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OPEN

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

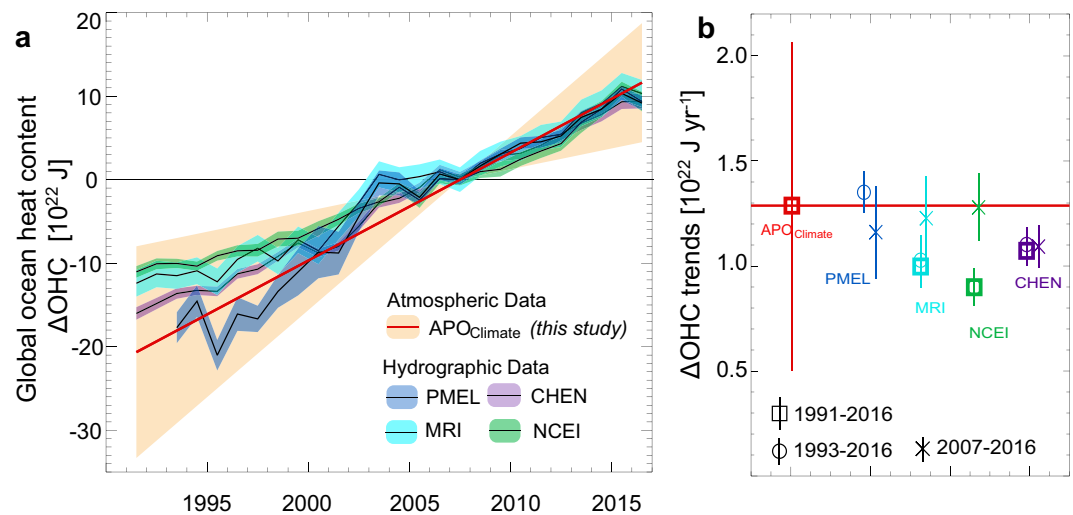
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The ocean is the main source of thermal inertia in the climate system. Ocean heat uptake during recent decades has been quantified using ocean temperature measurements. However, these estimates all use the same imperfect ocean dataset and share additional uncertainty due to sparse coverage, especially before 2007. 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  $1.29 \pm 0.79 \times 10^{22}$  Joules of heat per year between 1991 and 2016, equivalent to a planetary energy imbalance of  $0.80 \pm 0.49$  W 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> atmospheric measurements dating back to 1991 – leverages an integrative Earth system approach and provides much needed independent confirmation of heat uptake estimated from ocean data.

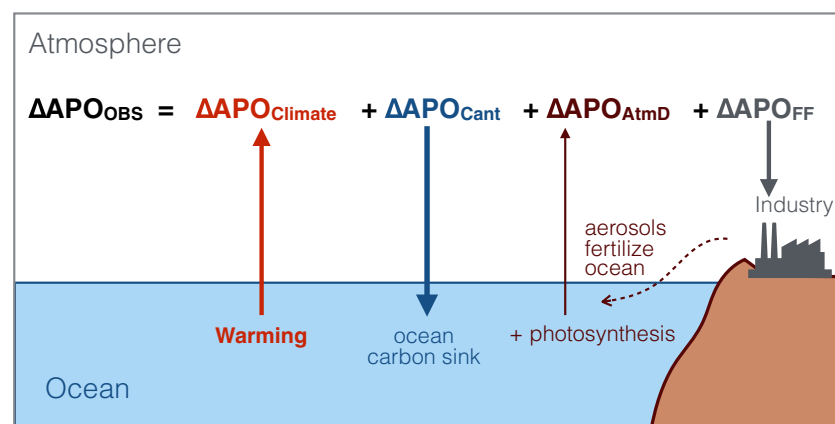
A fundamental measure of global warming is the heat uptake by the ocean, which represents more than 90% of the excess energy gained by the Earth<sup>1</sup>. This ocean warming has been quantified using hydrographic temperature measurements, including data from the Argo float program, which expanded coverage after 2007<sup>2–4</sup>. As shown in Fig. 1, the most recent temperature-based estimates of ocean warming<sup>5–8</sup> show good agreement for 2007–2016 ( $1.09 \pm 0.10$  to  $1.16 \pm 0.2 \times 10^{22}$  J yr<sup>−1</sup>), but a larger spread when extending back to include the sparser 1990s data ( $0.90 \pm 0.09$  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>5,8,9</sup>, which together introduce uncertainties up to 25–50% in warming trends<sup>10</sup>. Because temperature-based estimates use the same upper-ocean observations and linear warming trend for depths below 2000 m (ref. 6), they may share additional unknown systematic errors<sup>8</sup>. An alternative method based on the top of the atmosphere energy balance<sup>11</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>11–14</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 seawater. 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>15</sup> and glacial-interglacial<sup>16</sup> time-scales have been reconstructed using measurements of noble gases in modern or ancient air. Our method is similar, but instead of relying on noble gases (e.g. Ar/N<sub>2</sub>), which lack sufficient accuracy as yet<sup>15</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 similar to a noble gas<sup>17</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

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**Figure 1.** Change in global ocean heat content ( $\Delta\text{OHC}$ ). (a)  $\Delta\text{OHC}$  derived from hydrographic and atmospheric observations (normalized to zero in 2007,  $\pm 1-\sigma$  uncertainty). (b) Linear least-squares trends for 1991–2016, 1993–2016 and 2007–2016 ( $\pm 1-\sigma$  uncertainty). Hydrography-based  $\Delta\text{OHC}$  estimates combine warming rates at ocean depths of 0 to 2,000 m warming rates (Cheng and co-authors (CHEN)<sup>8</sup>, Pacific Marine Environmental Laboratory (PMEL)<sup>6</sup>, Meteorological Research Institute (MRI)<sup>5</sup> and National Center for Environmental Information (NCEI)<sup>47</sup> with revised deep ocean (depth of more than 2000 m) of ref. <sup>7</sup> (Tables S1, S2). The atmospheric-based estimate which uses observed atmospheric potential oxygen trends ( $\Delta\text{APO}_{\text{Climate}}$ ) and model-based  $\Delta\text{APO}_{\text{Climate}}$ -to- $\Delta\text{OHC}$  ratio, does not resolve interannual variations.



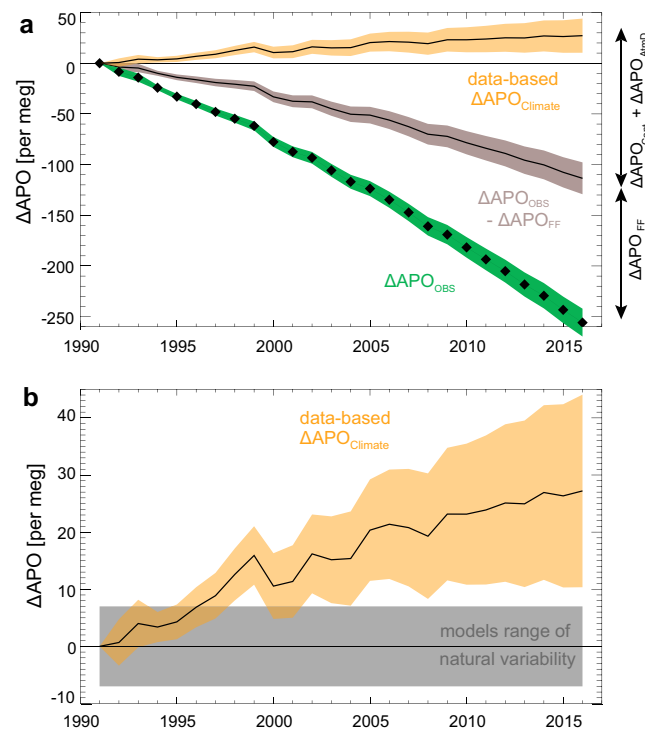
**Figure 2.** Processes contributing to observed changes in atmospheric potential oxygen ( $\Delta\text{APO}_{\text{OBS}}$ ). Industrial processes (fossil fuel burning and cement production,  $\Delta\text{APO}_{\text{FF}}$ ) and ocean sink for anthropogenic carbon ( $\Delta\text{APO}_{\text{Cant}}$ )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

the ocean can be quantified with the complementary change observed in the atmosphere. Precise atmospheric  $\text{O}_2$  measurements began in 1991 ( $\text{CO}_2$  in 1958), enabling APO-based ocean heat content reconstructions that span nearly three decades<sup>18</sup>.

## Results

**Atmospheric potential oxygen trend and components.** APO ( $=\text{O}_2 + 1.05 \times \text{CO}_2$ ) is computed using observed atmospheric  $\text{O}_2/\text{N}_2$  mole ratios and  $\text{CO}_2$  mole fractions (see Methods)<sup>18–20</sup>. By design, APO is insensitive to exchanges with land ecosystems, which produce changes in  $\text{O}_2$  and  $\text{CO}_2$  that largely cancel in APO owing to their approximate 1.05  $\text{O}_2:\text{C}$  oxidative ratio. Time-series measurements at remote sites show a global long-term decline in APO  $\Delta\text{APO}_{\text{OBS}} = 255.93 \pm 13.74$  per meg (units defined in Methods) between 1991 and 2016.  $\Delta\text{APO}_{\text{OBS}}$  is driven by four primary contributors, illustrated in Fig. 2:

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

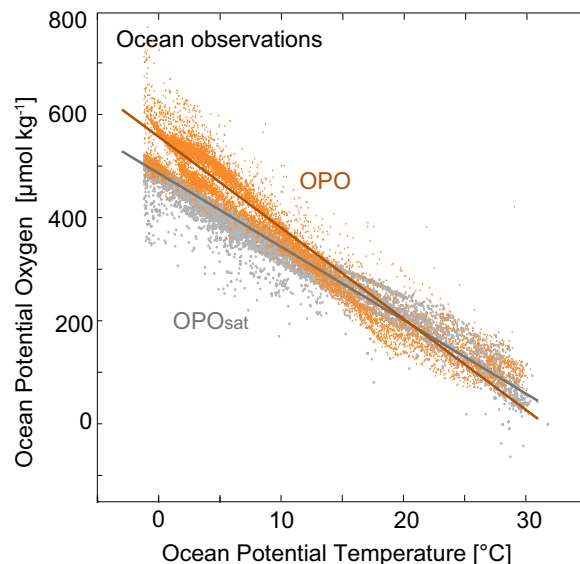


**Figure 3.** Data-based estimates of global  $\Delta\text{APO}_{\text{Climate}}$ . **(a)**  $\Delta\text{APO}_{\text{Climate}}$  estimated from observed APO ( $\Delta\text{APO}_{\text{OBS}}$ ) from the Scripps Institution of Oceanography network (1991–2016), and corrected for fossil fuel burning, ocean anthropogenic carbon uptake and anthropogenic aerosols deposition ( $\Delta\text{APO}_{\text{Climate}} = \Delta\text{APO}_{\text{OBS}} - \Delta\text{APO}_{\text{FF}} - \Delta\text{APO}_{\text{Cant}} - \Delta\text{APO}_{\text{AtmD}}$ ) and their  $1\sigma$  uncertainty ranges. **(b)** The increase in global  $\Delta\text{APO}_{\text{Climate}}$  ( $\pm 1\sigma$  interval) exceeds the range of 26-year trends expected from natural variations in four earth system models (CESM, GFDL, IPSL and UVic, shown in grey). Uncertainties and contributions to  $\Delta\text{APO}_{\text{Climate}}$  are given in Tables S3, S4 and S6.

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 1.05 moles of  $\text{O}_2$  for each mole of  $\text{CO}_2$  released;  $\Delta\text{APO}_{\text{Cant}}$  accounts for the oceanic uptake of excess anthropogenic atmospheric  $\text{CO}_2$ ;  $\text{APO}_{\text{AtmD}}$  accounts for for air-sea exchanges driven by ocean fertilization from anthropogenic aerosol deposition (increased fertilization leads to increased photosynthesis, with a concomitant release of  $\text{O}_2$  and uptake of  $\text{CO}_2$ ); and  $\Delta\text{APO}_{\text{Climate}}$  accounts for air-sea fluxes of  $\text{O}_2$ ,  $\text{CO}_2$  and  $\text{N}_2$  driven by ocean processes, including warming-induced changes in solubility, in ocean circulation, and in photosynthesis and respiration ( $\text{N}_2$  influences  $\text{O}_2/\text{N}_2$  ratios). Here, we derive  $\Delta\text{APO}_{\text{Climate}}$  from Eq. (1) and show that it tracks ocean warming.

We estimate  $\Delta\text{APO}_{\text{FF}}$  using fossil fuel and cement inventories<sup>21</sup>, finding  $\Delta\text{APO}_{\text{FF}} = -142.38 \pm 7.65$  per meg (Fig. 3).  $\Delta\text{APO}_{\text{Cant}}$  is controlled by the increase in atmospheric  $\text{CO}_2$  and by ocean mixing, which is quantified by transient tracers distributions including chlorofluorocarbons (CFCs)<sup>22</sup>, we find that  $\Delta\text{APO}_{\text{Cant}} = -147.75 \pm 3.69$  per meg.  $\Delta\text{APO}_{\text{Cant}}$  is relatively precise because it excludes the effect 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 aerosols fertilization (phosphate, iron and nitrogen; Fig. S1)<sup>23</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 Eq. (1), we therefore find  $\Delta\text{APO}_{\text{Climate}} = 27.21 \pm 16.85$  per meg, corresponding to a least-squares linear trend of  $+1.11 \pm 0.68$  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 show in Fig. 3, a clear increase in  $\Delta\text{APO}_{\text{Climate}}$  emerges over the period January 1991 to the end of December 2016.

**Linking atmospheric potential oxygen to ocean warming.** A starting point for understanding  $\Delta\text{APO}_{\text{Climate}}$  is to imagine that  $\text{O}_2$  and  $\text{CO}_2$  behave like inert gases, such that the air-sea fluxes are dominated by temperature-driven solubility changes. In this case, APO would increase by around 0.8 per meg per  $10^{22}$  J of warming, with  $\text{O}_2$  and  $\text{CO}_2$  solubility changes accounting for an increase of  $+1.0$  per meg per  $10^{22}$  J partly offset by the  $\text{N}_2$  contribution of  $-0.2$  per meg per  $10^{22}$  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>17</sup>. Observed OPO abundance is strongly tied to ocean potential temperature (Fig. 4): warming induces OPO loss and cooling induces OPO gain. The OPO-to-temperature trend of  $-4.43 \text{ nmol J}^{-1}$  is within 18% of the trend of  $-3.65$



**Figure 4.** Observed link between potential oxygen and ocean heat. OPO concentrations *in-situ* (OPO, yellow) and at saturation based on  $O_2$  and  $CO_2$  solubility ( $OPO_{sat}$ , grey) as a function of ocean temperature in the GLODAPv2 database<sup>49</sup>.

$nmol\ J^{-1}$  expected from solubility alone ( $OPO_{sat}$ -to-temperature). Biological effects (related to changes in ocean circulation and photosynthesis/respiration) on  $CO_2$  and  $O_2$  substantially cancel in OPO (Fig. S2), while thermal impacts reinforce each other, with warming waters releasing both  $O_2$  and  $CO_2$  to the atmosphere and increasing  $\Delta APO_{Climate}$ .

Further support for the dominance of solubility on  $\Delta APO_{Climate}$  is found on multidecadal timescales in four Earth system models mentioned above, which yield OPO-to-temperature ratios between  $-4.69$  and  $-4.36\ nmol\ J^{-1}$ , bracketing the ratio of  $-4.43\ nmol\ J^{-1}$  found in hydrographic observations (Fig. S3). The models also simulate a very close relationship between  $\Delta APO_{Climate}$  and the change in global ocean heat content ( $\Delta OHC$ ) that occurs during the simulations (1920–2100), with an atmospheric build-up in APO between  $0.82$  and  $0.98$  per meg per  $10^{22}\ J$  (Figs. S3 and S4) – close to the ratio expected from temperature-driven solubility changes alone ( $0.8$  per meg per  $10^{22}\ J$ ). By dividing the simulated APO change into separate biological and thermal components, we show that solubility changes account for more than 80% of  $\Delta APO_{Climate}$ , while biologically driven changes account for 5% to 20% (Fig. S4). This partitioning found in response to transient warming is very similar to the partitioning found in hydrographic data (where solubility and biology contribute 82% and 18% respectively, to the OPO-to-temperature ratio, Fig. 4).

Small differences between individual model  $\Delta APO_{Climate}$ -to- $\Delta OHC$  relationships ( $0.82$  to  $0.98$  per meg per  $10^{22}\ J$ ) 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  $\Delta APO_{Climate}$ -to- $\Delta OHC$ , Fig. S3b). Using this relationship, we find that a  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratio of  $0.86 \pm 0.03$  per meg per  $10^{22}\ J$  is compatible with observed OPO-to-temperature ratio. Combining this constrained  $\Delta APO_{Climate}$ -to- $\Delta OHC$  ratio ( $0.86 \pm 0.03$  per meg per  $10^{22}\ J$ ) with the observation-based trend in  $\Delta APO_{Climate}$  ( $1.11 \pm 0.68$  per meg/y), yields a warming trend of  $1.29 \pm 0.79 \times 10^{22}\ J\ yr^{-1}$  between 1991 and 2016. As shown in Fig. 1, this APO-based estimate of ocean heat uptake is centered near the high end of recent ocean-based estimates, while agreeing with all estimates to within the uncertainties. In our estimate, the largest single source of uncertainty is the scale error from the span calibration of the  $O_2/N_2$  analyzer (Table S6), which can be reduced via within-lab and inter-lab comparisons. The uncertainty in the estimated heat uptake since 1991 should therefore be significantly reduced in future updates.

## Discussion

Most recent ocean-based estimates of warming suggest an increase relative to prior estimates<sup>1,5,9</sup>. The independent APO estimate is fully in line with this upwards revision, although uncertainties are large. Our central value of  $\Delta OHC$  would call for a steric sea level rise of  $1.53\ mm\ y^{-1}$  (see Methods), also in agreement with satellite-derived constraints on thermal expansion, corrected for the fresh water contribution ( $1.50 \pm 0.40\ mm\ y^{-1}$ )<sup>24,25</sup>. Our result suggests that the ocean contributes  $0.80 \pm 0.49\ W/m^2$  to the Earth energy imbalance over the 1991–2016 period (Earth surface of  $5.1 \times 10^{14}\ m^2$ ), with implications for equilibrium climate sensitivity and the climate system lagged response to anthropogenic forcing<sup>26,27</sup>.

We find that the APO-heat coupling ( $\Delta APO_{Climate}$ ) is most robust on decadal and longer time-scales. Strong cancelation of biological  $O_2$  and  $CO_2$  fluxes is not expected on all temporal scales. On seasonal time-scales, air-sea  $O_2$  fluxes driven by marine photosynthesis are  $\sim 8$  times larger than those of  $CO_2$  due to slow equilibration of  $CO_2$ <sup>28</sup>. More complex coupling is also possible on interannual time-scales<sup>29</sup>, such as the weaker lagged air-sea  $CO_2$  flux compared to  $O_2$  during El Niño events<sup>30</sup>.

Atmospheric O<sub>2</sub> and CO<sub>2</sub> measurements have previously been applied to estimate global land and ocean CO<sub>2</sub> sinks, but relied on ocean heat content estimates and model-based oceanic O<sub>2</sub>-to-heat ratios to correct for climate-driven O<sub>2</sub> outgassing<sup>31–33</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 (See Methods for estimate of climate-driven ocean O<sub>2</sub> outgassing based on our results). 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 ocean and land carbon sinks using atmospheric O<sub>2</sub>.

The results presented here were previously published in *Nature*, 563, 105–108 (2018)<sup>34</sup>. The paper was subsequently retracted due to the underestimation of the uncertainties. Certain systematic errors were treated as random errors, and the uncertainty in the land oxidative ratio on the APO budget was neglected. These issues have been corrected in the current paper. The correction did not substantially change the central estimate of ocean warming but led to a roughly fourfold increase in uncertainties.

## Methods

**Observed changes in atmospheric potential oxygen ( $\Delta\text{APO}_{\text{OBS}}$ ).** A change in atmospheric potential oxygen concentration (in per meg) is defined following<sup>19</sup>:

$$\Delta(\delta\text{APO})[\text{per meg}] = \Delta(\delta\text{O}_2/\text{N}_2) + \frac{\alpha_B}{X_{\text{O}_2}} \cdot (X_{\text{CO}_2} - 350)$$

with

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

where  $\Delta(\delta\text{O}_2/\text{N}_2)$  is the atmospheric change in  $\delta\text{O}_2/\text{N}_2$  ratios (in per meg),  $X_{\text{CO}_2}$  is the CO<sub>2</sub> concentration in the air parcel (in ppm, i.e.  $\mu\text{mol mol}^{-1}$ ) and 350 is an arbitrary reference,  $\alpha_B (=1.05)$  is the approximate O<sub>2</sub>:CO<sub>2</sub> ratio of terrestrial ecosystems<sup>20</sup>, and  $X_{\text{O}_2} (=0.2094)$  is the reference value of atmospheric mole fraction of O<sub>2</sub> necessary to convert  $X_{\text{CO}_2}$  from ppm to per meg units. We use an oxidative ratio  $\alpha_B$  of 1.05 rather than 1.1, following the recommendation of ref. <sup>20</sup>, who argue that woody biomass, which has an oxidative ratio near 1.05, likely dominates the long-term carbon sources and sinks on land.

$\Delta\text{APO}_{\text{OBS}}$  is computed from *in situ* atmospheric changes in CO<sub>2</sub> concentrations and O<sub>2</sub>/N<sub>2</sub> ratios<sup>20</sup> measured at stations of the Scripps Institution of Oceanography network (available online at <http://scrippsco2.ucsd.edu>)<sup>18,35</sup>. The global average  $\Delta\text{APO}_{\text{OBS}}$  is based on a weighted average of the three stations with the longest record (1991 to 2016), i.e. La Jolla (32.9°N, 117°W), Alert (82.5°N, 62.5°W) and Cape Grim (40.5°S, 144.5°E), with weights of 0.25, 0.25 and 0.5 respectively<sup>36</sup>. Station's annual means are based on bi-monthly data fit to a four-harmonic seasonal cycle and a stiff long-term trend<sup>18</sup>. The uncertainty on  $\Delta\text{APO}_{\text{OBS}}$  was computed by generating 10<sup>6</sup> time-series with a noise scaled to the random and systematic errors of APO data detailed in Table S3. The uncertainty is taken as the 1-sigma interval ( $\pm 1$  standard deviation) from these 10<sup>6</sup> realizations (Fig. 3). To assess whether the weighted average of the 3 stations accurately reflects the global trend, we also compared other combinations of stations from 1999 onward when we have reliable data from 9 stations. The differences were consistently smaller than 0.05 per meg yr<sup>-1</sup> which is negligible compared to the other combined measurement uncertainties.

**Fossil fuel burning and cement production influence on APO ( $\Delta\text{APO}_{\text{FF}}$ ).**  $\Delta\text{APO}_{\text{FF}}$  is estimated using annual CO<sub>2</sub> emissions from oil, coal, gas, flaring and cement production ( $\Delta\text{CO}_{2(i)}$  in moles)<sup>21</sup> weighted by their O<sub>2</sub>:C combustion ratios  $R_i$ <sup>18</sup>:

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

where  $M_{\text{air}}$  is the number of moles of dry air in the atmosphere (convert moles of CO<sub>2</sub> to ppm).

The uncertainty on  $\Delta\text{APO}_{\text{FF}}$  includes uncertainties in CO<sub>2</sub> emissions ( $\Delta\text{CO}_{2(i)}$ )<sup>37</sup> and in combustion ratios ( $R_i$  in Tab S3)<sup>18</sup>. Uncertainties in  $\Delta\text{CO}_{2(i)}$  and  $R_i$  are considered not independent in time and were estimated using an autoregressive model with a correlation time-scale of 20 years following the method of ref. <sup>38</sup> (1000 realizations using Monte-Carlo approach). The uncertainty on  $\Delta\text{APO}_{\text{FF}}$  was then estimated by combining the 1000 realizations of  $\Delta\text{CO}_{2(i)}$  and the 1000 realizations of  $R_i$ , yielding a set of 10<sup>6</sup> estimates of  $\Delta\text{APO}_{\text{FF}}$ .

**Ocean anthropogenic carbon uptake influence on APO ( $\Delta\text{APO}_{\text{Cant}}$ ).** We can divide the ocean CO<sub>2</sub> uptake ( $\Delta\text{CO}_2$ ) as the sum of three contributions:

$$\Delta\text{CO}_2 = \Delta\text{Cant}_0 + \Delta\text{Cant}' + \Delta\text{CO}_{2\text{Climate}} \text{ (S1)}$$

where  $\Delta\text{Cant}_0$  is the flux driven by the rise in CO<sub>2</sub> 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, i.e. 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 CO<sub>2</sub> ( $\Delta\text{Cant}'$  is positive, i.e. 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  $\Delta\text{Cant}'$ :



$$\Delta\text{APO}_{\text{Cant}}[\text{per meg}] = \frac{\alpha_B}{X_{\text{O}_2} \times M_{\text{air}}} \times (\Delta\text{Cant}_0 + \Delta\text{Cant}') \quad (1)$$

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 the recent ocean inversion scheme with assimilation of observed potential temperature, salinity, radiocarbon, and CFC-11 of ref. <sup>22</sup> updated to 2016.  $\Delta\text{Cant}'$  cannot be derived from observations and was estimated to be  $0.05 \text{ PgC yr}^{-1}$ , equivalent to a trend of  $+0.11 \text{ per meg}^{-1}$ , using model simulations (see details in section Model-based  $\Delta\text{Cant}'$  below).

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>22</sup> using the 10 sensitivity experiments (on ocean vertical and isopycnal diffusivities, data constraint, gas exchange coefficient etc.) available for the ocean inversion and an estimate of the interannual variability in the ocean sink of a  $0.2 \text{ PgC yr}^{-1}$ . We also allow an additional 1% uncertainty ( $<0.03 \text{ PgC/yr}$ ) in  $\Delta\text{Cant}_0$  due to imperfectly known atmospheric  $\text{CO}_2$  history<sup>39</sup>, taking account of sensitivity to start date (1765 vs 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>40</sup>, and  $\text{CO}_2$  data from ref. <sup>41</sup> and the Scripps  $\text{CO}_2$  program (<https://doi.org/10.6075/J0542KSG>). Uncertainties on  $\Delta\text{Cant}'$  are assumed to be 100% of the model-based estimate of  $\Delta\text{Cant}'$  (see details in section Model-based  $\Delta\text{Cant}'$  below).

### Ocean fertilization and atmospheric deposition of anthropogenic aerosol ( $\Delta\text{APO}_{\text{AtmD}}$ ).

Deposition of anthropogenic aerosol from fossil fuel, biomass burning etc. fertilizes the ocean with nutrients, increases surface photosynthesis and sub-surface respiration<sup>42–44</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 time-scales of several 100 years<sup>45</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<sup>46</sup> using the difference between simulations with aerosols and a simulation in which the aerosol deposition is fixed to a constant pre-industrial value (equivalent to year 1850, Fig. S1)<sup>23</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 3 others only include their individual contribution (N-only, Fe-only or P-only, Fig. S1 and Table S5). Uncertainties at 1-sigma level on  $\Delta\text{APO}_{\text{AtmD}}$  are assumed to be  $\pm 50\%$ . See Table S4.

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 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 entire 1960–2007 simulation period, Fig. S1 and Table S5). The overall impact is  $\Delta\text{APO}_{\text{AtmD}} = +0.28 \text{ per meg yr}^{-1}$  over 27 years of simulation (1980 to 2007), which we extrapolate to our 1991 to 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  $-0.23 \text{ per meg yr}^{-1}$  ( $\text{APO}_{\text{AtmD}(\text{O}_2)}$  and  $\text{APO}_{\text{AtmD}(\text{CO}_2)}$  in Table S3).

The overall effect of N, Fe and P is smaller than the sum of the individual effects (Fig. S1), 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 are limiting (for example the Southern Ocean) (see Fig. 2 in ref. <sup>23</sup>).

To our knowledge this is the first estimate of the impact 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>18</sup> used anthropogenic aerosol N inventories and scaling arguments to estimate an ocean  $\text{O}_2$  loss due to anthropogenic N-deposition only of  $\sim 10 \pm 10 \text{ Tmol yr}^{-1}$ , slightly weaker 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

$$\Delta\text{APO}_{\text{Climate}} = \Delta\text{APO}_{\text{OBS}} - \Delta\text{APO}_{\text{FF}} - \Delta\text{APO}_{\text{Cant}} - \Delta\text{APO}_{\text{AtmD}} \quad (2)$$

We combine the estimates of  $\Delta\text{APO}_{\text{FF}}$ ,  $\Delta\text{APO}_{\text{Cant}}$  and  $\Delta\text{APO}_{\text{AtmD}}$  plus estimates of the contribution from variations in the oxidative ratio 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  $\Delta\text{APO}_{\text{Climate}}$  trend and its uncertainty based on the distribution of the unweighted least square fits to each of the  $10^6$  ensemble realizations of  $\Delta\text{APO}_{\text{Climate}}$  generated by combining all sources of uncertainty. We find a  $\Delta\text{APO}_{\text{Climate}}$  trend of  $1.11 \pm 0.68 \text{ per meg yr}^{-1}$  for 1991–2016. The individual contributions to the uncertainty are shown in Table S6.

**Hydrography-based estimates of ocean heat uptake ( $\Delta\text{OHC}$ ).** We used four global-ocean estimates of  $\Delta\text{OHC}$  based on hydrographic measurements in Fig. 1. Surface to 2000 m warming rates are from ref. <sup>6</sup> (PMEL), ref. <sup>5</sup> (MRI, [climate.mri-jma.go.jp/pub/ocean/ts/v7.2/](http://climate.mri-jma.go.jp/pub/ocean/ts/v7.2/)), an updated version of ref. <sup>47</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>8</sup> (CHEN, [http://159.226.119.60/cheng/images\\_files/TOA\\_OHC\\_errorbar\\_1940\\_2015\\_2.txt](http://159.226.119.60/cheng/images_files/TOA_OHC_errorbar_1940_2015_2.txt)), with the revised deep ocean (depth  $>2000 \text{ m}$ ) constant linear warming rate of  $0.10 \pm 0.03 \times 10^{22} \text{ J yr}^{-1}$  of ref. <sup>7</sup> based on the global ship-based sections program (GO-SHIP; <http://www.go-ship.org>)<sup>48</sup>.

**Ocean observations of ocean potential oxygen (OPO).** We used *in-situ* ocean observations from GLODAPv2<sup>49</sup> combined with an anthropogenic carbon estimate<sup>22</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 Oceanic Potential Oxygen (OPO)<sup>17</sup> via

$$\text{OPO} = \text{O}_2 + \alpha_B \times C_{\text{pi}}^*$$

where  $\text{O}_2^*$  and  $C_{\text{pi}}^*$  are the ocean conservative tracers related to air-sea fluxes of  $\text{O}_2$  and pre-industrial carbon<sup>50</sup>, and  $\alpha_B$  is the terrestrial oxidative ratio ( $\alpha_B = 1.05$ ). The thermal component (solubility-driven) of OPO ( $\text{OPO}_{\text{sat}}$ ) is computed as:

$$\text{OPO}_{\text{sat}} = \text{O}_{2\text{sat}} + \alpha_B \times C_{\text{pisat}}$$

where  $\text{O}_{2\text{sat}}$  is the dissolved  $\text{O}_2$  concentration at saturation with the observed temperature and salinity<sup>51</sup> and  $C_{\text{pisat}}$  is the dissolved inorganic carbon concentration expected at the observed temperature and salinity, and assuming equilibrium with a pre-industrial partial pressure of  $\text{CO}_2$  of 280 ppm and using pre-formed alkalinity<sup>52</sup>.

**Solubility-driven changes in oceanic and atmospheric potential oxygen.** Figure S2 shows a tight and quasi-linear link between observed OPO and potential temperature ( $-4.43 \text{ nmol/J}$ ,  $r^2 = 0.95$ ), similar to the link found between  $\text{OPO}_{\text{sat}}$  and potential temperature ( $-3.65 \text{ nmol/J}$ ,  $r^2 = 0.93$ ). This suggests that changes in OPO and hence  $\Delta\text{APO}_{\text{Climate}}$  are driven primarily by changes in thermal air-sea fluxes. In these observations, departures of dissolved oxygen and carbon concentrations ( $\text{O}_2^*$  and  $C_{\text{pi}}^*$ ) from their respective saturation curves ( $\text{O}_{2\text{sat}}$  and  $C_{\text{pisat}}$ ) due to biological activity tend to balance (Fig. S2). By contrast, thermal effects reinforce each other ( $\text{O}_{2\text{sat}}$  and  $C_{\text{pisat}}$  both decrease with increasing temperature) and biological effects compensate each other ( $\text{O}_2^* > \text{O}_{2\text{sat}}$  and  $C_{\text{pi}}^* < C_{\text{pisat}}$ ).

Changes in APO expected from changes in gas solubility in the ocean is an increase of 2.95 nmol per J of warming, which includes the outgassing of  $\text{O}_2$  and  $\text{CO}_2$  following  $\text{OPO}_{\text{sat}}$  (3.65 nmol/J) and the release of  $\text{N}_2$  (0.6 nmol/J) (Fig. S2b). A change of 2.95 nmol per J of warming is equivalent to an increase of 0.8 per meg/ $10^{22}$  J of APO in the atmosphere ( $= (2.95 \times 10^{-9}) / (3.7 \times 10^{19}) \times 10^{22} = 1.0 \times 10^{-6} = 0.8 \text{ per meg per } 10^{22} \text{ J}$ , with  $3.7 \times 10^{19}$  the number of moles of  $\text{O}_2$  in the atmosphere).  $\text{O}_2$  and  $\text{CO}_2$  solubility alone yield an increase in APO of 1.2 per meg/ $10^{22}$  J, which is partly counterbalanced by the outgassing of  $\text{N}_2$  that decreases APO by 0.2 per meg/ $10^{22}$  J (via the increase of the  $\text{O}_2/\text{N}_2$  ratio).

**Earth system model experiments.** We used 4 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>53,54</sup>, the Institut Pierre-Simon Laplace Coupled Model 5 version IPSL-CM5A-LR (IPSL here)<sup>55</sup>, the Community Earth System Model large ensemble CESM-LE (CESM here)<sup>56</sup> and the UVic model version 2.9 (UVic here)<sup>57</sup>. Evaluation of these models and their biogeochemical components can be found in previous studies<sup>54,56,58–60</sup>. GFDL, IPSL and UVic participated in the Coupled Model Intercomparison Project Phase 5 (CMIP5)<sup>61</sup>.

For GFDL, IPSL and UVic, we used the CMIP5 business as usual “historical-RCP8.5” scenario, the feedback experiment “esmFdbk3” that only includes warming-driven changes associated with anthropogenic emissions (e.g. radiation effects) and the fixed-climate experiment “esmFixClim3” that only includes the direct biogeochemical effects of increasing atmospheric  $\text{CO}_2$  (e.g. uptake of anthropogenic carbon, acidification etc.). For CESM, we also used the historical and RCP8.5 experiments 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  $\text{CO}_2$  on the air-sea  $\text{O}_2$  and  $\text{CO}_2$  exchanges, whereas the natural carbon tracer separation approach used for CESM still includes the biogeochemical impacts of increasing atmospheric  $\text{CO}_2$  on the carbon cycle (e.g. acidification) even while it excludes the anthropogenic carbon itself. However, we expect this effect to be small and negligible on our results.

We also used the multicentury preindustrial control simulation “piControl” with no increase in atmospheric  $\text{CO}_2$  to correct for model drift and to estimate the natural internal variability of  $\Delta\text{APO}_{\text{Climate}}$  (Fig. 2). We used model results over the 1920–2100 period, which were available for the four models.

Model OPO was computed as in the observations. Note that for CESM we removed subsurface regions of high denitrification in the eastern equatorial Pacific and Bay of Bengal where oxygen and  $\text{O}_2^*$  in this model have unrealistic values<sup>62</sup>.

**Model anthropogenic  $\Delta\text{Cant}'$ .** The component  $\Delta\text{Cant}'$  was derived from Eq. (S1) ( $\Delta\text{Cant}' = \Delta\text{CO}_2 - \Delta\text{Cant}_0 - \Delta\text{CO}_{2\text{Climate}}$ ) using CMIP5 model simulations.  $\Delta\text{CO}_2$  was taken from experiment RCP8.5,  $\Delta\text{Cant}_0$  from experiment esmFixClim3, and  $\Delta\text{CO}_{2\text{Climate}}$  from experiment esmFdbk3. Note that the control simulation was also used to correct model drift. We estimated  $\Delta\text{Cant}' = 0.05 \pm 0.05 \text{ PgC yr}^{-1}$  for the 1991 to 2016, based on the results of the three models, which individually yields  $\Delta\text{Cant}'$  of 0.0 PgC  $\text{yr}^{-1}$  (IPSL), 0.11 PgC  $\text{yr}^{-1}$  (GFDL) and 0.11 PgC  $\text{yr}^{-1}$  (UVic), and assuming an uncertainty of  $\pm 100\%$ . This corresponds to a trend of  $0.11 \pm 0.11 \text{ per meg yr}^{-1}$ .

**Model  $\Delta\text{APO}_{\text{Climate}}$  to  $\Delta\text{OHC}$  ratios and uncertainty.** Model  $\Delta\text{APO}_{\text{Climate}}$  is computed using individual contributions from  $\text{O}_2$ ,  $\text{CO}_2$  and  $\text{N}_2$  via:



$$\Delta \text{APO}_{\text{Climate}}[\text{per meg}] = \Delta \text{APO}_{(\text{O}_2)} + \Delta \text{APO}_{(\text{CO}_2)} + \Delta \text{APO}_{(\text{N}_2)}$$

$$\Delta \text{APO}_{\text{Climate}}[\text{per meg}] = \frac{1}{X_{\text{O}_2} M_{\text{air}}} \times \left( \Delta F_{\text{O}_2} + \alpha_B \times \Delta F_{\text{CO}_2} - \frac{X_{\text{O}_2}}{X_{\text{N}_2}} \times \Delta F_{\text{N}_2} \right)$$

where  $\Delta F_{\text{O}_2}$ ,  $\Delta F_{\text{CO}_2}$  and  $\Delta F_{\text{N}_2}$  are the changes in air-sea fluxes of  $\text{O}_2$ ,  $\text{CO}_2$  and  $\text{N}_2$  respectively (in moles),  $M_{\text{air}}$  is the number of moles of dry air in the atmosphere and  $X_{\text{N}_2}$  and  $X_{\text{O}_2}$  are the reference atmospheric mixing ratio of  $\text{N}_2$  and  $\text{O}_2$  respectively<sup>63</sup>.  $\text{O}_2$  and  $\text{CO}_2$  fluxes are simulated in the models.  $\text{N}_2$  air-sea fluxes, which impact the  $\text{O}_2$  atmospheric mixing ratio (because  $\text{O}_2$  is ~20% of the atmospheric composition), are quantified from the global ocean temporal changes in  $\text{N}_2$  solubility computed from model changes in temperature and salinity<sup>64</sup>.

The link between long-term changes in  $\text{APO}_{\text{Climate}}$  and ocean heat content, i.e.  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC}$  ratios, were computed for each model using the 180 years of simulations (1920 to 2100). Resulting  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC}$  ratios vary between 0.82 and 0.98 per meg per  $10^{22}$  J of warming (Fig. S3). These ratios include uncertainty in the natural climate variations at interannual and decadal time-scales and uncertainty in the  $\text{O}_2$ :C oxidative ratio associated with global gains and losses of  $\text{O}_2$  and  $\text{CO}_2$  by terrestrial ecosystems ( $\alpha_B = 1.05 \pm 0.05$ ). The uncertainty due to interannual variations was evaluated by computing  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC}$  ratios using multiple 26 year-long segments from the 180 year simulations. We obtained 616  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC}$  ratios (154 time-series of 26 years per model) and used the standard deviation between these ratios as a measure of the uncertainty.

We combine results from all models along with modeled and observed hydrographic relationships between OPO and potential temperature to establish an observationally-constrained estimate of the  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC}$  ratio, as shown in Fig. S3b. We first carry out this procedure with a fixed oxidative ratio  $\alpha_B$  of 1.05, which yields  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC} = 0.86 \pm 0.03$  per mer per  $10^{22}$  J. Repeating the same procedure using oxidative ratios  $\alpha_B$  of 1.0 and 1.1 applied to both APO and OPO yield differences in  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC}$  smaller than 0.01 per meg per  $10^{22}$  J, showing that sensitivity to the oxidative ratio is very small. Note that on Fig. S3b, the model error bars include two contributions to the uncertainties on the simulated  $\Delta \text{APO}_{\text{Climate}}$ -to- $\Delta \text{OHC}$  ratios (interannual variations and  $\text{O}_2$ :C ratio) that combine to yield  $\pm 0.01$  per meg per  $10^{22}$  J for the CESM and GFDL models,  $\pm 0.02$  per meg per  $10^{22}$  J for the UVic model and  $\pm 0.05$  per meg per  $10^{22}$  J for the IPSL model (1-sigma).

**Steric component of sea level rise.** We evaluated the steric component of sea level rise associated with a OHC of  $1.3 \times 10^{22}$  J  $\text{yr}^{-1}$  to be  $1.53 \text{ mm yr}^{-1}$ . Following ref. <sup>65</sup>, this calculation assumes that 45% of the warming occurs below 700 m and that the steric rise is 1 mm per  $0.60 \times 10^{22}$  J above 700 m and 1 mm per  $1.15 \times 10^{22}$  J below 700 m (i.e. global steric rise of 1 mm per  $0.84 \times 10^{22}$  J). Assuming that 48% of the warming occurs below 700 m (ref. <sup>6</sup>) would yield a global steric rise of 1 mm per  $0.86 \times 10^{22}$  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>66</sup>.

**Link to global ocean de-oxygenation.** Our application of  $\text{O}_2$  atmospheric measurements to constrain long-term ocean warming can be compared with earlier work considering warming-driven oceanic  $\text{O}_2$  outgassing. Multiplying our warming rate of  $1.29 \pm 0.79 \times 10^{22}$  J  $\text{yr}^{-1}$  by the  $\text{O}_2$ -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  $48 \pm 30 \text{ Tmol O}_2 \text{ yr}^{-1}$ . Adding a loss of  $\sim 19 \text{ Tmol O}_2 \text{ yr}^{-1}$  due to anthropogenic aerosols (Table S5), yields a global ocean outgassing of  $67 \pm 35 \text{ Tmol O}_2 \text{ yr}^{-1}$ , in the range of previous estimates based on atmospheric data<sup>67</sup> ( $\sim 40 \text{ Tmol O}_2 \text{ yr}^{-1}$ ), ocean data above 1000 m<sup>68,69</sup> ( $55\text{--}65 \text{ Tmol O}_2 \text{ yr}^{-1}$ ) and global ocean data<sup>70</sup> ( $96 \pm 42 \text{ Tmol O}_2 \text{ yr}^{-1}$ ). This calculation implies that ocean  $\text{CO}_2$  uptake is reduced by warming at a ratio of  $\sim 0.70 \text{ nmol of CO}_2$  per Joule (difference between  $\text{O}_2$ -to-heat ratio of  $3.70 \text{ nmol J}^{-1}$  and OPO-to-heat ratio of  $4.43 \text{ nmol J}^{-1}$ ).

### Data availability

Scripps APO data are available at [scripps2.ucsd.edu/apo-data](https://scripps2.ucsd.edu/apo-data). 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) and W. K. (UVic).

### Code availability

The code used to compute  $\text{APO}_{\text{Climate}}$  (<https://doi.org/10.5281/zenodo.2571986>) is available at <https://zenodo.org/record/2571986#.XQ1FYZNKhBw>. Earth-system model 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](http://mdl-mom5.herokuapp.com/web/docs/project/quickstart)), UVic ([climate.uvic.ca/model](http://climate.uvic.ca/model)) and CESM ([cesm.ucar.edu/models/](http://cesm.ucar.edu/models/)).

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## Author contributions

L.R. directed the analysis of the several data sets and models used here and shared responsibility for writing the manuscript; R.F.K. shared responsibility for writing the manuscript; R.W. performed the simulations of anthropogenic aerosols; L.B., J.P.D., M.C.L., W.K. and A.O. provided model results. All authors contributed to the final version of the manuscript.

## Competing interests

The authors declare no competing interests.

## Additional information

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