

# Quantification of ocean heat uptake from changes in atmospheric O<sub>2</sub> and CO<sub>2</sub> composition

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**The ocean is the main source of thermal inertia in the climate system<sup>1</sup>. During recent decades, ocean heat uptake has been quantified by using hydrographic temperature measurements and data from the Argo float program, which expanded its coverage after 2007<sup>2,3</sup>. However, these estimates all use the same imperfect ocean dataset and share additional uncertainties resulting from sparse coverage, especially before 2007<sup>4,5</sup>. Here we provide an independent estimate by using measurements of atmospheric oxygen (O<sub>2</sub>) and carbon dioxide (CO<sub>2</sub>)—levels of which increase as the ocean warms and releases gases—as a whole-ocean thermometer. We show that the ocean gained  $1.33 \pm 0.20 \times 10^{22}$  joules of heat per year between 1991 and 2016, equivalent to a planetary energy imbalance of  $0.83 \pm 0.11$  watts per square metre of Earth's surface. We also find that the ocean-warming effect that led to the outgassing of O<sub>2</sub> and CO<sub>2</sub> can be isolated from the direct effects of anthropogenic emissions and CO<sub>2</sub> sinks. Our result—which relies on high-precision O<sub>2</sub> measurements dating back to 1991<sup>6</sup>—suggests that ocean warming is at the high end of previous estimates, with implications for policy-relevant measurements of the Earth response to climate change, such as climate sensitivity to greenhouse gases<sup>7</sup> and the thermal component of sea-level rise<sup>8</sup>.**

As shown in Fig. 1, recent temperature-based hydrographic estimates of ocean warming<sup>9–12</sup> show good agreement for the years 2007–2016 ( $1.09 \pm 0.10 \times 10^{22}$  to  $1.16 \pm 0.20 \times 10^{22}$  J yr<sup>-1</sup>), but a larger spread when extending back to include the sparser data of the 1990s ( $0.90 \pm 0.09 \times 10^{22}$  to  $1.36 \pm 0.10 \times 10^{22}$  J yr<sup>-1</sup> for 1993–2015). The spread is mostly caused by gap-filling methods and systematic errors<sup>5,9</sup>, which together introduce uncertainties of up to 25%–50% in warming trends<sup>4</sup>. Because temperature-based estimates also use the same upper-ocean observations and linear warming trend for depths below 2,000 m (ref. <sup>11</sup>), they may share additional unknown systematic errors<sup>12</sup>. An alternative method based on the top of the atmosphere energy balance<sup>13</sup> is also not truly independent, because it is subject to large systematic errors when estimating long-term trends and therefore depends on the same hydrographic measurements for calibration<sup>13–15</sup>. Here we introduce a third method, based on changes in the abundances of gases in the atmosphere, which respond to whole-ocean warming through the temperature dependence of gas solubility in sea water. This method is not limited by data sparseness, because fast mixing in the atmosphere efficiently integrates the global ocean signal.

Changes in ocean heat content on seasonal<sup>16</sup> and glacial–interglacial<sup>17</sup> timescales have been reconstructed using measurements of noble gases in modern or ancient air. Our method is similar, but instead of relying on noble gases (for example, ratios of argon to nitrogen), which lack sufficient accuracy as yet<sup>16</sup>, we rely on measurements of atmospheric O<sub>2</sub> and CO<sub>2</sub>, which can be summed to yield a tracer ‘atmospheric potential oxygen’ (APO) that responds to warming similarly to a noble gas<sup>18</sup>. When the ocean warms, the solubility of O<sub>2</sub> and CO<sub>2</sub> drops, and the amount of gas lost by the ocean can be quantified with the complementary change observed in the atmosphere. Precise atmospheric

O<sub>2</sub> measurements began in 1991 (CO<sub>2</sub> in 1958), enabling APO-based reconstructions of ocean heat content that span nearly three decades<sup>6</sup>.

APO (O<sub>2</sub> +  $1.1 \times$  CO<sub>2</sub>) is computed using observed atmospheric O<sub>2</sub>/N<sub>2</sub> molar ratios and CO<sub>2</sub> molar fractions (see Methods)<sup>6,19</sup>. By design, APO is insensitive to exchanges with land ecosystems, which produce changes in O<sub>2</sub> and CO<sub>2</sub> that largely cancel in APO owing to their approximate 1.1 O<sub>2</sub>/C oxidative ratio. Time-series measurements at remote sites show a global long-term decline in APO, with  $\Delta\text{APO}_{\text{OBS}}$  being  $-243.70 \pm 10.10$  per meg (units defined in the Methods) between 1991 and 2016.  $\Delta\text{APO}_{\text{OBS}}$  is driven by four primary contributors, illustrated in Fig. 2:

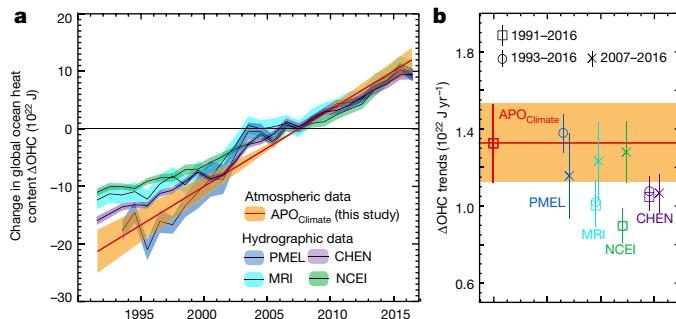
$$\Delta\text{APO}_{\text{OBS}} = \Delta\text{APO}_{\text{FF}} + \Delta\text{APO}_{\text{Cant}} + \Delta\text{APO}_{\text{AtmD}} + \Delta\text{APO}_{\text{Climate}} \quad (1)$$

where  $\Delta\text{APO}_{\text{FF}}$  is the decrease in APO caused by industrial processes (fossil-fuel burning and cement production), which in aggregate consume more than 1.1 moles of O<sub>2</sub> for each mole of CO<sub>2</sub> released;  $\Delta\text{APO}_{\text{Cant}}$  accounts for the oceanic uptake of excess anthropogenic atmospheric CO<sub>2</sub>;  $\Delta\text{APO}_{\text{AtmD}}$  accounts for air-sea exchanges driven by ocean fertilization from anthropogenic aerosol deposition (increased fertilization leads to increased photosynthesis, with a concomitant release of O<sub>2</sub> and uptake of CO<sub>2</sub>); and  $\Delta\text{APO}_{\text{Climate}}$  accounts for air-sea fluxes of O<sub>2</sub>, CO<sub>2</sub> and N<sub>2</sub> driven by ocean processes, including warming-induced changes in solubility, in ocean circulation, and in photosynthesis and respiration (N<sub>2</sub> influences O<sub>2</sub>/N<sub>2</sub> ratios). Here, we derive  $\Delta\text{APO}_{\text{Climate}}$  from equation (1) and show that it tracks ocean warming.

We estimate  $\Delta\text{APO}_{\text{FF}}$  using fossil-fuel and cement inventories<sup>20</sup>, finding  $\Delta\text{APO}_{\text{FF}} = -119.70 \pm 4.00$  per meg (Fig. 3).  $\Delta\text{APO}_{\text{Cant}}$  is controlled by the increase in atmospheric CO<sub>2</sub> and by ocean mixing, which is quantified by the distribution of transient tracers including chlorofluorocarbons (CFCs)<sup>21</sup>; we find that  $\Delta\text{APO}_{\text{Cant}} = -154.30 \pm 4.20$  per meg.  $\Delta\text{APO}_{\text{Cant}}$  is relatively precise because it excludes the effects of changing ocean biology and circulation on natural carbon fluxes that are included in  $\Delta\text{APO}_{\text{Climate}}$ .  $\Delta\text{APO}_{\text{AtmD}}$  is derived from ocean model simulations with and without aerosol fertilization (phosphate, iron and nitrogen; Extended Data Fig. 1)<sup>22</sup>.  $\Delta\text{APO}_{\text{AtmD}}$  is uncertain, owing in part to uncertainties in iron availability to photosynthetic organisms, but is relatively small compared with the other terms:  $\Delta\text{APO}_{\text{AtmD}} = 7.00 \pm 3.50$  per meg. From equation (1), we thereby find that  $\Delta\text{APO}_{\text{Climate}} = 23.20 \pm 12.20$  per meg, corresponding to a least-squares linear trend of  $+1.16 \pm 0.15$  per meg per year—larger than the trends expected from 26-year natural variations alone in four Earth-system models (the Community Earth System Model (CESM) and the Geophysical Fluid Dynamics Laboratory (GFDL), Institut Pierre Simon Laplace (IPSL) and University of Victoria (UVic) models). As shown in Fig. 3, a clear increase in  $\Delta\text{APO}_{\text{Climate}}$  emerges over the period January 1991 to the end of December 2016.

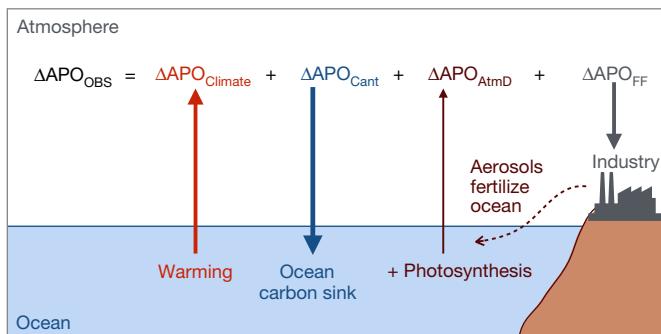
A starting point for understanding  $\Delta\text{APO}_{\text{Climate}}$  is to imagine that O<sub>2</sub> and CO<sub>2</sub> behave like inert gases, such that the air-sea fluxes are dominated by temperature-driven solubility changes. In this case,

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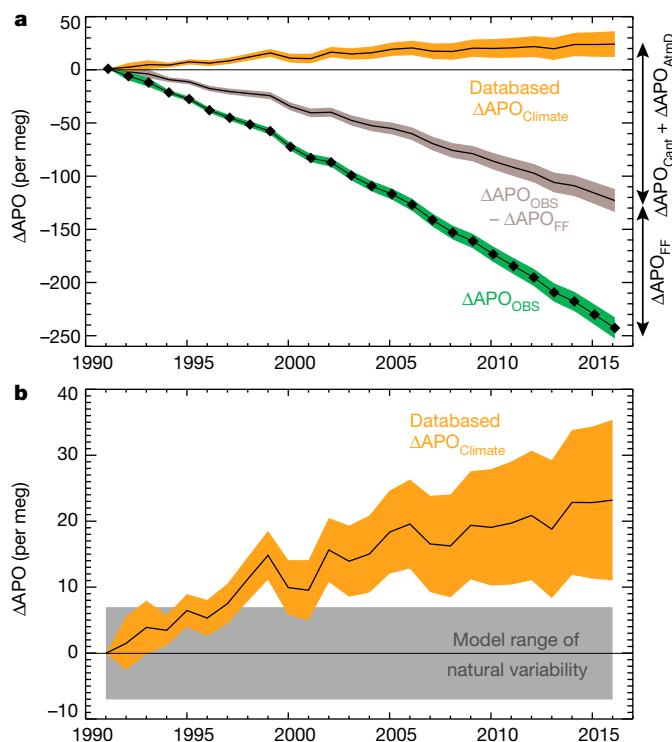


APO would increase by around 0.8 per meg per  $10^{22}\text{ J}$  of warming, with changes in  $\text{O}_2$  and  $\text{CO}_2$  solubility accounting for an increase of +1.0 per meg per  $10^{22}\text{ J}$ , partly offset by the  $\text{N}_2$  contribution of −0.2 per meg per  $10^{22}\text{ J}$  (Methods). Support for the dominance of solubility in  $\Delta\text{APO}_{\text{Climate}}$  can be found in the natural distribution of  $\text{O}_2$  and carbon in the ocean. Ocean potential oxygen (OPO) is a dissolved tracer that mirrors  $\text{APO}_{\text{Climate}}$  and tracks changes in air-sea  $\text{O}_2$  and  $\text{CO}_2$  fluxes<sup>18</sup>. Observed OPO abundance is strongly tied to ocean potential temperature (Fig. 4): warming induces OPO loss, and cooling induces OPO gain. The observed OPO-to-temperature trend of  $-4.45\text{ nmol J}^{-1}$  is within 17% of the trend of  $-3.70\text{ nmol J}^{-1}$  expected from solubility alone (OPO<sub>sat</sub>-to-temperature). Biological effects (related to changes in ocean circulation and photosynthesis/respiration) on  $\text{CO}_2$  and  $\text{O}_2$  substantially cancel in OPO (Extended Data Fig. 2), while thermal impacts reinforce each other, with warming waters releasing both  $\text{O}_2$  and  $\text{CO}_2$  to the atmosphere and increasing  $\Delta\text{APO}_{\text{Climate}}$ .

Further support for the dominance of solubility in  $\Delta\text{APO}_{\text{Climate}}$  is found on multidecadal timescales in the four Earth-system models mentioned above, which yield OPO-to-temperature ratios of between  $-4.71$  and  $-4.38\text{ nmol J}^{-1}$ , bracketing the ratio of  $-4.45\text{ nmol J}^{-1}$ .

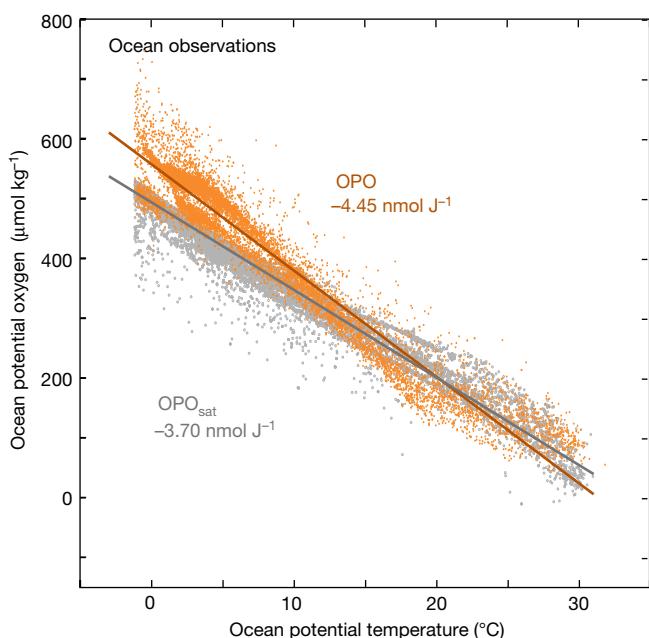


**Fig. 2 | Processes contributing to observed changes in atmospheric potential oxygen ( $\Delta\text{APO}_{\text{Obs}}$ ).** Industrial processes (fossil-fuel burning and cement production;  $\Delta\text{APO}_{\text{FF}}$ ) and the ocean sink for anthropogenic carbon ( $\Delta\text{APO}_{\text{Cant}}$ ) remove APO from the atmosphere. The fertilization effect of anthropogenic aerosol deposition ( $\Delta\text{APO}_{\text{AtmD}}$ )—which promotes marine photosynthesis—and the changes in solubility, biology and ocean circulation due to warming ( $\Delta\text{APO}_{\text{Climate}}$ ) release APO into the atmosphere. Our study shows that  $\Delta\text{APO}_{\text{Climate}}$  can be used to estimate long-term changes in global ocean warming.



found in hydrographic observations (Extended Data Fig. 3). The models also simulate a very close relationship between  $\Delta\text{APO}_{\text{Climate}}$  and the change in global ocean heat content ( $\Delta\text{OHC}$ ) that occurs during the simulations (1920–2100), with an atmospheric build-up in APO of between 0.83 and 0.99 per meg per  $10^{22}\text{ J}$  (Extended Data Figs. 3, 4)—close to the ratio expected from temperature-driven solubility changes alone (0.8 per meg per  $10^{22}\text{ J}$ ). By dividing the simulated APO change into separate biological and thermal components, we show that solubility changes account for more than 80% of  $\Delta\text{APO}_{\text{Climate}}$ , while biologically driven changes account for 5% to 20% (Extended Data Fig. 4). This partitioning found in response to transient warming is very similar to the partitioning found in hydrographic data (where solubility and biology contribute 83% and 17%, respectively, to the OPO-to-temperature ratio; Fig. 4).

Small differences between individual model  $\Delta\text{APO}_{\text{Climate}}$ -to- $\Delta\text{OHC}$  relationships (0.83 to 0.99 per meg per  $10^{22}\text{ J}$ ) reflect systematic differences in biological fluxes. Models with stronger biological effects (IPSL and UVic) yield stronger oceanic loss of OPO and stronger release of APO for a given ocean warming (more negative OPO-to-temperature and higher  $\Delta\text{APO}_{\text{Climate}}$ -to- $\Delta\text{OHC}$ ; Extended Data Fig. 3b). Using this relationship, we find that a  $\Delta\text{APO}_{\text{Climate}}$ -to- $\Delta\text{OHC}$  ratio of  $0.87 \pm 0.03$  per meg per  $10^{22}\text{ J}$  is compatible with the observed OPO-to-temperature ratio. Combining this constrained  $\Delta\text{APO}_{\text{Climate}}$ -to- $\Delta\text{OHC}$  ratio ( $0.87 \pm 0.03$  per meg per  $10^{22}\text{ J}$ ) with the observation-based trend in  $\Delta\text{APO}_{\text{Climate}}$  ( $1.16 \pm 0.18\text{ per meg yr}^{-1}$ ) yields a warming trend of  $1.33 \pm 0.20 \times 10^{22}\text{ J yr}^{-1}$  between 1991 and 2016. As shown in Fig. 1, this APO-based estimate of ocean heat uptake agrees well, within uncertainties, with the highest temperature-based estimates (from the Pacific Marine Environmental Laboratory (PMEL)<sup>10</sup>, available only for



**Fig. 4 | Observed link between potential oxygen and ocean heat.** OPO concentrations in situ (OPO, yellow) and at saturation based on O<sub>2</sub> and CO<sub>2</sub> solubility (OPO<sub>sat</sub>, grey) as a function of ocean temperature in the GLODAPv2 database<sup>32</sup>.

1993–2015) and marginally with the two next estimates (from Cheng et al.<sup>12</sup> (CHEN) and the Japanese Meteorological Institute (MRI)<sup>9</sup>).

The APO data provide a much-needed independent confirmation of the recent upward revisions in estimates of ocean warming<sup>5,9</sup>. A higher value of ΔOHC compatible with both APO<sub>Climate</sub> and in situ temperature approaches ( $1.13$  to  $1.46 \times 10^{22} \text{ J yr}^{-1}$ ) calls for a steric sea level rise of  $1.34$ – $1.74 \text{ mm yr}^{-1}$  (Methods), in full agreement with satellite constraints on thermal expansion, corrected for the freshwater contribution ( $1.50 \pm 0.40 \text{ mm yr}^{-1}$ )<sup>8,23</sup>.

A higher ΔOHC will also affect the equilibrium climate sensitivity, recently estimated at between +1.5 K and +4.5 K if CO<sub>2</sub> is doubled<sup>1</sup>. This estimated range reflects a decrease in the lower bound from 2 K to 1.5 K owing to downward revision of the aerosol cooling effect (in the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report, as compared with the Fourth Assessment Report)<sup>1,24</sup>, but relied on a low ΔOHC value ( $0.80 \times 10^{22} \text{ J yr}^{-1}$  for 1993–2010). An upward revision of the ocean heat gain by  $+0.5 \times 10^{22} \text{ J yr}^{-1}$  (to  $1.30 \times 10^{22} \text{ J yr}^{-1}$  from  $0.80 \times 10^{22} \text{ J yr}^{-1}$ ) would push up the lower bound of the equilibrium climate sensitivity from 1.5 K back to 2.0 K (stronger warming expected for given emissions), thereby reducing maximum allowable cumulative CO<sub>2</sub> emissions by 25% to stay within the 2 °C global warming target (see Methods).

We find that the APO–heat coupling (APO<sub>Climate</sub>) is most robust on decadal and longer timescales. Strong cancellation of biological O<sub>2</sub> and CO<sub>2</sub> fluxes is not expected on all temporal scales<sup>25</sup>. On seasonal timescales, air-sea O<sub>2</sub> fluxes driven by marine photosynthesis are around eight times larger than those of CO<sub>2</sub> owing to slow equilibration of CO<sub>2</sub> (ref. <sup>25</sup>). More complex coupling is also possible on interannual timescales<sup>26</sup>, such as the weaker lagged air-sea CO<sub>2</sub> flux compared with the O<sub>2</sub> flux during El Niño events<sup>27</sup>.

Atmospheric O<sub>2</sub> and CO<sub>2</sub> measurements have been applied previously to estimate global land and ocean CO<sub>2</sub> sinks, but relied on estimates of ocean heat content and model-based oceanic O<sub>2</sub>-to-heat ratios to correct for climate-driven O<sub>2</sub> outgassing<sup>28–30</sup>. Here we have reversed this logic, using estimates of other quantities to constrain the ocean heating. Our approach exploits the APO–heat relationship, which is stronger than the O<sub>2</sub>–heat relationship. Further work to constrain the separate contributions of O<sub>2</sub> and CO<sub>2</sub> to APO is needed to refine estimates of land and ocean carbon sinks using atmospheric O<sub>2</sub> and CO<sub>2</sub>.

## Online content

Any methods, additional references, Nature Research reporting summaries, source data, statements of data availability and associated accession codes are available at <https://doi.org/10.1038/s41586-018-0651-8>.

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**Competing interests** The authors declare no competing interests.

**Additional information**

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## METHODS

**Observed changes in APO.** A change in atmospheric potential oxygen concentration (in per meg) is defined following<sup>19</sup>:

$$(\delta\text{APO}) = (\delta\text{O}_2/\text{N}_2) + \frac{1.1}{X_{\text{O}_2}} \times (X_{\text{CO}_2} - 350)$$

with

$$(\delta\text{O}_2/\text{N}_2) = \frac{\text{O}_2/\text{N}_2(\text{sample})}{\text{O}_2/\text{N}_2(\text{reference})} - 1$$

where  $(\delta\text{O}_2/\text{N}_2)$  is the atmospheric change in  $\text{O}_2/\text{N}_2$  ratios (in per meg);  $X_{\text{CO}_2}$  is the  $\text{CO}_2$  concentration in the air parcel (in p.p.m., that is,  $\mu\text{mol mol}^{-1}$ ) and 350 is an arbitrary reference; 1.1 is the approximate  $\text{O}_2/\text{CO}_2$  ratio of terrestrial ecosystems<sup>33</sup>; and  $X_{\text{O}_2}$  ( $= 0.2094$ ) is the reference value of atmospheric mole fraction of  $\text{O}_2$  necessary to convert  $X_{\text{CO}_2}$  from p.p.m. to per meg units.

$\Delta\text{APO}_{\text{OBS}}$  is computed from in situ atmospheric changes in  $\text{CO}_2$  concentrations and  $\text{O}_2/\text{N}_2$  ratios<sup>19</sup> measured at stations of the Scripps Institution of Oceanography network (available online at <http://scrippso2.ucsd.edu>)<sup>6</sup>. The global average  $\Delta\text{APO}_{\text{OBS}}$  is based on data from the three stations with longest record (1991 to 2016), that is, La Jolla ( $32.9^\circ\text{N}, 117^\circ\text{W}$ ), Alert ( $82.5^\circ\text{N}, 62.5^\circ\text{W}$ ) and Cape Grim ( $40.5^\circ\text{S}, 144.5^\circ\text{E}$ ) and weighted by the stations' latitudinal distribution<sup>34</sup>. Station annual means are based on bimonthly data fit to a four-harmonic seasonal cycle and a stiff long-term trend<sup>6</sup>. The uncertainty on  $\Delta\text{APO}_{\text{OBS}}$  was computed by generating  $10^6$  time series with noise scaled to the random and systematic errors of APO data detailed in Extended Data Table 3. The uncertainty is taken as the  $1\sigma$  interval ( $\pm 1$  standard deviation) from these  $10^6$  realizations (Fig. 3).

**Effects of fossil-fuel burning and cement production on APO.**  $\Delta\text{APO}_{\text{FF}}$  is estimated using annual  $\text{CO}_2$  emissions from oil, coal, gas, flaring and cement production ( $\Delta\text{CO}_{2(i)}$  in moles)<sup>20</sup> weighted by their  $\text{O}_2/\text{C}$  combustion ratios,  $R_i$  (ref. <sup>6</sup>):

$$\Delta\text{APO}_{\text{FF}}(\text{per meg}) = \sum_i \frac{1.1 - R_i}{X_{\text{O}_2}} \times \frac{\Delta\text{CO}_{2(i)}}{M_{\text{air}}}$$

where  $M_{\text{air}}$  is the number of moles of dry air in the atmosphere (convert moles of  $\text{CO}_2$  to p.p.m.).

The uncertainty on  $\Delta\text{APO}_{\text{FF}}$  includes uncertainties in  $\text{CO}_2$  emissions ( $\Delta\text{CO}_{2(i)}$ )<sup>35</sup> and in combustion ratios ( $R_i$  in Extended Data Table 3)<sup>36</sup>. Uncertainties on  $\Delta\text{CO}_{2(i)}$  are not independent in time and were estimated using an autoregressive model<sup>37</sup> (1,000 realizations); uncertainties on  $R_i$  were computed using a Monte Carlo approach (1,000 realizations). The uncertainty on  $\Delta\text{APO}_{\text{FF}}$  was then estimated by combining the 1,000 realizations of  $\Delta\text{CO}_{2(i)}$  and the 1,000 realizations of  $R_i$ , yielding a set of  $10^6$  estimates of  $\Delta\text{APO}_{\text{FF}}$ .

**Effect of ocean anthropogenic carbon uptake on APO.** We represent the ocean  $\text{CO}_2$  uptake ( $\Delta\text{CO}_2$ ) as the sum of three contributions:

$$\Delta\text{CO}_2 = \Delta\text{Cant}_0 + \Delta\text{Cant}' + \Delta\text{CO}_{2\text{Climate}} \quad (2)$$

where  $\Delta\text{Cant}_0$  is the flux driven by the rise in  $\text{CO}_2$  assuming steady ocean circulation ( $\Delta\text{Cant}_0$  is negative, corresponding to uptake by the ocean);  $\Delta\text{CO}_{2\text{Climate}}$  is the flux driven by the action of climate on natural carbon in the ocean ( $\Delta\text{CO}_{2\text{Climate}}$  is positive, that is, warming reduces the uptake of natural carbon); and  $\Delta\text{Cant}'$  is the remainder, which accounts for impact of circulation changes on the uptake of carbon driven by rising  $\text{CO}_2$  ( $\Delta\text{Cant}'$  is positive, that is, warming reduces the uptake of  $\text{C}_{\text{ant}}$ ).  $\Delta\text{APO}_{\text{Cant}}$  can be expressed as the weighted sum of the two terms  $\Delta\text{Cant}_0$  and  $\Delta\text{Cant}'$ :

$$\Delta\text{APO}_{\text{Cant}}(\text{per meg}) = \frac{1.1}{X_{\text{O}_2} \times M_{\text{air}}} \times (\Delta\text{Cant}_0 + \Delta\text{Cant}')$$

where  $\Delta\text{Cant}_0$  and  $\Delta\text{Cant}'$  are in moles. Note that  $\Delta\text{CO}_{2\text{Climate}}$  is accounted for in  $\Delta\text{APO}_{\text{Climate}}$ .

$\Delta\text{Cant}_0$  is taken from a recent ocean inversion scheme with assimilation of observed potential temperature, salinity, radiocarbon and CFC-11 (ref. <sup>21</sup>), updated to 2016.  $\Delta\text{Cant}'$  cannot be derived from observations and was estimated at  $0.05 \text{ Pg C yr}^{-1}$ , equivalent to a trend of  $+0.2 \text{ per meg}^{-1}$ , using model simulations (see 'Model anthropogenic  $\Delta\text{Cant}'$ ').

The uncertainty on  $\Delta\text{APO}_{\text{Cant}}$  is related to uncertainties in  $\Delta\text{Cant}_0$  and  $\Delta\text{Cant}'$ . We allow for uncertainty in  $\Delta\text{Cant}_0$  following ref. <sup>21</sup>, using the ten sensitivity experiments (on ocean vertical and isopycnal diffusivities, data constraint, gas-exchange coefficient and so on) available for the ocean inversion and an estimate of the interannual variability in the ocean sink of a  $0.2 \text{ Pg C yr}^{-1}$ . We also allow an additional 1% uncertainty (less than  $0.03 \text{ Pg C yr}^{-1}$ ) in  $\Delta\text{Cant}_0$  resulting from imperfectly known atmospheric  $\text{CO}_2$  history<sup>38</sup>, taking account of sensitivity to start date (1765

versus 1791), to degree of temporal smoothing, and to using different versions of the record since 1958 (Mauna Loa record versus average of Mauna Loa and South Pole records). This estimate used a variant of the box-diffusion model<sup>39</sup>, and  $\text{CO}_2$  data from ref. <sup>40</sup> and the Scripps  $\text{CO}_2$  program (<https://library.ucsd.edu/dc/collection/bb3381541w>). Uncertainties on  $\Delta\text{Cant}'$  are assumed to be 100% of the model-based estimate of  $\Delta\text{Cant}'$ .

**Ocean fertilization and atmospheric deposition of aerosols.** Deposition of anthropogenic aerosol from fossil fuel, biomass burning and other processes fertilizes the ocean with nutrients and increases surface photosynthesis and subsurface respiration<sup>41–43</sup>. The effect of aerosol fertilization is partly counterbalanced by biological processes such as a decline in nitrogen fixation, which would be immediate, and an increase in denitrification in the water column, which would be on timescales of several hundred years<sup>44</sup>. Fixed anthropogenic nitrogen also fertilizes the land biosphere and coastal oceans by river runoffs, but, in these cases, efficient denitrification returns fixed nitrogen to the atmosphere and has little impact on the APO budget on the decadal timescales considered here. The impact of anthropogenic aerosol on  $\text{O}_2$ ,  $\text{CO}_2$  and APO air–sea fluxes is evaluated with the IPSL ocean model NEMO-PISCES v2 (ref. <sup>45</sup>), using the difference between simulations with aerosols and a simulation in which the aerosol deposition is fixed to a constant preindustrial value (equivalent to year 1850, Extended Data Fig. 1)<sup>22</sup>. We use four simulations with varying aerosols: one includes the combined effect of nitrogen (N), iron (Fe) and phosphorus (P) aerosol deposition, whereas the other three include only their individual contributions (N-only, Fe-only or P-only; Extended Data Fig. 1 and Extended Data Table 5). Uncertainties at the  $1\sigma$  level on  $\Delta\text{APO}_{\text{AtmD}}$  are assumed to be  $\pm 50\%$ . See Extended Data Table 4.

Combined, N, Fe and P deposition accounts for an  $\text{O}_2$  outgassing of  $19.0 \text{ Tmol yr}^{-1}$  for the 1980–2007 period ( $16 \text{ Tmol yr}^{-1}$  for the entire 1960–2007 simulation period) and an oceanic  $\text{CO}_2$  uptake of  $8.3 \text{ Tmol yr}^{-1}$  for the 1980–2007 period ( $6.8 \text{ Tmol yr}^{-1}$  for the entire 1960–2007 simulation period; Extended Data Fig. 1 and Extended Data Table 5). The overall impact on  $\Delta\text{APO}_{\text{AtmD}}$  is  $+0.27 \text{ per meg yr}^{-1}$  over 27 years of simulation (1980–2007), which we extrapolate to our 1991–2016 period. Increased  $\text{O}_2$  outgassing accounts for an increase in APO of  $+0.51 \text{ per meg yr}^{-1}$ , and  $\text{CO}_2$  uptake accounts for a change in APO of  $-0.24 \text{ per meg yr}^{-1}$  ( $\Delta\text{APO}_{\text{AtmD(O2)}}$  and  $\Delta\text{APO}_{\text{AtmD(CO2)}}$  in Extended Data Table 3).

The overall effect of N, Fe and P is smaller than the sum of the individual effects (Extended Data Fig. 1), because of the interplay between the aerosol deposition pattern and nutrient co-limitations in the ocean. Phytoplankton growth in the ocean depends on the availability of the most limiting nutrient. While more available N will promote photosynthesis in regions where N is limiting (for example, the tropical Atlantic Ocean), the effect is negligible in regions where Fe, P or any other nutrient is limiting (such as the Southern Ocean; see Fig. 2 in ref. <sup>22</sup>).

To our knowledge this is the first estimate of the effect of anthropogenic aerosol deposition on both  $\text{O}_2$  and  $\text{CO}_2$  air–sea fluxes at the global scale. Note however that ref. <sup>6</sup> used anthropogenic aerosol N inventories and scaling arguments to estimate an ocean  $\text{O}_2$  loss due to anthropogenic N deposition only of about  $10 \pm 10 \text{ Tmol yr}^{-1}$ , slightly lower than our model estimate of  $15.5 \text{ Tmol yr}^{-1}$ .

**$\Delta\text{APO}_{\text{Climate}}$  trends and uncertainty analysis.** We compute the APO response to climate change ( $\Delta\text{APO}_{\text{Climate}}$ ) via:

$$\Delta\text{APO}_{\text{Climate}} = \Delta\text{APO}_{\text{OBS}} - \Delta\text{APO}_{\text{FF}} - \Delta\text{APO}_{\text{Cant}} - \Delta\text{APO}_{\text{AtmD}}$$

We combine the estimates of  $\Delta\text{APO}_{\text{FF}}$ ,  $\Delta\text{APO}_{\text{Cant}}$  and  $\Delta\text{APO}_{\text{AtmD}}$  to obtain  $10^6$  time series of  $\Delta\text{APO}_{\text{FF}} + \Delta\text{APO}_{\text{Cant}} + \Delta\text{APO}_{\text{AtmD}}$ , and obtain  $10^6$  time series of  $\Delta\text{APO}_{\text{Climate}}$  using the  $10^6$  time series of  $\Delta\text{APO}_{\text{OBS}}$ . We computed the  $\Delta\text{APO}_{\text{Climate}}$  least-squares linear trend using the standard deviation of the  $10^6$  realizations of  $\Delta\text{APO}_{\text{Climate}}$  as the error. We find a  $\Delta\text{APO}_{\text{Climate}}$  trend of  $1.16 \pm 0.15 \text{ per meg yr}^{-1}$  for 1991–2016.

**Hydrography-based estimates of ocean heat uptake.** We used four global-ocean estimates of  $\Delta\text{OHC}$ , based on hydrographic measurements, in Fig. 1. Ocean warming rates from the surface to 2,000 m are from ref. <sup>10</sup> (PMEL), ref. <sup>9</sup> (MRI; <https://climate.mri-jma.go.jp/pub/ocean/ts/v7.2/>), an updated version of ref. <sup>31</sup> (NCEI; [www.nodc.noaa.gov/OC5/3M\\_HEAT\\_CONTENT/basin\\_avt\\_data.html](http://www.nodc.noaa.gov/OC5/3M_HEAT_CONTENT/basin_avt_data.html)) and ref. <sup>12</sup> (CHEN; [http://159.226.119.60/cheng/images\\_files/TOA\\_OHC\\_error-bar\\_1940\\_2015\\_2.txt](http://159.226.119.60/cheng/images_files/TOA_OHC_error-bar_1940_2015_2.txt)), with the revised deep-ocean (depths greater than 2,000 m) constant linear warming rate of  $0.10 \pm 0.03 \times 10^{22} \text{ J yr}^{-1}$  of ref. <sup>11</sup> being based on the global ship-based sections program (GO-SHIP; <http://www.go-ship.org>)<sup>46</sup>.

**Ocean observations of ocean potential oxygen.** We used in situ ocean observations from GLODAPv2 (ref. <sup>32</sup>) combined with an anthropogenic carbon estimate<sup>21</sup> interpolated at the location of each sample to compute 78,456 values (GLODAPv2 quality control = 0; marginal seas and coastal waters were removed) of  $\text{OPO}^{18}$  as follows:

$$\text{OPO} = \text{O}_2^* + 1.1 \times \text{C}_{pi}^*$$

where  $O_2^*$  and  $C_{pi}^*$  are ocean conservative tracers related to air-sea fluxes of  $O_2$  and preindustrial carbon<sup>47</sup>. The thermal component (solubility-driven) of OPO ( $OPO_{sat}$ ) is computed as:

$$OPO_{sat} = O_{2sat} + 1.1 \times C_{pisat}$$

where  $O_{2sat}$  is the dissolved  $O_2$  concentration at saturation with the observed temperature and salinity<sup>48</sup>, and  $C_{pisat}$  is the dissolved inorganic carbon concentration expected at the observed temperature and salinity and assuming equilibrium with a preindustrial partial pressure of  $CO_2$  of 280 p.p.m., using pre-formed alkalinity<sup>49</sup>.

**Solubility-driven changes in OPO and APO.** Extended Data Fig. 2 shows a tight and quasilinear link between observed OPO and potential temperature ( $-4.4 \text{ nmol J}^{-1}; r^2 = 0.95$ ), similar to the link found between  $OPO_{sat}$  and potential temperature ( $-3.7 \text{ nmol J}^{-1}; r^2 = 0.93$ ). This suggests that changes in OPO and hence  $\Delta APO_{Climate}$  are driven primarily by changes in thermal air-sea fluxes. In these observations, departures of dissolved oxygen and carbon concentrations ( $O_2^*$  and  $C_{pi}^*$ ) from their respective saturation curves ( $O_{2sat}$  and  $C_{pisat}$ ) due to biological activity tend to balance (Extended Data Fig. 2). By contrast, thermal effects reinforce each other ( $O_{2sat}$  and  $C_{pisat}$  both decrease with increasing temperature) and biological effects compensate for each other ( $O_2^* > O_{2sat}$  and  $C_{pi}^* < C_{pisat}$ ).

The change in APO expected from changes in gas solubility in the ocean is an increase of 3.0 nmol per J of warming, which includes the outgassing of  $O_2$  and  $CO_2$  following  $OPO_{sat}$  ( $3.7 \text{ nmol J}^{-1}$ ) and the release of  $N_2$  ( $0.7 \text{ nmol J}^{-1}$ ) (Extended Data Fig. 2b). A change of 3.0 nmol per J of warming is equivalent to an increase of  $1.0 \text{ per meg per } 10^{22} \text{ J} = (3.0 \times 10^{-9}) / (3.7 \times 10^{19}) \times 10^{22} = 0.8 \times 10^{-6} = 1.0 \text{ per meg per } 10^{22} \text{ J}$ , with  $3.7 \times 10^{19}$  being the number of moles of  $O_2$  in the atmosphere).  $O_2$  and  $CO_2$  solubility alone yields an increase in APO of 1.0 per meg per  $10^{22} \text{ J}$ , which is partly counterbalanced by the outgassing of  $N_2$  that decreases APO by 0.2 per meg per  $10^{22} \text{ J}$  (via the increase in the  $O_2/N_2$  ratio).

**Earth system model experiments.** We used four Earth-system models (ESMs): the Geophysical Fluid Dynamics Laboratory Earth System Models with a nominally level vertical coordinate version, GFDL-ESM2M (called GFDL here)<sup>50,51</sup>, the Institut Pierre-Simon Laplace Coupled Model 5 version IPSL-CM5A-LR (IPSL here)<sup>52</sup>, the Community Earth System Model large ensemble CESM-LE (CESM here)<sup>53</sup> and the UVic model version 2.9 (UVic here)<sup>54</sup>. Evaluation of these models and their biogeochemical components can be found in previous studies<sup>51,55–57</sup>. GFDL, IPSL and UVic participated in the Coupled Model Intercomparison Project Phase 5 (CMIP5)<sup>58</sup>.

For GFDL, IPSL and UVic, we used the CMIP5 business as usual ‘historical-RCP8.5’ scenario, the feedback experiment ‘esmFdbk3’ (which includes only warming-driven changes associated with anthropogenic emissions, such as radiation effects), and the fixed-climate experiment ‘esmFixClim3’, which includes only the direct biogeochemical effects of increasing atmospheric  $CO_2$  (for example, uptake of anthropogenic carbon, acidification and so on). For CESM, we also used the historical-RCP8.5 experiment and the separation between anthropogenic carbon from the natural carbon available in this model (carbon tracer separation approach). The feedback approach used for GFDL, IPSL and UVic removes all direct biogeochemical effects of rising atmospheric  $CO_2$  on the air-sea  $O_2$  and  $CO_2$  exchanges, whereas the natural carbon tracer separation approach used for CESM still includes the biogeochemical impacts of increasing atmospheric  $CO_2$  on the carbon cycle (for example, acidification) even while it excludes the anthropogenic carbon itself. However, we expect the effect on our results to be small and negligible.

We also used the multicentury preindustrial control simulation ‘piControl’ with no increase in atmospheric  $CO_2$  to correct for model drift and to estimate the natural internal variability of  $\Delta APO_{Climate}$  (Fig. 2). We used model results over the 1920–2100 period, which were available for the four models.

Model OPO was computed as for the observations. Note that for CESM we removed subsurface regions of high denitrification in the Eastern Equatorial Pacific Ocean and the Bay of Bengal, where oxygen and  $O_2^*$  in this model have unrealistic values<sup>59</sup>.

**Model anthropogenic  $\Delta Cant'$ .** The component  $\Delta Cant'$  was derived from equation (2) ( $\Delta Cant' = \Delta CO_2 - \Delta Cant_0 - \Delta CO_{2Climate}$ ) using CMIP5 model simulations.  $\Delta CO_2$  was taken from experiment RCP8.5,  $\Delta Cant_0$  from experiment esmFixClim3, and  $\Delta CO_{2Climate}$  from experiment esmFdbk3. Note that the control simulation was also used to correct model drift. We estimated  $\Delta Cant'$  to be  $0.05 \pm 0.05 \text{ Pg C yr}^{-1}$  for 1991–2016, based on the results of the three models—which individually yielded  $\Delta Cant'$  values of  $0.0 \text{ Pg C yr}^{-1}$  (IPSL),  $0.12 \text{ Pg C yr}^{-1}$  (GFDL) and  $0.12 \text{ Pg C yr}^{-1}$  (UVic)—and assuming an uncertainty of  $\pm 100\%$ . This corresponds to a trend of  $0.12 \pm 0.12 \text{ per meg yr}^{-1}$ .

**Model  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratios and uncertainty.** Model  $\Delta APO_{Climate}$  is computed using individual contributions from  $O_2$ ,  $CO_2$  and  $N_2$  as follows:

$$APO_{Climate}(\text{per meg}) = APO_{O_2} + APO_{CO_2} + APO_{N_2}$$

$$\begin{aligned} \Delta APO_{Climate}(\text{per meg}) &= \frac{1}{X_{O_2} 2pM_{air}} \\ &\times \left( \Delta F_{O_2} + 1.1 \times \Delta F_{CO_2} - \frac{X_{O_2}}{X_{N_2}} \times \Delta F_{N_2} \right) \end{aligned}$$

where  $\Delta F_{O_2}$ ,  $\Delta F_{CO_2}$  and  $\Delta F_{N_2}$  are the changes in air-sea fluxes of  $O_2$ ,  $CO_2$  and  $N_2$  respectively (in moles);  $M_{air}$  is the number of moles of dry air in the atmosphere; and  $X_{N_2}$  and  $X_{O_2}$  are the reference atmospheric mixing ratios of  $N_2$  and  $O_2$  respectively<sup>60</sup>.  $O_2$  and  $CO_2$  fluxes are simulated in the models.  $N_2$  air-sea fluxes, which affect the  $O_2$  atmospheric mixing ratio (because  $O_2$  constitutes around 20% of the atmospheric composition), are quantified from the global ocean temporal changes in  $N_2$  solubility computed from model changes in temperature and salinity<sup>61</sup>.

The link between long-term changes in  $APO_{Climate}$  and ocean heat content—that is,  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratios—were computed for each model using the 180 years of simulations (1920–2100). Resulting  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratios vary between 0.83 and 0.99 per meg per  $10^{22} \text{ J}$  of warming (Extended Data Fig. 3). These ratios include uncertainty in the natural climate variations on interannual and decadal timescales and uncertainty in the  $O_2/C$  oxidative ratio associated with global gains and losses of  $O_2$  and  $CO_2$  by terrestrial ecosystems. The uncertainty due to interannual variations was evaluated by computing  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratios using multiple 26-year-long segments from the 180-year simulations. We obtained 616  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratios (154 time series of 26 years per model), and used the standard deviation between these ratios as a measure of the uncertainty.

The  $O_2/C$  ratio is assumed to be 1.1 in our computation to follow the widely accepted definition of APO ( $APO = O_2 + 1.1 \times CO_2$ ), but is shown to have variations between 1 and 1.1 (ref. <sup>33</sup>). An oxidative ratio lower than 1.1 would yield a weaker  $\Delta APO_{Climate}$ -to- $\Delta OHC$  slope and hence a slightly higher estimate of  $\Delta OHC$  for a given  $\Delta APO_{Climate}$ . We evaluated the influence of the  $O_2/C$  ratio for each model by using the difference between  $\Delta APO_{Climate}$  computed with a ratio of 1.1 and  $\Delta APO_{Climate}$  computed with a ratio of 1. The two contributions to the uncertainties on the simulated  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratios (interannual variations and  $O_2/C$  ratio) combine to yield  $\pm 0.01$  per meg per  $10^{22} \text{ J}$  for the CESM and GFDL models,  $\pm 0.02$  per meg per  $10^{22} \text{ J}$  for the UVic model, and  $\pm 0.05$  per meg per  $10^{22} \text{ J}$  for the IPSL model ( $1\sigma$ ). These uncertainties are used in Extended Data Fig. 3.

**Steric component of sea-level rise.** We evaluated the steric component of sea-level rise associated with a  $\Delta OHC$  compatible with both  $APO_{Climate}$  and existing in situ temperature constraints (that is, between  $1.13 \times 10^{22} \text{ J yr}^{-1}$  and  $1.46 \times 10^{22} \text{ J yr}^{-1}$ ) to be between  $1.34 \text{ mm yr}^{-1}$  and  $1.74 \text{ mm yr}^{-1}$ . Following ref. <sup>62</sup>, this calculation assumes that 45% of the warming occurs below 700 m, and that the steric rise is 1 mm per  $0.60 \times 10^{22} \text{ J}$  above 700 m, and 1 mm per  $1.15 \times 10^{22} \text{ J}$  below 700 m (that is, a global steric rise of 1 mm per  $0.84 \times 10^{22} \text{ J}$ ). Assuming that 48% of the warming occurs below 700 m (ref. <sup>10</sup>) would yield a global steric rise of 1 mm per  $0.86 \times 10^{22} \text{ J}$  and change our estimate by less than 3%. Our estimate is also consistent with the recent hydrography-based estimate of the WCRP Global Sea Level Budget Group<sup>63</sup>.

**Ocean heat uptake, sea level and climate sensitivity.** Climate sensitivity has been estimated to fall within the range of  $+1.5 \text{ K}$  to  $+4.5 \text{ K}$  for a doubling of  $CO_2$  (ref. <sup>1</sup>). The impact of an increase in the ocean heat uptake on the effective equilibrium climate sensitivity (the apparent equilibrium climate sensitivity diagnosed from nonequilibrium conditions) can be estimated using a cumulative approach on the Earth energy balance (see Fig. 2 in ref. <sup>1</sup>):

$$N = F - \alpha \Delta T \quad (3)$$

where  $N$  is the global heat imbalance, which mostly consists of the ocean heat uptake;  $F$  is the radiative forcing (in  $\text{W m}^{-2}$ );  $\Delta T$  is the increase in surface temperature (in K) above a natural steady state; and  $\alpha$  is the climate feedback parameter (in  $\text{W m}^{-2} \text{ K}^{-1}$ ), which is inversely proportional to the effective equilibrium climate sensitivity<sup>1</sup>. All terms in equation (3) are time integrated over the period of interest.

The IPCC Fifth Assessment Report gives a  $\Delta OHC$  of  $0.80 \times 10^{22} \text{ J yr}^{-1}$  for 1993–2010, which is about  $0.5 \times 10^{22} \text{ J yr}^{-1}$  lower than the  $\Delta OHC$  that is compatible with both APO and hydrographic constraints. By applying equation (3)<sup>1</sup> to surface temperature data over the period 1991–2016 (HadCrut4 version 4.5, ref. <sup>64</sup>, with a 1860–1879 preindustrial baseline), we found that the upward revision of the global heat imbalance,  $N$ , by  $+0.5 \times 10^{22} \text{ J yr}^{-1}$  pushes up the lower bound of the equilibrium climate sensitivity from  $1.5 \text{ K}$  back to  $2.0 \text{ K}$ . An increase of the lower bound from  $1.5 \text{ K}$  to  $2 \text{ K}$  corresponds to a need to reduce maximum emissions by 25% to stay within the  $2^\circ \text{C}$  global warming target (because of the almost linear

relationship between warming and cumulative emissions; see Fig. SPM.10 in ref.<sup>1</sup>). This corresponds to a reduction in maximum allowable cumulative CO<sub>2</sub> emissions from 4,760 Gt CO<sub>2</sub> to 3,570 Gt CO<sub>2</sub>.

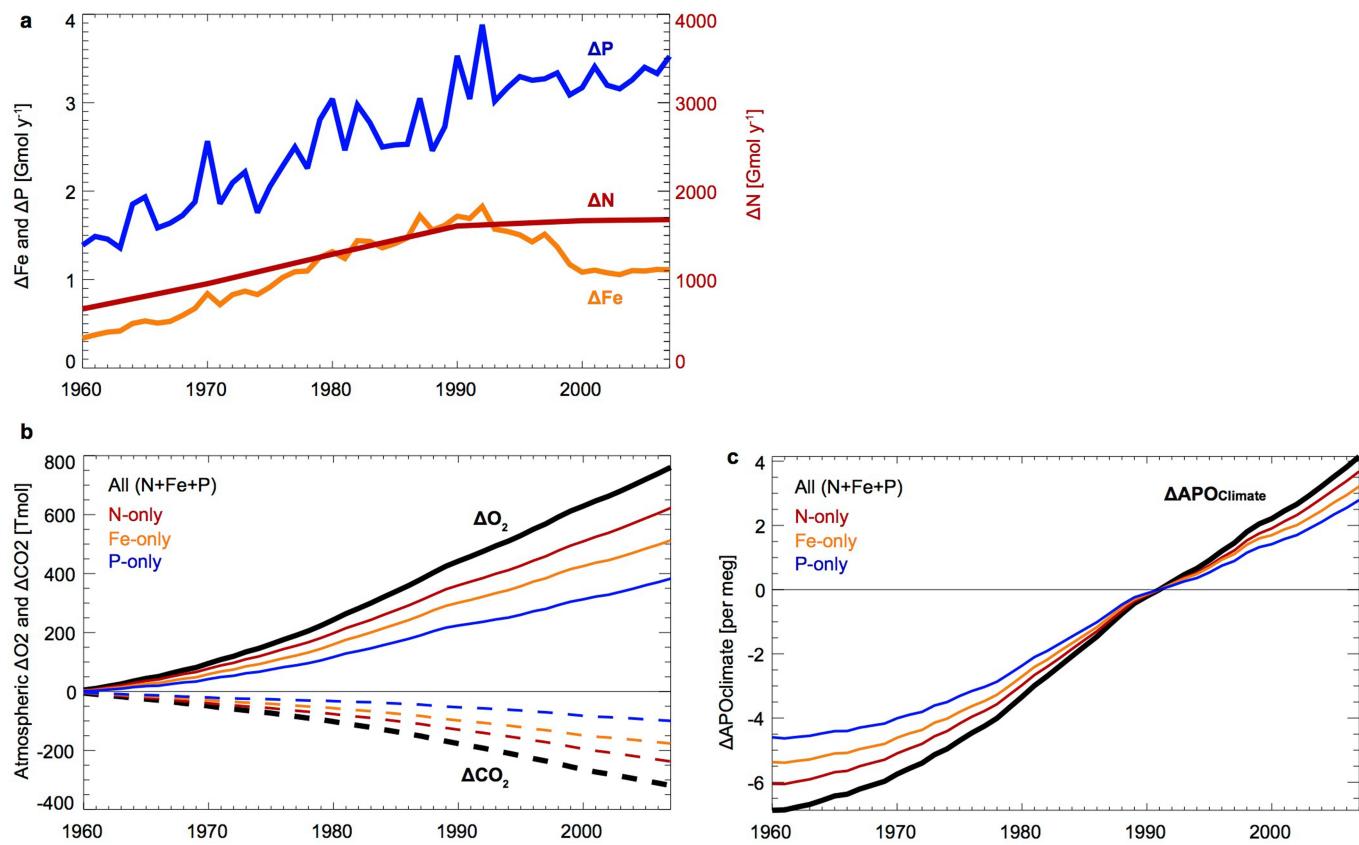
We tested the sensitivity of the climate sensitivity by using three alternate temperature datasets (NASA GISS Surface Temperature Analysis GISTEMP<sup>65</sup>, available at <https://data.giss.nasa.gov/gistemp>; the NOAA/OAR/ESRL global surface temperature data<sup>66</sup> v4.0.1, available at <https://www.esrl.noaa.gov/psd/>; and the ocean + land product of Berkeley Earth, available at [berkeleyearth.lbl.gov/auto/Global](http://berkeleyearth.lbl.gov/auto/Global); all data were accessed on 7 August 2018) as well as two preindustrial baseline periods (1860–1879 and 1880–1899). We find changes in the climate sensitivity of the order of 5% owing to the choice of temperature dataset, and less than 1% due to the choice of preindustrial baseline.

**Link to global ocean deoxygenation.** Our application of O<sub>2</sub> atmospheric measurements to constrain long-term ocean warming can be compared with earlier work that considers warming-driven oceanic O<sub>2</sub> outgassing. Multiplying our warming rate of  $1.33 \pm 0.20 \times 10^{22} \text{ J yr}^{-1}$  by the O<sub>2</sub>-to-heat ratios simulated by the four ESMs ( $-3.70 \pm 0.80 \text{ nmol O}_2 \text{ J}^{-1}$ ) yields an ocean loss of  $49 \pm 13 \text{ Tmol O}_2 \text{ yr}^{-1}$ . Adding a loss of around  $19 \pm 19 \text{ Tmol O}_2 \text{ yr}^{-1}$  due to anthropogenic aerosols (Extended Data Table 5) yields a global ocean outgassing of  $68 \pm 23 \text{ Tmol O}_2 \text{ yr}^{-1}$ , in the range of previous estimates based on atmospheric data<sup>67</sup> (about  $40 \text{ Tmol O}_2 \text{ yr}^{-1}$ ), ocean data above 1,000 m (55–65 Tmol O<sub>2</sub> yr<sup>-1</sup>, refs.<sup>68,69</sup>) and global ocean data<sup>70</sup> ( $96 \pm 42 \text{ Tmol O}_2 \text{ yr}^{-1}$ ). This calculation suggests that ocean CO<sub>2</sub> uptake is reduced by warming at a ratio of around 0.70 nmol of CO<sub>2</sub> per joule (the difference between the O<sub>2</sub>-to-heat ratio of  $3.70 \text{ nmol J}^{-1}$  and the OPO-to-heat ratio of  $4.45 \text{ nmol J}^{-1}$ ). **Code availability.** ESM codes are available online for IPSL-CM5A-LR ([cmc.ipsl.fr/ipsl-climate-models](http://cmc.ipsl.fr/ipsl-climate-models)), GFDL-ESM2M ([mdl-mom5.herokuapp.com/web/docs/project/quickstart](https://mdl-mom5.herokuapp.com/web/docs/project/quickstart)), UVic ([climate.uvic.ca/model](http://climate.uvic.ca/model)) and CESM ([www.cesm.ucar.edu/models/](http://www.cesm.ucar.edu/models/)).

## Data availability

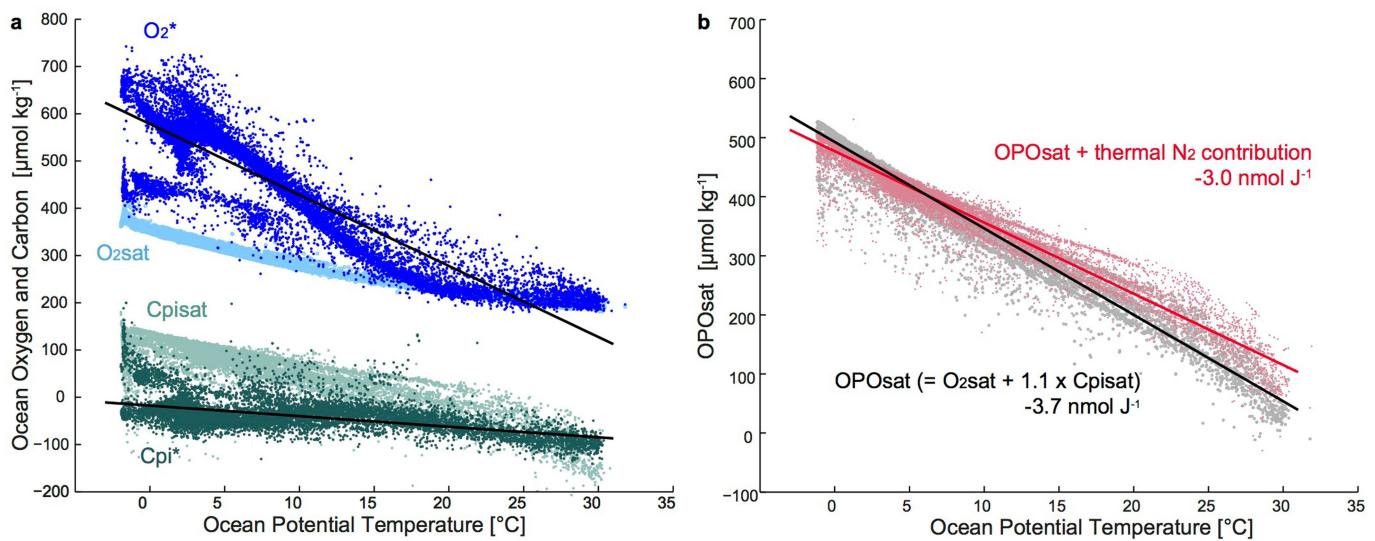
Scripps APO data are available at <http://scrippso2.ucsd.edu/apo-data>. APO<sub>Climate</sub> data, contributions to APO<sub>OBS</sub> and ocean heat content time series are available in Extended Data Figs. 1–4 and Extended Data Tables 1–5. Model results are available upon reasonable request to R.W. (IPSL anthropogenic aerosol simulations), L.B. (IPSL-CM5A-LR), M.C.L. (CESM-LE), J.P.D. (GFDL-ESM2M) or W.K. (UVic).

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**Extended Data Fig. 1 | Effects of anthropogenic aerosols on APO.**

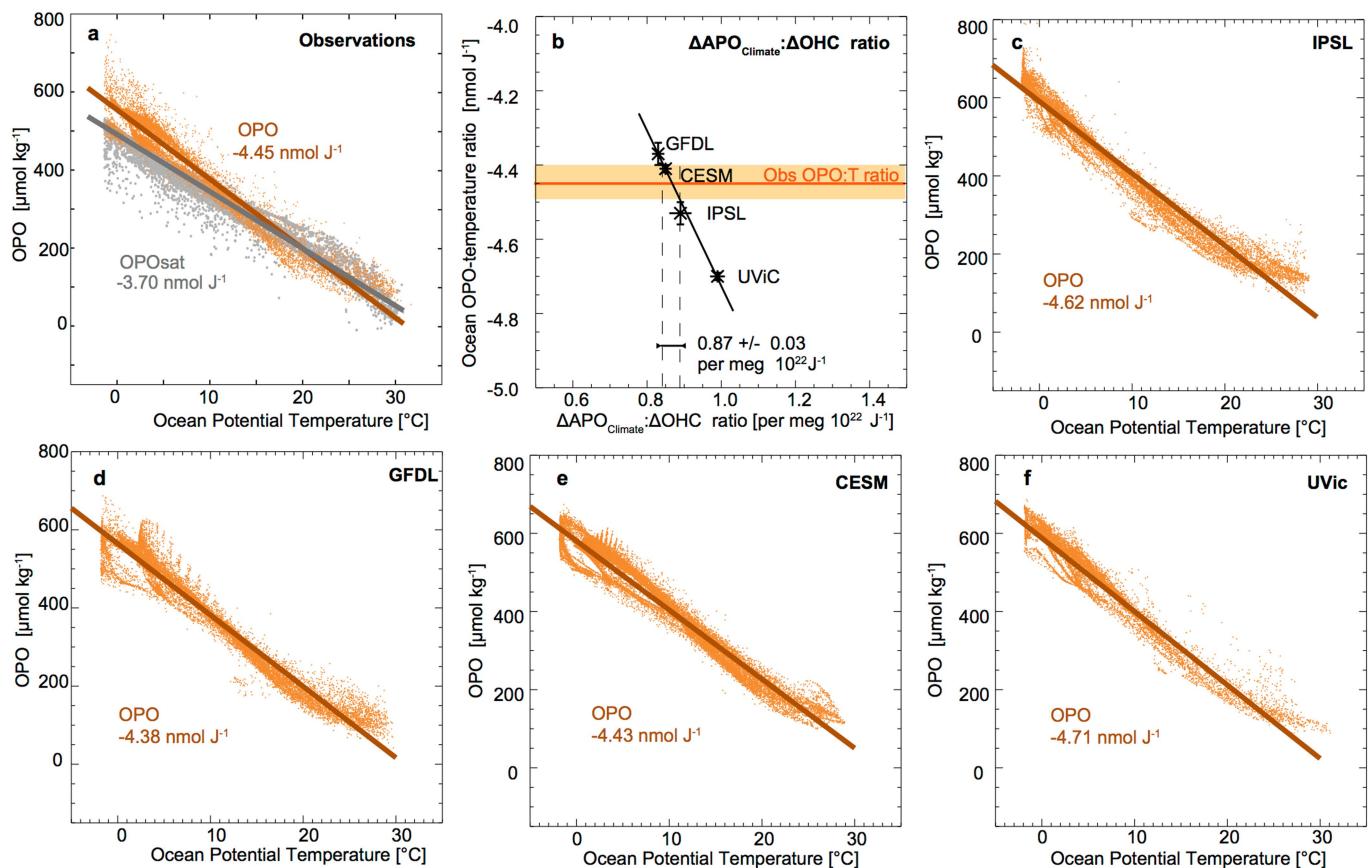
a, Anomaly, relative to 1850 levels, in deposition of atmospheric anthropogenic aerosols (N, P and Fe) at the air-sea interface between 1960 and 2007, derived from model simulations with and without aerosols<sup>22</sup>.

b, Impact of aerosol eutrophication on atmospheric  $\text{O}_2$  (solid lines) and  $\text{CO}_2$  (dashed lines) for all aerosols (black lines) and for each aerosol taken individually (coloured lines). c, Overall impact of aerosol eutrophication on  $\Delta\text{APO}_{\text{Climate}}$  referenced to the first year that has observations (1991).



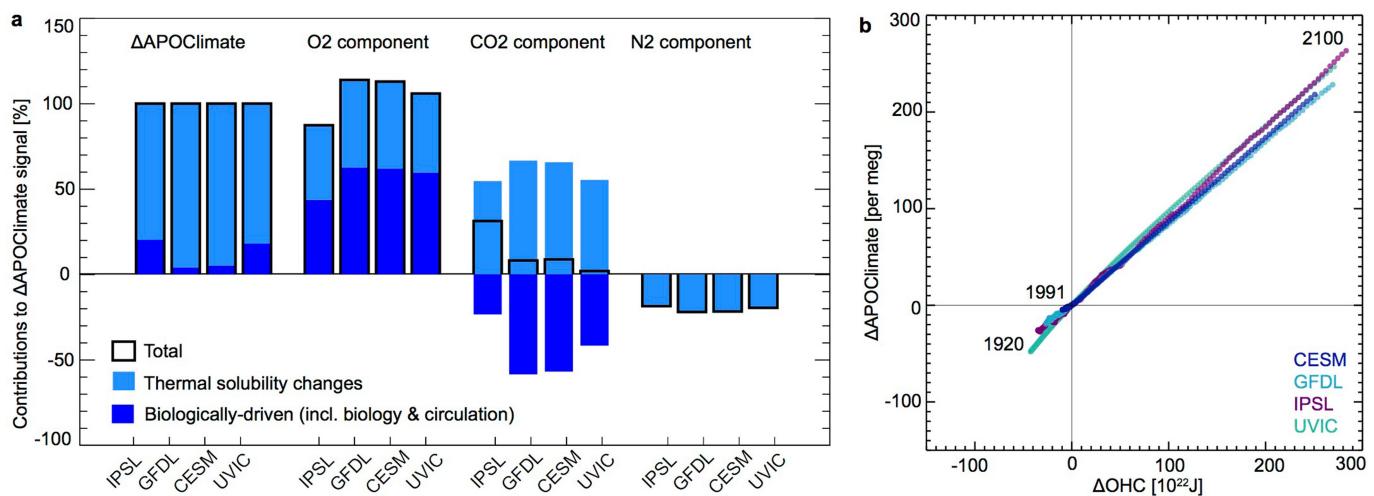
**Extended Data Fig. 2 | Solubility-driven changes in ocean oxygen and carbon concentrations.** **a**, Ocean observations of  $\text{O}_2^*$ ,  $\text{O}_{2\text{sat}}$ ,  $\text{C}_{\text{pi}}^*$  and  $\text{C}_{\text{pisat}}$  as a function of potential temperature in the Glodapv2 database<sup>32</sup>. **b**, OPO<sub>sat</sub> ( $= \text{O}_{2\text{sat}} + 1.1 \text{C}_{\text{pisat}}$ , in grey) and the expected effects on APO owing to the combined effects of OPO<sub>sat</sub> and the thermal exchanges of  $\text{N}_2$  ( $= \text{O}_{2\text{sat}} + 1.1 \text{C}_{\text{pisat}} - X_{\text{O}_2} / X_{\text{N}_2} [\text{N}_2 - \text{mean}(\text{N}_2)]$ , in red). For clarity only  $16 \times 10^3$  points randomly picked out of the 78,456 data points available

are shown for each variable. Note that very low values of  $\text{O}_2^*$  (around  $450 \mu\text{mol kg}^{-1}$ ) at low temperature (less than  $10^{\circ}\text{C}$ ) correspond to data collected in the Arctic Ocean, where phosphate concentrations (used for  $\text{O}_2^*$  calculation) are comparatively lower than in other cold ocean regions. Low  $\text{O}_2^*$  values in the Arctic explain the relatively low values of OPO shown in Extended Data Fig. 3a at temperatures below  $10^{\circ}\text{C}$ .



**Extended Data Fig. 3 | Link between OPO, APO<sub>Climate</sub> and ocean heat.** **a, c–f**, OPO concentrations (yellow) and OPO concentrations at saturation based on O<sub>2</sub> and CO<sub>2</sub> solubility (OPO<sub>sat</sub>, grey) as a function of ocean temperature in the GLODAPv2 database<sup>32</sup> (**a**) and four Earth-system models (IPSL, GFDL, CESM and UVic; **c–f**). Slopes give the OPO-to-temperature ratios in nmol J<sup>-1</sup>. **b**, The link between ΔAPO<sub>Climate</sub> and

changes in ocean heat content (that is, ΔAPO<sub>Climate</sub>-to-ΔOHC ratio) in the four models is tied to their OPO-to-temperature ratios and can be constrained using the observed OPO-to-temperature of 4.45 nmol J<sup>-1</sup> (vertical dashed lines). To avoid visual saturation, only 16,000 points, picked randomly, are shown for OPO.



**Extended Data Fig. 4 | Changes in APO<sub>Climate</sub> ( $\Delta\text{APO}_{\text{Climate}}$ ) and ocean heat content ( $\Delta\text{OHC}$ ) in four Earth-system models.** **a**, Simulated  $\Delta\text{APO}_{\text{Climate}}$  (black outlines) are decomposed into the contributions (percentage of total) from changes in ocean thermal saturation (light blue) and biologically driven changes (dark blue), the latter including changes in photosynthesis/respiration and changes in ocean circulation that transport and mix gradients of biological origin. For each model,  $\Delta\text{APO}_{\text{Climate}}$  is

further decomposed into its O<sub>2</sub>, CO<sub>2</sub> and N<sub>2</sub> components—that is, how much of  $\Delta\text{APO}_{\text{Climate}}$  is explained by changes in O<sub>2</sub>, CO<sub>2</sub> and N<sub>2</sub> air-sea fluxes due to ocean saturation changes and biologically driven changes.

**b**, Model  $\Delta\text{APO}_{\text{Climate}}$ -to- $\Delta\text{OHC}$  ratios over the 180 years of simulation (referenced to year 1991) in per meg per  $10^{22}\text{J}$  units are:  $0.85 \pm 0.01$  (CESM),  $0.83 \pm 0.01$  (GFDL),  $0.89 \pm 0.03$  (IPSL) and  $0.99 \pm 0.02$  (UVic).

**Extended Data Table 1 | Sources of the hydrographic databased estimates of global changes in ocean heat content ( $\Delta\text{OHC}$ ) used in Fig. 1**

Label in Fig 1	0 to 2000 m depth range	2000 to 6000 m depth range
PMEL	Ref. 10	Ref. 11
MRI	Ref. 9	Ref. 11
NCEI	Update of Ref. 31	Ref. 11
CHEN	Ref. 12	Ref. 11

The estimates are taken from refs 9–12.

**Extended Data Table 2 | Linear trends in global ocean heat content**

	<b>1991-2016</b> ΔOHC trend ( $\pm 1\sigma$ )	<b>1993-2016</b> ΔOHC trend ( $\pm 1\sigma$ )	<b>2007-2016</b> ΔOHC trend ( $\pm 1\sigma$ )
APO <sub>Climate</sub>	$1.33 \pm 0.20$	-	-
PMEL	-	$1.35 \pm 0.10$	$1.16 \pm 0.20$
MRI	$1.00 \pm 0.11$	$1.03 \pm 0.12$	$1.23 \pm 0.22$
NCEI	$0.89 \pm 0.08$	$0.90 \pm 0.09$	$1.28 \pm 0.16$
CHEN	$1.07 \pm 0.07$	$1.10 \pm 0.08$	$1.09 \pm 0.10$

Units are  $10^{22} \text{ J yr}^{-1}$ . Trends and  $\pm 1\sigma$  uncertainty ranges are given for hydrographic (in situ temperature) and atmospheric (APO) data over the depth range 0–6,000 m. See Extended Data Table 1 for literature sources of estimates.

**Extended Data Table 3 | Contributions to  $\Delta\text{APO}_{\text{OBS}}$ ,  $\Delta\text{APO}_{\text{FF}}$  and  $\Delta\text{APO}_{\text{Cant}}$  and associated uncertainties ( $\pm 1\sigma$ ) during the observation period 1991–2016**

Mean value	References	1- $\sigma$ uncertainty	References
$\Delta\text{APO}_{\text{OBS}}$			
Corrosion		$\pm 0.3 \text{ per meg yr}^{-1}$	
Leakage		$\pm 0.2 \text{ per meg yr}^{-1}$	
Desorption		$\pm 0.1 \text{ per meg yr}^{-1}$	
Thermal fractionation		$\pm 2 \text{ per meg}$ ( $\pm 4$ before July 1992)	Ref. 36
Scale systematic error		2% on $\delta(\text{O}_2/\text{N}_2)$ contribution	
$\Delta\text{APO}_{\text{FF}}$			
Oxidative Ratios $R_i$			
Coal	1.17	$\pm 0.03$	
Oil	1.44	$\pm 0.03$	
Gas	1.95	$\pm 0.04$	Ref. 36
Cement	0.0	$\pm 0.00$	
Flaring	1.98	$\pm 0.07$	
Emissions $\Delta\text{CO}_2$			
Coal		$\pm 7.0\%$	
Oil	Time varying	$\pm 5.5\%$	Ref. 35
Gas		$\pm 6.5\%$	
Cement		$\pm 12\%$	
Flaring		$\pm 12\%$	
$\Delta\text{APO}_{\text{Cant}}$			
$\Delta\text{Cant}_0$	Time varying (~2 to 3 PgC $\text{yr}^{-1}$ )	Ref. 21  $\pm 1\%$ uncertainty (<0.03 PgC $\text{yr}^{-1}$ ) (atmospheric $\text{CO}_2$ history)	Ref. 21  <i>this study</i>
$\Delta\text{Cant}'$	0.05 PgC $\text{yr}^{-1}$ (0.12 per meg $\text{yr}^{-1}$ )	<i>this study</i>  $\pm 0.05 \text{ PgC yr}^{-1}$ ( $\pm 0.12 \text{ per meg yr}^{-1}$ )	<i>this study</i>

The estimates are taken from refs 20,21,35,36.

Extended Data Table 4 | Temporal evolution of the cumulative contributions to global APO changes and their  $1\sigma$  uncertainties

year	$\Delta\text{APO}_{\text{Climate}}$	$1-\sigma$	$\Delta\text{APO}_{\text{OBS}}$	$1-\sigma$	$\Delta\text{APO}_{\text{FF}}$	$1-\sigma$	$\Delta\text{APO}_{\text{Cant}}$	$1-\sigma$	$\Delta\text{APO}_{\text{AtmD}}$	$1-\sigma$
1991	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1992	1.50	4.00	-6.80	4.00	-4.00	0.30	-4.90	0.10	0.50	0.30
1993	3.90	4.10	-12.90	4.00	-8.00	0.40	-9.70	0.30	0.80	0.40
1994	3.50	2.30	-22.30	2.20	-12.10	0.50	-14.80	0.40	1.10	0.50
1995	6.50	2.50	-28.40	2.30	-16.20	0.60	-20.10	0.50	1.30	0.70
1996	5.40	2.70	-39.00	2.50	-20.40	0.70	-25.50	0.70	1.60	0.80
1997	7.50	3.00	-46.10	2.70	-24.70	0.80	-30.80	0.90	1.90	0.90
1998	11.30	3.40	-52.40	2.90	-29.00	0.90	-36.80	1.10	2.10	1.10
1999	14.90	3.70	-58.70	3.20	-33.40	1.10	-42.60	1.20	2.40	1.20
2000	10.00	4.20	-73.40	3.60	-37.90	1.20	-48.20	1.40	2.70	1.30
2001	9.60	4.50	-83.90	3.90	-42.50	1.30	-54.00	1.60	2.90	1.50
2002	15.70	4.80	-87.90	4.10	-47.10	1.40	-59.80	1.70	3.20	1.60
2003	14.00	5.40	-100.40	4.50	-51.80	1.60	-66.10	1.90	3.50	1.70
2004	15.10	5.80	-110.20	4.90	-56.70	1.70	-72.40	2.00	3.70	1.90
2005	18.40	6.30	-117.80	5.20	-61.70	1.80	-78.60	2.30	4.00	2.00
2006	19.60	6.70	-127.90	5.60	-66.80	2.00	-85.10	2.40	4.30	2.10
2007	16.60	7.30	-142.10	6.10	-71.90	2.20	-91.60	2.60	4.50	2.30
2008	16.30	7.80	-153.90	6.50	-77.10	2.40	-98.10	2.90	4.80	2.40
2009	19.40	8.20	-162.00	6.80	-82.20	2.60	-104.50	3.00	5.10	2.50
2010	19.10	8.80	-174.30	7.20	-87.40	2.80	-111.50	3.20	5.40	2.70
2011	19.70	9.30	-185.40	7.60	-92.70	3.00	-118.30	3.40	5.60	2.80
2012	20.90	9.80	-196.20	8.10	-98.10	3.20	-125.10	3.50	5.90	3.00
2013	18.80	10.50	-210.30	8.60	-103.40	3.40	-132.10	3.80	6.20	3.10
2014	22.90	11.00	-218.70	9.00	-108.70	3.60	-139.50	3.90	6.50	3.20
2015	22.90	11.50	-231.10	9.50	-114.20	3.80	-146.70	4.20	6.70	3.40
2016	23.20	12.20	-243.70	10.10	-119.70	4.00	-154.30	4.20	7.00	3.50

Units are per meg.

Extended Data Table 5 | Trends in air-sea flux of O<sub>2</sub>, CO<sub>2</sub> and APO due to anthropogenic aerosol deposition

Trends 1980 to 2007	N-only	Fe-only	P-only	All (N+Fe+P)
O <sub>2</sub> [Tmol y <sup>-1</sup> ]	15.5	12.9	9.6	19.0
CO <sub>2</sub> [Tmol y <sup>-1</sup> ]	-6.1	-4.6	-2.6	-8.3
<b>APO<sub>AtmD(O2)</sub> [per meg y<sup>-1</sup>]</b>	<b>0.42</b>	<b>0.35</b>	<b>0.26</b>	<b>0.51</b>
<b>APO<sub>AtmD(CO2)</sub> [per meg y<sup>-1</sup>]</b>	<b>-0.18</b>	<b>-0.14</b>	<b>-0.08</b>	<b>-0.24</b>
<b>APO<sub>AtmD</sub> [per meg y<sup>-1</sup>]</b>	<b>0.24</b>	<b>0.21</b>	<b>0.18</b>	<b>0.27</b>

Trends in APO due to atmospheric deposition ( $\Delta\text{APO}_{\text{AtmD}}$ ) are decomposed into contributions from the O<sub>2</sub> flux only ( $\text{APO}_{\text{AtmD}(\text{O}_2)}$ ) and the CO<sub>2</sub> flux only ( $\text{APO}_{\text{AtmD}(\text{CO}_2)}$ ). Results are from model simulations<sup>22</sup>. Anomalies in air-sea flux are positive towards the atmosphere. The total trend used in this study is in bold type.