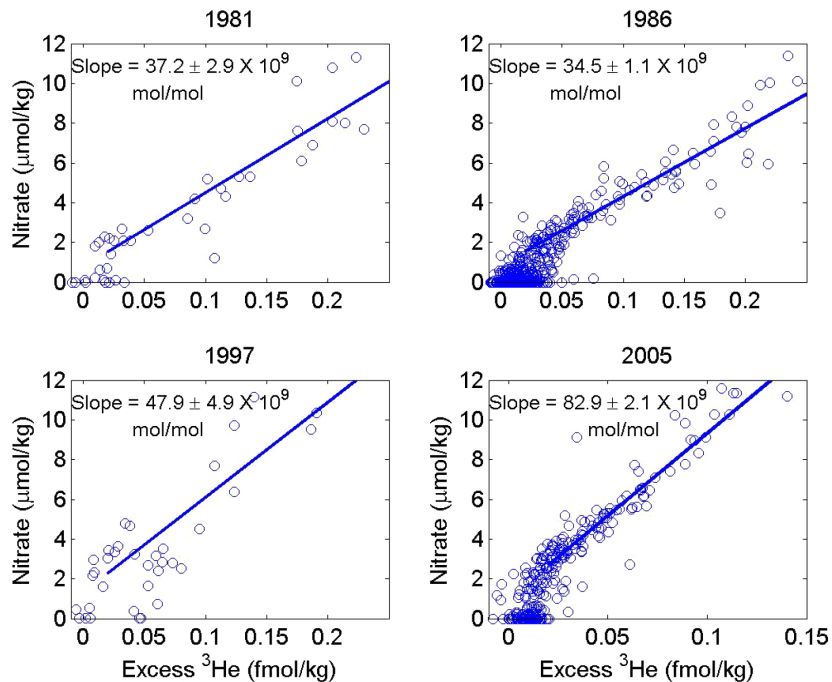


Figure 3. The observed relationship between excess (tritiogenic) ^3He and dissolved inorganic nitrate near Bermuda at four points in time. The 1986 and 2005 relations are based on approximately 3 year time series occupations near Bermuda (the former at Hydrostation S and the latter at BATS). The 1981 and 1997 data sets are from cruise stations within ~ 500 km of the site. Only samples with potential density anomalies less than 26.8 kg m^{-3} are plotted and used. Note the “waterfall” effect at low ^3He and nitrate concentrations in the euphotic zone, where the two tracers become uncoupled due to differing boundary conditions. The straight lines, from which the slopes are obtained, are type II linear regressions of points with nitrate concentrations in excess of $2 \mu\text{mol kg}^{-1}$. The lower bound nitrate limit was chose to avoid the tracer-decoupled points.

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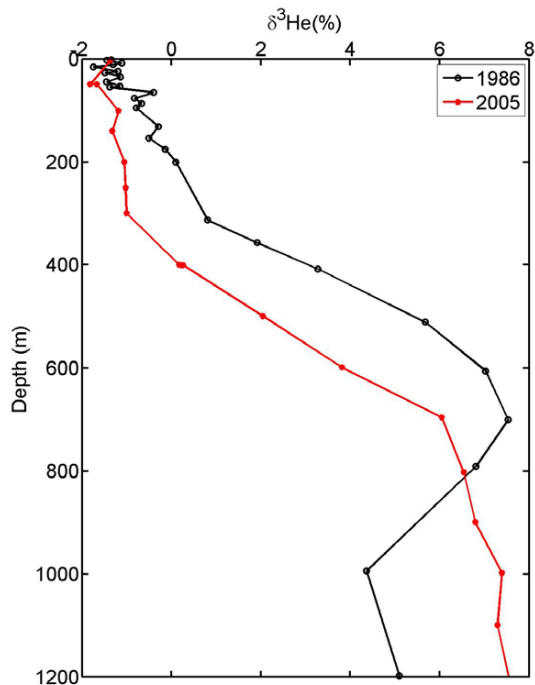


Figure 4. Representative profiles of $\delta^3\text{He}$ in the upper 1200 m of the water column in 1986 (black) and 2003–2006 (red). The profiles illustrate that in 1986 there was much higher $\delta^3\text{He}$ in the main thermocline and a larger gradient between the thermocline and the mixed layer than there was in 2006. This drives the observed greater ^3He flux in the 1980s compared to the 2000's.

The ^3He flux gauge in the Sargasso Sea

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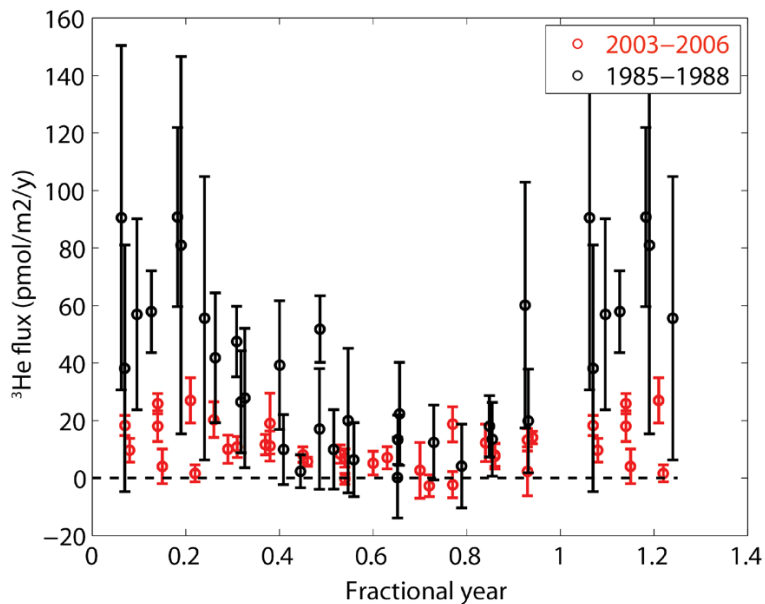


Figure 5. The ^3He flux as a function of fractional year for the 2003–2006 time period (red) and the 1985–1988 time period (black). The time period between 0 and 0.4 has been replicated from 1 to 1.4 in order to better visualize a seasonal cycle.

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