| 1 | Carbon sequestration in the deep Atlantic enhanced by Saharan dust |
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| 11 | Enhanced atmospheric input of dust-borne nutrients and minerals to the remote surface |
| 12 | ocean can potentially increase carbon uptake and sequestration at depth. Nutrients can |
| 13 | enhance primary productivity, and mineral particles act as ballast, increasing sinking |
| 14 | rates of particulate organic matter. Here we present a unique 2-year time-series of |
| 15 | sediment-trap observations of particulate organic carbon flux to 3000 m depth, measured |
| 16 | directly in two locations: the dust-rich central North Atlantic gyre and the dust-poor |
| 17 | South Atlantic gyre. We find that carbon fluxes are twice as high and a higher proportion |
| 18 | of primary production is exported to depth in the dust-rich North Atlantic gyre. Low |
| 19 | stable nitrogen isotope ratios suggest that high fluxes result from the stimulation of |
| 20 | nitrogen fixation and productivity following the deposition of dust-borne nutrients. |
| 21 | Sediment traps in the northern gyre also collected intact colonies of nitrogen-fixing |
| 22 | Trichodesmium species. Whereas ballast in the southern gyre is predominantly biogenic, |
| 23 | dust-derived mineral particles constitute the dominant ballast element during the |
| 24 | enhanced carbon fluxes in the northern gyre. We conclude that dust deposition increases |

carbon sequestration in the North Atlantic gyre through the fertilisation of the nitrogen fixing community in surface waters and mineral ballasting of sinking particles.

Flux of airborne desert dust into the surface ocean can increase the amount of 27 photosynthetically fixed carbon dioxide (CO₂) by reducing nutrient limitation of primary 28 production and thus increase the flux of particulate organic carbon (POC) to the deep ocean¹. 29 Dense dust-derived lithogenic particles can also increase particle size through aggregation and 30 enhance sinking velocity and preservation of POC through ballasting, allowing more carbon to 31 penetrate deeper into the ocean's interior². The impact of dust input on downward POC flux 32 can be especially important in the subtropical low-nutrient low-chlorophyll (oligotrophic) 33 gyres which occupy 60% of the global ocean surface³ and thus are likely large sinks for 34 atmospheric CO₂. Even relatively small changes in downward POC flux in these immense areas 35 would significantly affect the global carbon budget. However, the transport of organic carbon 36 (i.e. Biological Carbon Pump) in oligotrophic regions is very poorly understood, and large 37 uncertainties remain over the impact of enhanced dust deposition on the magnitude of POC 38 39 flux below the depth of winter mixing (sequestration).

40 We tested the hypothesis that enhanced dust deposition increases POC sequestration in remote low-nutrient low-chlorophyll provinces by directly measuring downward deep POC 41 flux in the centres of the subtropical North and South Atlantic gyres. The study regions 42 represent permanently stratified systems characterised by restricted nutrient advection, and 43 hence extremely low surface concentrations of macronutrients (nitrate and phosphate) and 44 chlorophyll. Here, picoplankton dominate community structure⁴, while heterotrophic bacteria 45 and cyanobacteria govern ecosystem metabolism, channelling a large proportion of POC into 46 the microbial loop⁵, thus diminishing its export out of the euphotic zone. The subtropical North 47 Atlantic, however, receives large depositional fluxes of Saharan dust with associated essential 48 nutrients (e.g. nitrogen, phosphorus, iron)¹ blocked from the South Atlantic region by the Inter-49

Tropical Convergence Zone⁶. The Fe-rich surface waters of the northern gyre are favoured by 50 N_2 fixing microbes (diazotrophs) that generate bioavailable nitrogen for other phytoplankton⁷. 51 thereby allowing for a higher proportion of primary production to be converted into sinking 52 POC than would otherwise occur. Phosphate co-limits the Fe-induced N₂ fixation and 53 production fuelled by atmospheric nitrogen^{8, 9}. This can exert an important control over the 54 amount of POC ultimately produced from diazotrophic and atmospheric sources and 55 subsequently available for export. Along with this fertilisation effect, increased lithogenic 56 particle concentration following dust input can also facilitate POC flux to depth through 57 additional incorporation of dense dust particles¹⁰. Biomineral ballasting is otherwise regulated 58 by calcite which is typically found in both gyres¹¹. However, the degree to which lithogenic 59 ballasting can drive the increased POC sedimentation would itself be limited by the amount of 60 POC present¹². 61

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Field observations in the central Atlantic gyres

We directly captured POC flux in the central Atlantic gyres from 2007 to 2010 using 63 sediment traps moored at 3000 m depth at sites NOG (23°N 41°W) and SOG (18°S 25°W) 64 (Fig.1). During this period, NOG was subjected to, on average, ten-fold higher dust deposition 65 compared to SOG (Fig. 2a), as inferred from dust concentration measurements over Barbados¹³ 66 for NOG and modelled data^{14, 15} for SOG (Methods). At both sites, the average surface 67 production rates derived from a Vertically Generalised Production Model (VGPM)¹⁶ were 68 lower than much of the global ocean¹⁷, and on average 23% higher at NOG than at SOG (Fig. 69 2b). The observed POC fluxes to the trap at NOG (0.40-2.7 mg C m⁻² d^{-1;} mean = 1.06 mg C m⁻² 70 2 d⁻¹) were always at least two-fold higher than at SOG (0.21-0.95 mg C m⁻² d⁻¹; mean = 0.49 71 mg C $m^{-2} d^{-1}$) (Fig. 2d, 3). The POC fluxes at NOG and SOG were significantly lower than the 72 depth-normalised values reported for the oligotrophic sites in the western North Atlantic gyre 73 (station OFP (BATS))¹⁸ and subtropical North Pacific gyre (station ALOHA)¹⁹, and hence they 74

75 are the lowest measured in the global ocean. From the ratios of POC flux to VGPM primary production (both variables were averaged over the trap deployment period) we calculate almost 76 double the fraction of surface production reaching 3000 m depth at NOG (0.60%) compared to 77 SOG (0.37%). These very low values are similar to the records at BATS $(0.59\%)^{18}$ and imply 78 an overall more efficient downward POC transport in the dusty northern gyre. Lithogenic flux 79 determined from aluminium concentrations in trap material was significantly lower at SOG 80 than at NOG (Fig. 2c) and elsewhere in the subtropical North Atlantic^{18, 20}, indicating that the 81 inter-basin differences in dust deposition propagated to depth. The NOG data bridge the 82 83 previous observations of deep lithogenic flux in the eastern and western parts of the northern gyre^{18, 20} showing the westward gradient of decreasing deep lithogenic fluxes driven by the 84 weakening of the Saharan dust transport towards the northwest-Atlantic¹³. 85

86 Although higher at NOG, at both sites, POC flux was enhanced during late summerautumn (>120% of the annual mean value; Fig. 3), a period of warm sea-surface temperature 87 (25.0 - 28.2 °C), relatively shallow mixed layer (<50 m), and low surface chlorophyll 88 concentrations (<0.04 mg m⁻³; Supplementary Fig. 1). At NOG, the average dust input during 89 summer-autumn (14.4 \pm 8.9 mg m⁻² d⁻¹) exceeded the wintertime values (8.87 \pm 11.6 mg m⁻² d⁻¹) 90 (Fig. 3a). An enhanced input of dust-borne nitrogen, phosphorus and iron has likely occurred 91 during this period. After nitrogen is exhausted by the dust-stimulated primary producers, the 92 93 warm and strongly stratified water column would offer optimal conditions for enhanced N₂ fixation provided there is enough iron and phosphate present to satisfy cellular demands of 94 diazotrophs^{7, 8, 9}. Bloom-forming *Trichodesmium* spp. dominate diazotrophic biomass in the 95 region of NOG^{21} . Elevated N₂ fixation rates by these diazotrophs were reported during 96 summer-autumn (median 34.9 µmol N m⁻² d⁻¹) compared to winter-spring (median 12.2 µmol 97 N m⁻² d⁻¹) (refs^{21, 22, 23, 24, 25}). This coincides with higher fluxes of aerosol iron in autumn than 98 in spring²⁶ and higher surface concentrations of dissolved iron in the early autumn²⁷ (1.0-1.3) 99

nmol L⁻¹) than in winter²⁸ (0.18-0.54 nmol L⁻¹). Lower phosphate concentrations measured in 100 the central northern gyre during summer have also been attributed to the enhanced diazotrophic 101 activity exhausting the phosphate pool⁸. Remarkably, we find a strikingly high POC flux of up 102 to 2.7 mg C m⁻² d⁻¹ in August-September 2009 at NOG (Fig. 3a). This relatively short POC 103 export pulse, never seen at SOG, accounted for 29% of total POC sequestered at NOG during 104 2007-2009 and greatly exceeded the mean wintertime POC flux at NOG (0.88 \pm 0.13 mg C m⁻² 105 d⁻¹) and the daily flux at SOG. A notable presence of some intact *Trichodesmium* "tufts" (Figs. 106 3a, 4) within this pulse suggests a potential involvement of these diazotrophs in driving the 107 108 extreme POC sequestration event at NOG. Similarly short and efficient POC export pulses to > 2800 m depth have been regularly observed at ALOHA following a summertime increase in 109 productivity and biomass of diatom-diazotroph symbiotic phytoplankton¹⁹. 110

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Fertilisation effect of dust

We measured markedly low stable nitrogen isotope ratios in the trap material ($\delta^{15}N_{PN}$, 112 in ‰ relative to air) from the dust-rich NOG (range 0.40-1.32‰; mass-weighed mean 0.77‰), 113 indicating that isotopically light nitrogen introduced by enhanced N₂ fixation and potentially 114 atmospheric deposition²⁹ significantly contributed to sinking particles. Some of this low $\delta^{15}N$ 115 signal might have originated south of NOG (10°-16°N), before being transported to and 116 accumulated at the NOG thermocline as low $\delta^{15}N_{\text{nitrate}}$ during northward water mass transit^{7, 30}. 117 However, a strong inverse correlation between $\delta^{15}N_{PN}$ and POC flux (R²=0.67, p=0.001) with 118 *Trichodesmium* "tufts" present at the lowest $\delta^{15}N_{PN}$ values (Figs. 4, 5), is suggestive of a direct 119 link between elevated POC flux at NOG and a local supply of newly fixed nitrogen by 120 diazotrophs whose activity was likely stimulated by substantial inputs of dust-borne iron and 121 phosphorus. Observations at NOG are qualitatively similar to those at ALOHA¹⁹, where $\delta^{15}N_{PN}$ 122 minima and diazotroph-driven particulate POC flux maxima are closely associated. Dust 123

124 deposition, which is a substantial source of isotopically light nitrogen in the region (8.5 μ mol 125 m⁻² d⁻¹; ref³¹) could augment the deep POC flux lowering its δ^{15} N signature.

126 In contrast to NOG, sinking particles from the dust-poor SOG carried significantly 127 heavier $\delta^{15}N_{PN}$ of 3.70% to 4.41% (mass-weighted mean 4.07%). This is similar to the oceanic 128 average $\delta^{15}N$ of deep-water nitrate (4.8%; ref²⁹), and hence this source was probably fuelling 129 primary production at SOG.

130 The deep $\delta^{15}N_{PN}$ at NOG and SOG fit a broad range of $\delta^{15}N$ values reported for 131 particulate nitrogen in the upper waters of the central North and South Atlantic gyres^{32, 33} 132 (Supplementary Fig. 2). At both sites, trap material was ¹⁵N-enriched compared to the particles 133 suspended in the euphotic zone (top 130 m) likely due to fractionation resulting from 134 remineralisation processes in both the surface and mesopelagic (Ref 34). Similar $\delta^{15}N$ values 135 for trap material and particles from 150-160 m depth may also point to a potentially important 136 contribution of heavier $\delta^{15}N$ signal formed at the deep chlorophyll maximum to $\delta^{15}N_{PN}$.

We estimated the contribution of different nitrogen sources to $\delta^{15}N_{PN}$ at NOG and SOG 137 using a two-end member nitrogen mass-balance model²⁹ (see Methods and references therein). 138 We assumed that the isotope budget of the mixed layer in the permanently oligotrophic gyres 139 incorporates nitrogen supplied by diazotrophs, by vertical diffusion across the nitrate 140 concentration gradient, and from dust (NOG only). We also assumed negligible isotopic 141 fractionation following complete nitrogen assimilation by phytoplankton. The average isotopic 142 signature of diazotrophic biomass ($-1\pm1\%$) was used as the N₂ fixation endmember. The upper 143 thermocline nitrate endmember was represented by δ^{15} N-nitrate averaged over the depth of the 144 nitrate gradient spanning the euphotic layer at NOG (2.73±0.36‰) and SOG (6.22±0.35‰). 145 The dust-derived nitrogen endmember was assigned $\delta^{15}N$ of -3.1% based on the average 146 isotopic composition of bulk aerosols influenced by Saharan dust. Using these endmember 147 values, we find that local N_2 fixation could contribute on average 50.4±8.4% to the isotopic 148

149 signal of nitrogen sequestration at NOG, while aerosol nitrogen alone (if all bioavailable) could account for 32.4±5.4% (Supplementary Table 1). The relative contribution of diazotrophs to 150 $\delta^{15}N_{PN}$ at NOG was higher than that at BATS (33%; at average $\delta^{15}N_{PN} = +1\%$ (ref³⁴) and nitrate 151 $\delta^{15}N=+2.6\%$ (ref³⁵)) and at ALOHA (range 21-48%; refs^{19, 36}), where eddy transfer and lateral 152 advection are important mechanisms of nitrogen supply^{36, 37}. At SOG, newly fixed nitrogen 153 contributed a smaller, yet considerable portion of $\delta^{15}N_{PN}$ (29.7±3.1%), possibly owing to the 154 activity of unicellular cyanobacteria, major N₂ fixers in the South Atlantic^{7, 21}. We, however, 155 acknowledge a significant uncertainty of these results due to an overall lack of time-resolved 156 δ^{15} N data for the surface nitrate and dust at the trap sites. Moreover, our budgets did not account 157 for a possible origin of particles from a specific trophic level (*e.g.* faecal pellets) and alteration 158 of $\delta^{15}N_{PN}$ due to isotopic fractionation during particle remineralisation and transformation in 159 the mesopelagic. However, regardless of these uncertainties, the isotope budgets suggest a large 160 systematic difference in the contribution of newly fixed local nitrogen inputs between the North 161 and South Atlantic gyres which likely contriutes to the two-fold inter-basin difference in POC 162 sequestration. Our observations thus set an important quantitative constraint on the downward 163 flux of low δ^{15} N material sinking to the subtropical North Atlantic. They provide compelling 164 evidence for the origin of an isotopically light nitrate reservoir in the subtropical North Atlantic 165 supporting previous observations (e.g. ref^{30}). 166

167 The unique presence of intact *Trichodesmium* colonies in the deep particles at NOG 168 (Fig. 4) indicates that *Trichodesmium* biomass is not always lost in the surface waters as 169 previously assumed^{38, 39}, but can leave the euphotic zone and contribute to POC export. It is 170 possible that the "tufts" reached the abyssal depth at NOG in a rapidly sinking (>200 m d⁻¹) 171 *Trichodesmium* bloom, collapsed through viral lysis or programmed cell death³⁹. Since Fe 172 starvation at NOG is unlikely, exhaustion of bioavailable phosphrous⁸ during the summer 173 might be major triggers of the bloom collapse. Alternatively, the "tufts" might represent 174 *Trichodesmium* populations that migrated towards the phosphocline to "mine" phosphate but 175 were unable to return to the light⁴⁰. Finally, *Trichodesmium* can retain dust particles within 176 their morphologically intricate colonies to accelerate Fe dissolution from dust⁴¹. Trapped dust 177 particles may therefore "ballast" *Trichodesmium* colonies, increasing their density and 178 allowing them to sink rapidly to depth and avoid remineralisation or grazing. This could partly 179 explain the temporal coherence between low δ^{15} N, elevated dust, POC, and lithogenic fluxes 180 during late summer at NOG (Fig. 3a).

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Ballasting effect of dust

Higher dust input significantly altered the composition of particles at NOG compared 182 to SOG (Fig. 2e). Dust-derived lithogenic material was the second largest contributor 183 184 (34.3±11.6%) to the total mass at NOG after calcite, whereas at SOG this value was 4.7±2.3%, 185 consistent with the difference in the amount of dust being deposited at each site (Fig. 2a). Although the seasonal signal of elevated dust flux at both sites was largely lost at 3000 m depth, 186 we still observed elevated lithogenic flux at NOG (>120% of the annual average) in winter 187 2008 and summer-autumn 2008 and 2009 concurrently with the increased POC flux and 188 following high dust input (Fig. 3a). Assuming that this temporal coherence was not accidental, 189 we investigated the relative involvement of lithogenic and biogenic (opal + calcite) ballast 190 phases in enhanced POC sequestration. Based on the outputs of the mineral-associated POC 191 flux model and multiple linear regression analysis^{2, 42} (Methods), 41.0% of POC flux at SOG 192 was ballasted by lithogenic material. This, however, might be an overestimation driven by a 193 relatively large carrying coefficient for lithogenic ballast (0.371) which resulted from a nearly 194 1:1 ratio of POC to lithogenic flux and their strong positive correlation (Spearman's p=0.91). 195 At NOG the percentage of POC ballasted by lithogenic particles increased from 45.7% during 196 low POC flux to 70.1% during high flux in the summer-autumn (Supplementary Table 2). 197 Overall, lithogenic material appears to be a more important ballast for POC in the central 198

199 northern gyre compared to its western boundary (25%), where lithogenic fluxes are lower and opal fluxes are ten times higher¹⁸. We suggest that at NOG elevated dust inputs may shift the 200 dominant ballasting phase from biogenic to lithogenic, increasing POC flux to the deep ocean. 201 202 This is likely achieved through a sudden increase in mineral particle concentration following dust deposition and subsequent stimulation of aggregation of organic matter, including that of 203 diazotrophs, in the surface waters¹⁰. Moreover, clay particles, constituting >60% of the aerosol 204 dust over the central North Atlantic⁴³, are denser (2.79 g cm⁻³) than biomineral calcite (2.65 g 205 cm⁻³) and opal (2.1 g cm⁻³), and thus would likely increase sinking velocity of POC upon 206 aggregation. Although currently debated in the literature (e.g. refs.^{44, 45}), lithogenic ballast 207 might have also exert an enhanced protective effect on POC compared to calcite. Recent 208 laboratory experiments^{45, 46} demonstrated slower degradation rates for clay-ballasted POC 209 relative to calcite-ballasted POC. The existence of such protective effect of lithogenic material 210 is yet to be shown in the field. 211

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Mechanism of dust-induced enhancement of carbon sequestration

Lithogenic particles did not represent the main ballasting phase for POC during periods 213 of high and low lithogenic fluxes and were not associated with the biomineral fluxes at NOG 214 (Supplementary Table 2). The ballasting ability of lithogenic particles at NOG appears to be 215 confined to the summer-autumn period (Fig. 3a) when the surface fertilisation by dust was 216 potentially the strongest. This tight temporal coupling suggests that the presence of additional 217 218 fresh organic (i.e. fertilisation effect) matter might be required to activate effective lithogenic ballasting while lithogenic particles are critical to transport the fertilisation effect to the deep 219 ocean. The variability in mineralogy and morphology of dust arriving at NOG from different 220 locations in the Sahara during winter⁴⁷ and summer may have also impacted both fertilisation 221 and ballasting properties of dust. 222

223 Overall, enhanced POC sequestration in the dust-rich NOG suggests that in the vast nutrient-limited Atlantic, the strength of the biological carbon pump could be significantly 224 lower without concurrent dust-induced fertilisation and ballasting. The observed two-fold 225 226 enhancement of POC sequestration under a ten-fold higher dust (iron) input at NOG further points to a potentially important role of phosphate in setting the upper bound for the Fe-driven 227 enhancement of POC export. However, fertilisation could also stimulate the activity of 228 heterotrophic bacteria, increasing remineralisation and a corresponding reduction of carbon 229 export⁵. 230

231 Under the current climatic trends, the subtropical oligotrophic gyres are predicted to expand over the coming centuries⁴⁸. Multi-decadal observations of dust concentrations over 232 Barbados have already revealed a weakening of dust transport from North Africa to the North 233 Atlantic as a function of increasing sea-surface temperature¹³. Predicted changes in wind 234 patterns are expected to continue altering dust deposition into the ocean and hence input of 235 nutrients and mineral ballast⁴⁹. In parallel, ongoing ocean acidification might affect 236 bioavailability of essential nutrients, including iron⁵⁰. All these perturbations will certainly 237 alter POC sequestration in the oligotrophic gyres, and hence global climate, in the coming 238 centuries. Therefore, our study urges for a better understanding of the present Biological 239 Carbon Pump functioning in the nutrient-limited oceans. 240

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434

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460 **Figure captions**

Figure 1| Chlorophyll and dust deposition flux in the Atlantic Ocean. a, annual composite 461 Moderate-resolution Imaging Spectroradiometer chlorophyll-*a* concentration (mg m⁻³) in 2009. 462 Oligotrophic gyres are represented by dark blue areas of low chlorophyll concentrations (<0.1 463 mg m⁻³). **b**, basin-wide annually averaged (1974-2004) modelled dust deposition flux re-plotted 464 from ref¹⁴. Yellow triangles indicate the locations of the NOG and SOG sediment trap 465 moorings, which are also on the annually repeated Atlantic Meridional Transect (AMT) line 466 467 (www.amt-uk.org). The black solid line shows the AMT-19 cruise track (Oct-Nov 2009) passing through the NOG and SOG sites. Dashed lines indicate an approximate north-south 468 boundary of the Inter-Tropical Convergence Zone. 469

Figure 2| Surface ocean and deep particle flux data for the study sites. a-d, mean ± standard
deviation values over the respective trap deployment periods. a, dust deposition flux (n=25 for
NOG and n=26 for SOG). b, depth-integrated primary production derived from the
chlorophyll-based Vertically Generalised Production Model¹⁶ (Methods) c, lithogenic flux
(n=40 for both sites). d, POC flux (n=40 for both sites). e, composition of sediment trap
material. The height of the stacked bars represents total particle mass flux.

Figure 3 Time-series fluxes at NOG (a) and SOG (b). The dust deposition (monthly values) and aluminium-derived lithogenic fluxes are presented on a logarithmic scale. The uncertainty of the dust flux to the South Atlantic is estimated to be at least a factor of 10 (ref¹⁴). For POC and lithogenic fluxes, the width of each bar corresponds to 14- or 21-day collection interval. Red circles depict stable nitrogen isotopic composition of particles ($\delta^{15}N_{PN}$) from the selected cups. Arrows and a letter "T" indicate the cups where *Trichodesmium* spp. "tufts" were found. Summer-autumn periods are highlighted in yellow.

Figure 4| *Trichodesmium* spp. "tufts" from the summer POC flux pulse at NOG. Tufted
colonies of *Trichodesmium* spp. cells were identified in the cups collecting in August and
September 2009. This is the first record of *Trichodesmium* being exported to bathypelagic
depth (>1500 m).

Figure 5| POC flux *vs.* isotopic composition of the trap material ($\delta^{15}N_{PN}$) from NOG (blue circles) and SOG (red circles). The black line is the best fit line of the linear model. Arrows with letter "T" mark the cups where *Trichodesmium* tufts were found. The strong inverse relationship between the magnitude of POC flux and $\delta^{15}N_{PN}$ at NOG signifies a potentially important role of local input of isotopically light N from N₂ fixation (and dust deposition) in enhancing carbon sequestration at this site.

495 Methods

Particle collection and processing. Sinking particles were collected using 21-cup time-series 496 Parflux Mark 78H-21 sediment traps (McLane Research Laboratories, USA) deployed on a 497 bottom-tethered mooring at a depth of 3000 m in a water depth of >4200 m. At NOG, the traps 498 were deployed from November 4, 2007 to October 5, 2008 and from November 23, 2008 to 499 October 25, 2010, collecting particles over a total of 672 days. At SOG, the traps operated from 500 May 11, 2008 to May 20, 2009 and from May 24, 2009 to June 20, 2010, collecting particles 501 over a total of 766 days. Each trap cup collected for 14 or 21 days. Sample preservative 502 consisted of a solution of sodium chloride (5 g L^{-1}), di-sodium tetra-borate (0.25 g L^{-1}), and 503 formalin (5% vol/vol) made up with deep seawater. Upon recovery, pH was measured and 504 found to be between 8.0 and 8.3. One mL of concentrated formalin solution was then added to 505 the cups to supplement the existing formalin. Sample processing was carried out under dust-506 507 and metal-free conditions in a laminar flow cabinet using plastic- or glass-ware only. Prior to all analyses, zooplankton "swimmers" were identified under stereo-microscope (Meiji Techno, 508 509 Japan) fitted with a photo-camera (Canon EOS-1000, Japan) and handpicked using PTFE-510 coated tweezers (Dumont, Switzerland) and a plastic pipette (Fisher Scientific, UK). The preservative/particle mixture in each cup was then split into 8 sub-samples using a custom-511 built rotary PVC splitter. Individual sub-samples from each cup were filtered, dried at 40°C 512 and analysed for particulate organic carbon, opal, calcite, and trace metals including 513 aluminium. Selected sub-samples were also analysed for stable nitrogen isotope composition. 514

515 **Chemical analyses of the trap material.** Particulate organic carbon (POC) was measured in 516 tin capsules (HEKAtech GmbH) after removing carbonate by *in situ* acidification⁵¹ with 517 concentrated hydrochloric acid and using a high-temperature combustion technique on a CHN 518 analyser (HEKAtech GmbH EURO EA CHNS-O Elemental Analyser) with analytical 519 precision of <0.1%). The median filter blank contribution to POC signal was 2.7%. The

520 calculated limit of detection (LoD; based on three times standard deviations of the filter blanks) was 8.26 µg (n=20). Particulate Organic Matter (POM) was calculated as 2.2×POC (ref²). Splits 521 for calcite were prepared by leaching in 0.4 mol L⁻¹ nitric acid with calcium content measured 522 by inductively coupled plasma optical emission spectrometry⁵² (Perkin-Elmer Optima 4300DV 523 ICP-OES; analytical precision of <1%). Procedural blanks consisting of unused polycarbonate 524 membranes treated with nitric acid contributed <1% to Ca signal. The LoD of the blank-525 corrected Ca measurements was wavelength-dependent, ranging from 0.012 to 0.015 µg 526 (n=10). Calcite mass flux was calculated by multiplying calcium-derived flux of particulate 527 inorganic carbon by a factor of 8.3. Samples for opal were digested in 0.2 mol L⁻¹ sodium 528 hydroxide, neutralized with 0.1 mol L⁻¹ hydrochloric acid and analysed as dissolved silicate on 529 a SEAL QuAATro auto-analyser^{52, 53}. The detection limit of the instrument was 0.3 µg. The 530 median contribution of procedural blanks was 3.1%. The LoD of the filter-blank corrected 531 samples was run-dependent ranging from 1.19 to 11.5 µg (n=9). Opal was calculated to be 2.4 532 \times biogenic silica flux assuming 10% water content^{52, 53}. Labile and refractory fractions of 533 aluminium in trap material were determined⁵⁴. The labile fraction was extracted with 25% 534 (vol/vol) acetic acid at room temperature, and then the more refractory fraction was fully 535 digested in a mixture of concentrated nitric and hydrofluoric acids at 150°C. The residues of 536 both fractions were redissolved in 0.5 mol L^{-1} nitric acid and analysed by inductively coupled 537 plasma-mass spectrometry (Thermo Fisher Scientific Element 2 XR HR-ICPMS). The LoD of 538 blank corrected aluminium measurements was 0.12 ng g^{-1} (n=10); the concentrations in acid 539 mix and blank filters were 0.764 ± 0.8 ng g⁻¹; (n=10) and 0.843 ± 0.917 ng g⁻¹ (n=8), respectively. 540 The accuracy of the measurements was established using a range of Certified Reference 541 Materials, including HISS-1, NIST-1648a and NIST-1573a. The recoveries in these reference 542 materials were 97.3-104.1% for aluminium. Total trace metal concentration was determined by 543 adding leach and digest metal fractions. Total aluminium mass flux was used to calculate 544

lithogenic mass flux based on aluminium content of 7.1% in Saharan dust⁵⁵ and 7.7% in 545 Patagonian dust⁵⁶ for NOG and SOG samples respectively. Stable nitrogen isotopic 546 composition of sinking particulate nitrogen pool ($\delta^{15}N_{PN}$) was determined from ${}^{14}N/{}^{15}N$ mass 547 ratio measured using Micro Cube elemental analyser (Elementar Analysensysteme GmbH, 548 Hanau, Germany) interfaced to a PDZ Europa 20-20 isotope ratio mass spectrometer (Sercon 549 Ltd., Cheshire, UK). The accuracy of the measurements was established using a set of 550 laboratory standards calibrated against NIST Standard Reference Materials (IAEA-N1, IAEA-551 N2, IAEA-N3, USGS-40, and USGS-41). The analytical precision of the δ^{15} N_{PN} measurements 552 553 was <0.1‰, while the difference between duplicates ranged between 3.1 and 11% (n=4). Measurements were performed at the UC Davies Stable Isotope Facility, USA. 554

555

Dust deposition flux. Direct and time-resolved measurements of dust deposition at NOG and 556 557 SOG are not available. At SOG we obtained monthly estimates of dust deposition using an atmospheric model^{14, 15}, which utilizes reanalysis data (a combination of model and 558 559 observations) to drive a dust chemical transport model, and was compared to long-term 560 measurements of aerosol concentration. Dust deposition flux was modelled in four bins with the size distribution range of 0.1-0.5, 0.5-1.0, 1.0-2.5, and 2.5-10 µm. Dust deposition velocities 561 were calculated within the model as a function of meteorological conditions and resulted in 562 averages of 0.01, 0.029, 0.115, and 0.674 cm s⁻¹ over our region. The modelled dust deposition 563 fluxes were averaged for $3^{\circ} \times 3^{\circ}$ area centred at the SOG location. The uncertainty of the model 564 output for the South Atlantic Ocean is hypothesised to be at least a factor of 10 due to scarcity 565 and uncertainties in observational data and uncertainties in model source, transport and 566 deposition processes¹⁴. Dust deposition flux at NOG was inferred from time-series dust 567 concentrations measured over Barbados which is heavily influenced by air-masses from Sahara 568 and Sahel deserts¹³. The details of dust sampling and processing are described in ref¹³. Dust 569

⁵⁷⁰ deposition flux was calculated by multiplying dust concentrations by a range of deposition ⁵⁷¹ velocities (0.01-1.2 cm s⁻¹) characteristic of relatively fine mineral dust aerosols of $<5 \mu$ m in ⁵⁷² size typically arriving to the remote open ocean⁵⁷. The resulting average dust deposition flux ⁵⁷³ at NOG ranged from 0.085 to 10.2 mg m⁻² s⁻¹. Assuming a deposition velocity of 1 cm s⁻¹, dust ⁵⁷⁴ deposition flux is similar in magnitude to deep lithogenic flux at NOG. Thus, we considered ⁵⁷⁵ this deposition velocity to be the most appropriate for calculations of daily dust deposition flux ⁵⁷⁶ at NOG.

577

Upper ocean hydrography. Eight-day composite sea-surface temperature (SST) data were recorded by the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor of NASA's Aqua satellite at 9 km resolution and averaged for $3^{\circ} \times 3^{\circ}$ box centred at each trap location. The annual cycle of mixed layer depth at the trap sites was derived from the ARGO-based climatology⁵⁸ averaged for $3^{\circ} \times 3^{\circ}$ area over the trap sites. The base of the mixed layer was defined as the depth at which the density was 0.03 kg m⁻³ less than that at 10 m.

584

Ancillary biogeochemical datasets were provided by the British Oceanographic Data Centre (BODC) and include vertical profiles of chlorophyll (archived data under accession numbers SOC050136 and SOC110235), nitrate concentrations (refs^{59, 60} and archived dataset under accession number MIT130172), isotopic composition of total nitrate (refs^{61, 62, 63}), nitrogen fixation rates (refs^{22, 23, 24, 25}), ¹⁴C-based primary production rates (ref⁶⁴ and archived data with accession numbers PP-PML090162, PP-PML110236 and PP-PML120146),

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Primary production. Depth-integrated daily rates of primary production for the relevant time period were estimated from the chlorophyll-based eight-day resolved Vertically Generalized Production Model (VGPM)¹⁶ and averaged for the $3^{\circ} \times 3^{\circ}$ area centred at the trap sites. The 595 VGPM data downloaded from the Ocean Productivity website were (http://www.science.oregonstate.edu/ocean.productivity/). Within relevant time periods, the 596 VGPM-based productivity rates at NOG (160±14 mg C m⁻² d⁻¹) and SOG (139±18 mg C m⁻² 597 d⁻¹) were comparable to the values measured directly at the trap sites in October-November 598 2008-2011 (240 \pm 96 mg C m⁻² d⁻¹ at NOG and 204 \pm 84 mg C m⁻² d⁻¹ at SOG (see ancillary 599 biogeochemical datasets above). 600

601

602 **Surface chlorophyll-***a* **concentration.** Eight-day composite surface chlorophyll-*a* data were 603 recorded by MODIS Aqua at 9 km resolution and averaged for $3^{\circ} \times 3^{\circ}$ box centred at each trap 604 location. MODIS Aqua calculates near-surface chlorophyll concentrations from a model of 605 ocean colour using an empirical relationship.

606

Contribution of newly fixed nitrogen to the stable nitrogen isotope signal in trap material. 607 The δ^{15} N of the trap material reflects both the autotrophic particle formation and the subsequent 608 heterotrophic transformations. In the latter, the diagenetic fractionation can potentially alter 609 δ^{15} N of the bulk nitrogen export and sequestration. No significant relationship was observed 610 between C/N ratios and δ^{15} N of nitrogen export at NOG (r²=0.02, n=12) while at SOG, this 611 relationship was positive but weak and insignificant ($r^2=0.25$, n=12). This suggests that the 612 observed variations in δ^{15} N of the trap material were determined predominantly during algal 613 production, with no significant influence from detrital material and/or non-phytoplankton 614 organisms⁶⁵. At both sites isotopic fractionation following nitrogen assimilation is expected to 615 be negligible due to constant nitrogen limitation in the surface waters²⁹. Therefore, $\delta^{15}N$ of the 616 produced organic matter should reflect the composition of dominating nitrogen sources to the 617 euphotic zone, namely, upward diffusive flux of deep-water nitrate and N₂ fixation both having 618 distinct isotopic signals. In addition, in the northern gyre, atmospheric dust deposition can 619

significantly contribute to the total pool of new nitrogen^{31, 66}. Using equation (1) we describe
isotopic composition of nitrogen export as mixing between diffused nitrogen from the upper
thermocline and nitrogen from external source, represented by either diazotrophy or
atmospheric deposition at NOG, and diazotrophy only at SOG:

$$\delta^{15} N_{PN} = (f_1 \times \delta^{15} N_{f1}) + (f_2 \times \delta^{15} N_{f2})$$
(1)

where f_1 and f_2 and $\delta^{15}N$ denote fractions and isotopic signatures of dominant nitrogen sources. 624 We estimate the percent contribution of these sources from a single choice of their respective 625 endmember δ^{15} N values: +2.73‰ (NOG) and +6.22‰ (SOG) for nitrate diffusing from the 626 shallow thermocline across the concentration gradient; -1.0‰ for N₂ fixation (both sites), -627 3.1‰ for bulk aerosol input (NOG only). Due to sensitivity of the two-endmember mixing 628 model to the values of the chosen endmembers, we performed sensitivity analyses to account 629 for uncertainty of the changing δ^{15} N endmembers on the fraction of δ^{15} N_{PN} (in %) originating 630 from this source at each site, similar to isotopic assessment in ref²⁸. The choices of $\delta^{15}N$ 631 endmembers for each nitrogen source and those used in the sensitivity tests are described in 632 the section below and the results are summarised in Supplementary Table 1. 633

634 Sensitivity analyses and δ^{15} N endmember choice.

635 **Nitrate endmember:** The choice of nitrate δ^{15} N endmember was based on the biogeochemical 636 data (nitrate δ^{15} N, nitrate and chlorophyll concentrations, PAR) obtained at the NOG and SOG 637 sites during AMT cruises in May-June 2005 and October 2005, and US-GEOTRACES cruise 638 GA03 in December 2011 (see ancillary biogeochemical datasets above).

At the permanently oligotrophic NOG and SOG sites, winter mixing is weak, and thermocline nitrate is supplied into the euphotic zone largely by turbulence-driven upward diffusion⁶⁷. The magnitude of diffusive nitrate flux is governed by nitrate concentration gradients as the changes in turbulent diffusivity are relatively small⁶⁷. At both sites, nitrate concentrations remain at 643 nanomolar levels (< 0.01 μ mol L⁻¹) throughout the top 130-150 m and increase below, 644 signifying the position of the nitracline (defined by a nitrate concentration of 0.1 μ mol L⁻¹ (*e.g.* 645 ref⁶⁸). The largest nitrate flux with a characteristic δ^{15} N signature is therefore expected at the 646 depth of the maximum nitrate concentration gradient typically found at depths near the base of 647 the euphotic zone (0.1 % surface PAR; includes the deep chlorophyll maximum).

Referring to vertical profiles of nitrate and chlorophyll concentrations, we calculate 648 concentration-weighted average nitrate δ^{15} N (ref³³) from the top of the nitracline, where nitrate 649 concentrations begin to consistently increase, to the base of the euphotic zone. At NOG this 650 yields nitrate δ^{15} N of +2.73±0.36 ‰ (n=5) for the depth range of 137-191 m. In our isotopic 651 budgets this value represents an isotopic signal of nitrogen pool influenced by N₂ fixation and 652 atmospheric deposition, and sustained over time in the shallow thermocline. This is achieved 653 through both the internal cycle of low- $\delta^{15}N$ nitrate assimilation and subsequent 654 remineralisation and 2) accumulation of low- δ^{15} N nitrate imported during the northward water 655 mass transit^{7, 30}. This nitrogen pool has not yet been homogenised with the large global ocean 656 nitrate reservoir (~4.8‰) or ¹⁵N-enriched through denitrification^{29, 30}. At SOG the most 657 relevant depth range for measured nitrate δ^{15} N spanned 226-230 m, substantially deeper than 658 the base of the euphotic zone. The corresponding mean $\delta^{15}N$ of +6.22±0.35‰ may thus 659 overestimate the value for the shallower waters, where preferential remineralisation of ¹⁴N may 660 introduce a ¹⁵N-depleted signal to the nitrogen pool²⁹. 661

For the primary sensitivity test (Supplementary Table 1) we used the minimal nitrate $\delta^{15}N$ observed in the upper thermocline at NOG (+0.96‰ at 137 m depth) to estimate the least contribution of local N₂ fixation to $\delta^{15}N_{PN}$. We also tested nitrate $\delta^{15}N$ averaged from the top of nitracline down to 26.8 kg m⁻³ isopycnal surface, which marks the main thermocline depth at the study sites^{7, 69}. The corresponding value at NOG was +3.53±0.40‰ (n=13) for the 136667 421 m depth range; the SOG value was +6.35±0.32 ‰ (n=3) for 226-306 m depth range. 668 Finally, we included the oceanic global mean $\delta^{15}N$ (+4.8‰; ref²⁹) to compare our isotope 669 budgets with published data.

Nitrogen fixation endmember: We chose the mean $\delta^{15}N$ for diazotrophic biomass (-1±1‰) to represent the N₂ fixation endmember at both sites ^{29, 70, 71}. Assuming the mean nitrate $\delta^{15}N$ signal in the shallow thermocline, the range of the isotopic signal for diazotrophic nitrogen (-2‰ to 0‰) generates average contributions of 39.8-68.5% and 26.1-34.4% to $\delta^{15}N_{PN}$ at NOG and SOG, respectively (Supplementary Table 1).

Atmospheric deposition endmember: Atmospheric fluxes supply approximately $\sim 9.9 \times 10^9$ 675 mol N yr⁻¹ to the central North Atlantic gyre and 5.8×10^9 mol N yr⁻¹ to the South Atlantic 676 gyre⁷². Although these values are notably smaller than regional estimates of new nitrogen 677 inputs from diazotrophy (20×10¹¹ mol N yr⁻¹; ref⁷³), recent studies^{30, 31, 35} suggest that 678 deposition fluxes can significantly lower the $\delta^{15}N$ of the nitrogen pool. The published data on 679 δ^{15} N in atmospheric fluxes in the open Atlantic Ocean is extremely scarce. Previous studies^{31,} 680 ^{35, 74, 75, 76} report a wide range of δ^{15} N in bulk aerosol and rainfall samples (-6.8‰ to +1.7‰). 681 Given that dry deposition dominates atmospheric input at NOG, a value close to an average 682 683 isotopic signal of bulk aerosols seems the most appropriate to represent the dust endmember at NOG. We thus choose $\delta^{15}N$ of -3.1‰, based on the mean $\delta^{15}N$ values measured in the Sahara-684 influenced aerosol samples collected the sub-tropical North Atlantic^{31, 74} and Crete⁷⁶. For the 685 sensitivity test, we varied δ^{15} N of aerosol N across the full range, also including annual (-4.5%) 686 and seasonal cold (-6.8%; October-March) and warm (-1.9%; April-September) averages 687 measured in the Bermuda rainfall^{35, 74, 75} (Supplementary Table 1). We find that with the nitrate 688 $\delta^{15}N$ of 2.73‰, aerosol nitrogen can account for a sizable fraction of $\delta^{15}N$ of nitrogen export 689 at NOG (21.7-176%). Therefore, with nitrogen input equal or greater to magnitude of N₂ 690 691 fixation, dust deposition can have a similar or greater effect on the isotopic budget of trap 692 material from NOG. Hence, future studies should include the measurements of both magnitude 693 and δ^{15} N of dust deposition and N₂ fixation to avoid under- or over-estimation of the 694 importance of each of source.

Assessment of ballast effect of lithogenic flux. We examined the relationship between POC and (bio)mineral at NOG and SOG using POC flux model by ref^2 . The model divides POC flux into fractions ballasted by biomineral (opal + calcite; POC_{bio}) and lithogenic (POC_{lith}) particles, and freely sinking POC (POC_{free}). We use multiple linear regression to fit the particle flux data into equation (2) and determine correlation coefficients *a*, *b* and *c* (hereafter, carrying coefficients) for each fraction, following approach in refs^{2, 42, 77}.

$$POC flux = a \times POC_{bio} + b \times POC_{lith} + c \times POC_{free}$$
(2)

Carrying coefficients only reflect the size of the ballast-normalized fraction of POC flux, but 701 not their absolute magnitudes, and are used to calculate the relative fraction (in %) of POC 702 associated with each ballast type^{42, 77}. We further assume that the POC_{free} fraction is negligible 703 at 3000 m depth and force multiple linear regression to pass through zero². The strong temporal 704 705 variability of dust deposition limits the relevance of the annual-scale approach for estimating the role of lithogenic ballast to POC flux. Hence, we first assess the effect of lithogenic ballast 706 based on different POC sequestration scenarios, namely, (1) elevated POC flux (≥120% of 707 annual mean) at NOG, (2) POC flux at NOG outside scenario (1), (3) POC flux at SOG. We 708 evaluated the sensitivity of these results by performing multiple linear regression on the NOG 709 flux dataset separated according to the high and low lithogenic fluxes (Supplementary Table 710 2). Our approach differs from that applied previously by refs^{2, 42, 77} in which carrying 711 coefficients for both calcite and opal were determined. This is due to strong collinearity 712 observed between calcite and opal in all POC-based groups, violating the independence 713 assumption of multiple linear regression, as further determined by ridge regression analysis. 714 The resulting carrying coefficients and calculated proportion of ballast-associated POC flux in 715

716 each surveyed group are summarised in Supplementary Table 2. The carrying coefficients for lithogenic material compared well with the global and the north Atlantic means (0.052, and 717 $(0.058, respectively)^2$ during low POC flux, but exceeded these values during high fluxes and 718 overall at SOG. We acknowledge that the relatively large carrying coefficient for lithogenic 719 ballast in the SOG group compared to the NOG groups and other time-series might be an 720 overestimation introduced by a nearly 1:1 ratio between POC and lithogenic fluxes and a their 721 strong positive correlation (Spearman's p = 0.91). As a result, the proportion of POC flux 722 ballasted by lithogenic material appears to be comparable between SOG and scenario (2) at 723 NOG, despite the significant difference in their lithogenic fluxes (Supplementary Table 2). 724

Data availability: The data analysed during this study are available from the corresponding
author upon reasonable request. The supporting data for this study are available from the
repository of the British Oceanographic Data Centre upon request.

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FIGURES

842 Figure 1











851 Figure 4





858 SUPPLEMENTARY INFORMATION

859 Supplementary Table 1| Sensitivity analysis of the choice endmember $\delta^{15}N$ value on 860 calculated source contribution (%) to isotopic signal of nitrogen flux at 3000 m depth

| Nitrate endmember | N ₂ fixation (%) | | | Dry deposition (%) | | | Rainfall (%) | | |
|-----------------------|-----------------------------|--------|------|--------------------|--------|-------|--------------|--------|------|
| δ ¹⁵ N (‰) | Min | Choice | Max | Min | Choice | Max | Min | Choice | Max |
| NOG | -2 | -1 | 0 | -6 | -3.1 | +1.7 | -6.8 | -4.5 | -1.9 |
| +0.96 (min NOG) | 2.82 | 4.25 | 8.68 | 1.20 | 2.1 | -11.3 | 1.07 | 1.53 | 2.91 |
| +2.73 (137- 191 m) | 39.8 | 50.4 | 68.5 | 21.7 | 32.4 | 176.2 | 19.8 | 26.2 | 40.7 |
| +3.53 (137-421 m) | 48.0 | 58.6 | 75.2 | 27.4 | 40.0 | 145.0 | 25.7 | 33.0 | 48.9 |
| +4.8* | 57.7 | 67.7 | 81.7 | 36.3 | 49.7 | 126.6 | 33.8 | 42.2 | 58.6 |
| SOG | -2 | -1 | 0 | | | | | | |
| +6.22 (226-230 m) | 26.1 | 29.7 | 34.4 | | | | | | |
| +6.35 (226-306 m) | 27.2 | 30.9 | 35.8 | | | | | | |
| +4.8* | 10.6 | 12.5 | 15.1 | | | | | | |

The source and choice of $\delta^{15}N$ endmember values are described in Methods. Depth-range over which measured nitrate $\delta^{15}N$ values were averaged (nitrate concentration weighted) is given in parentheses. Bold values show the percentage contribution values calculated with the preferred endmember $\delta^{15}N$ values.

*global average δ^{15} N of deep-water nitrate (ref²⁹)

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863 Supplementary Table 2| Results of multiple linear regression model

| | Mean flux (mg m ⁻² d ⁻¹) | | | Carrying | coefficient* | Ballasted POC flux (%) | | |
|-------------------|---|--------------------------|-----------|---------------------|---------------------|------------------------|------------|-----------------|
| Group | POC | POC Opal + Calcite Li | | Opal + Calcite | | Opal + Calcite | Lithogenic | R ^{2‡} |
| <u>NOG groups</u> | | | | | | | | |
| High POC (10) | 1.64±0.43 | 17.4±6.7 | 9.14±1.9 | 0.027 ^{ns} | 0.126 | 29.1 | 70.1 | 0.94 |
| Low POC (31) | 0.88±0.21 | 13.0±3.5 | 8.38±2.2 | 0.036 | 0.048 | 53.7 | 45.7 | 0.98 |
| | | | | | | | | |
| High Lith (9) | 1.31±0.58 | 15.6±1.81 | 11.3±1.44 | 0.067 ns | 0.025 ^{ns} | 79.7 | 21.2 | 0.87 |
| Low Lith (32) | 0.99±0.36 | 13.7±5.27 | 7.80±1.62 | 0.045 | 0.048 | 62.3 | 37.6 | 0.95 |
| SOG group (40) | 0.49±0.17 | 8.62±2.96 | 0.54±0.21 | 0.032 | 0.371 | 57.2 | 41.0 | 0.96 |

Values in parentheses indicate number of data points (=collection cups) pulled into each group. Statistically not significant carrying

coefficients (p>0.01) are marked with 'ns'.

* Correlation coefficients of multiple linear regression determined from equation (2) (see Methods).

[‡]Overall model fit









Supplementary Figure 2 Nitrogen isotope data for water column total nitrate, suspended
particles and trap material near the study. Concentrations of nitrate and chlorophyll
measured at the study sites are also shown. Data sources are described in the figure legend with
complete references provided in the Methods section.