

Quantification of ocean heat uptake from changes in atmospheric O₂ and CO₂ composition

L. Resplandy^{1*}, R. F. Keeling², Y. Eddebbar², M. K. Brooks², R. Wang³, L. Bopp⁴, M. C. Long⁵, J. P. Dunne⁶, W. Koeve⁷ & A. Oschlies⁷

The ocean is the main source of thermal inertia in the climate system¹. 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^{2,3}. However, these estimates all use the same imperfect ocean dataset and share additional uncertainties resulting from sparse coverage, especially before 2007^{4,5}. Here we provide an independent estimate by using measurements of atmospheric oxygen (O₂) and carbon dioxide (CO₂)—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 ± 0.11 watts per square metre of Earth's surface. We also find that the ocean-warming effect that led to the outgassing of O₂ and CO₂ can be isolated from the direct effects of anthropogenic emissions and CO₂ sinks. Our result—which relies on high-precision O₂ measurements dating back to 1991⁶—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 gases⁷ and the thermal component of sea-level rise⁸.

As shown in Fig. 1, recent temperature-based hydrographic estimates of ocean warming^{9–12} 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⁻¹), 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⁻¹ for 1993–2015). The spread is mostly caused by gap-filling methods and systematic errors^{5,9}, which together introduce uncertainties of up to 25%–50% in warming trends⁴. Because temperature-based estimates also use the same upper-ocean observations and linear warming trend for depths below 2,000 m (ref. ¹¹), they may share additional unknown systematic errors¹². An alternative method based on the top of the atmosphere energy balance¹³ 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^{13–15}. 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 seasonal¹⁶ and glacial–interglacial¹⁷ 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 yet¹⁶, we rely on measurements of atmospheric O₂ and CO₂, which can be summed to yield a tracer ‘atmospheric potential oxygen’ (APO) that responds to warming similarly to a noble gas¹⁸. When the ocean warms, the solubility of O₂ and CO₂ drops, and the amount of gas lost by the ocean can be quantified with the complementary change observed in the atmosphere. Precise atmospheric

O₂ measurements began in 1991 (CO₂ in 1958), enabling APO-based reconstructions of ocean heat content that span nearly three decades⁶.

APO (O₂ + 1.1 × CO₂) is computed using observed atmospheric O₂/N₂ molar ratios and CO₂ molar fractions (see Methods)^{6,19}. By design, APO is insensitive to exchanges with land ecosystems, which produce changes in O₂ and CO₂ that largely cancel in APO owing to their approximate 1.1 O₂/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 ± 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 O₂ for each mole of CO₂ released; $\Delta\text{APO}_{\text{Cant}}$ accounts for the oceanic uptake of excess anthropogenic atmospheric CO₂; $\Delta\text{APO}_{\text{AtmD}}$ accounts for air–sea exchanges driven by ocean fertilization from anthropogenic aerosol deposition (increased fertilization leads to increased photosynthesis, with a concomitant release of O₂ and uptake of CO₂); and $\Delta\text{APO}_{\text{Climate}}$ accounts for air–sea fluxes of O₂, CO₂ and N₂ driven by ocean processes, including warming-induced changes in solubility, in ocean circulation, and in photosynthesis and respiration (N₂ influences O₂/N₂ 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 inventories²⁰, 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 CO₂ and by ocean mixing, which is quantified by the distribution of transient tracers including chlorofluorocarbons (CFCs)²¹; 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)²². $\Delta\text{APO}_{\text{AtmD}}$ 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 O₂ and CO₂ behave like inert gases, such that the air–sea fluxes are dominated by temperature-driven solubility changes. In this case,

¹Department of Geosciences and Princeton Environmental Institute, Princeton University, Princeton, NJ, USA. ²Scripps Institution of Oceanography, University of California San Diego, La Jolla, CA, USA. ³Department of Environmental Science and Engineering, Fudan University, Shanghai, China. ⁴LMD/IPSL, ENS, PSL Research University, École Polytechnique, Sorbonne Université, CNRS, Paris, France. ⁵National Center for Atmospheric Research, Boulder, CO, USA. ⁶NOAA, Geophysical Fluid Dynamics Laboratory, Princeton, NJ, USA. ⁷GEOMAR Helmholtz Centre for Ocean Research Kiel, Kiel, Germany. *e-mail: laurer@princeton.edu

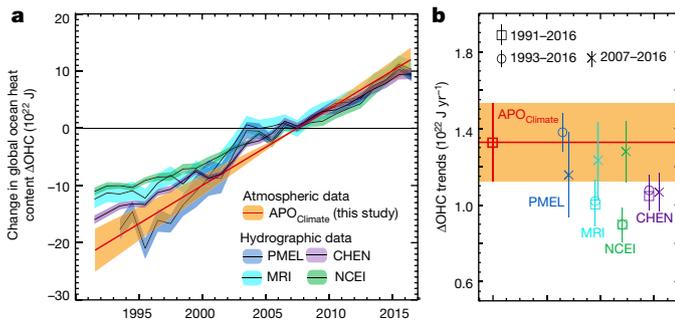


Fig. 1 | Change in global ocean heat content (ΔOHC). **a**, ΔOHC derived from hydrographic and atmospheric observations (normalized to zero in 2007, $\pm 1\sigma$ uncertainty). **b**, Linear least-squares trends for 1991–2016, 1993–2016 and 2007–2016 ($\pm 1\sigma$ uncertainty). Hydrography-based ΔOHC estimates combine warming rates at ocean depths of 0 to 2,000 m (from Cheng and co-authors (CHEN)¹², Pacific Marine Environmental Laboratory (PMEL)¹⁰, Meteorological Research Institute (MRI)⁹ and National Centers for Environmental Information (NCEI)³¹ estimates) with the revised deep ocean warming (at depths of more than 2,000 m) of ref.¹¹ (Extended Data Tables 1 and 2). The atmospheric-based estimate (this study), which uses observed atmospheric potential oxygen trends ($\Delta\text{APO}_{\text{Climate}}$) and model-based $\Delta\text{APO}_{\text{Climate}}$ -to- ΔOHC ratios, does not resolve interannual variations.

APO would increase by around 0.8 per meg per 10^{22} J of warming, with changes in O_2 and CO_2 solubility accounting for an increase of +1.0 per meg per 10^{22} J, partly offset by the 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 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 O_2 and CO_2 fluxes¹⁸. 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_{sat}-to-temperature). Biological effects (related to changes in ocean circulation and photosynthesis/respiration) on CO_2 and O_2 substantially cancel in OPO (Extended Data Fig. 2), while thermal impacts reinforce each other, with warming waters releasing both O_2 and 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 $-4.45 \text{ nmol J}^{-1}$

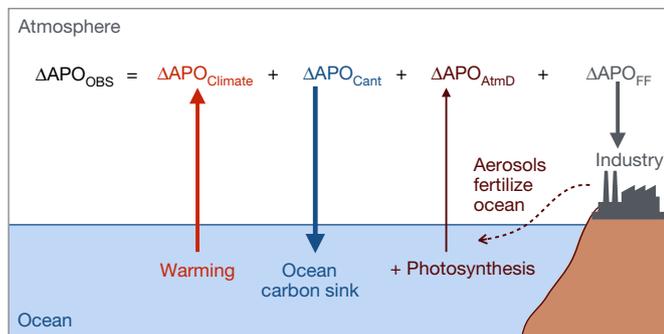


Fig. 2 | Processes contributing to observed changes in atmospheric potential oxygen ($\Delta\text{APO}_{\text{OBS}}$). Industrial processes (fossil-fuel burning and cement production; $\Delta\text{APO}_{\text{FF}}$) and the ocean sink for anthropogenic carbon ($\Delta\text{APO}_{\text{Cant}}$) remove 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.

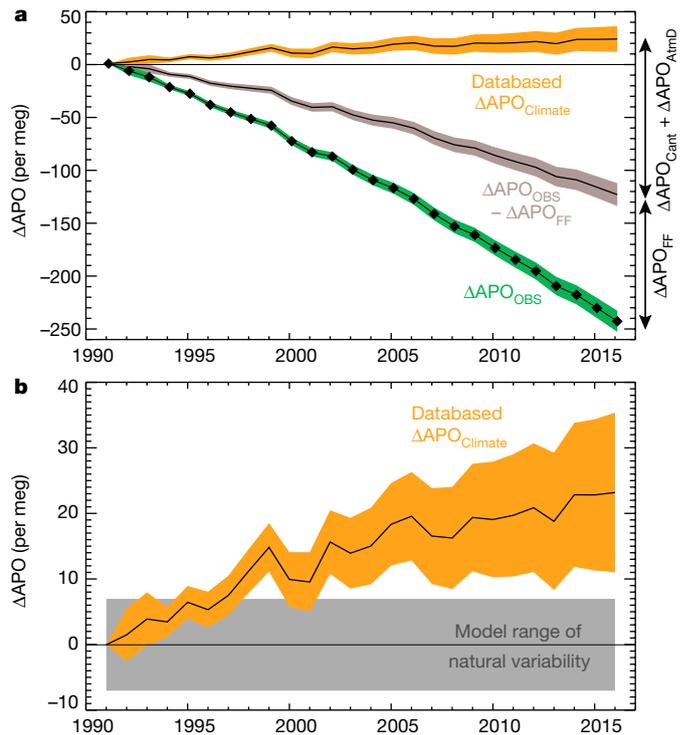


Fig. 3 | Databased estimates of global $\Delta\text{APO}_{\text{Climate}}$. **a**, $\Delta\text{APO}_{\text{Climate}}$ estimated from observed APO ($\Delta\text{APO}_{\text{OBS}}$) from the Scripps Institution of Oceanography network (1991–2016), and corrected by taking into account fossil-fuel burning, ocean anthropogenic carbon uptake and anthropogenic aerosol 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 1σ 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 the natural variations in four Earth system models (CESM, GFDL, IPSL and UVic, shown in grey). Sources of uncertainty and contributions to $\Delta\text{APO}_{\text{OBS}}$, $\Delta\text{APO}_{\text{FF}}$ and $\Delta\text{APO}_{\text{Cant}}$ are given in Extended Data Tables 3 and 4.

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 (ΔOHC) 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} J (Extended Data Figs. 3, 4)—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 $\Delta\text{APO}_{\text{Climate}}$, 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- ΔOHC relationships (0.83 to 0.99 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 $\Delta\text{APO}_{\text{Climate}}$ -to- ΔOHC ; Extended Data Fig. 3b). Using this relationship, we find that a $\Delta\text{APO}_{\text{Climate}}$ -to- ΔOHC ratio of 0.87 ± 0.03 per meg per 10^{22} J is compatible with the observed OPO-to-temperature ratio. Combining this constrained $\Delta\text{APO}_{\text{Climate}}$ -to- ΔOHC ratio (0.87 ± 0.03 per meg per 10^{22} J) with the observation-based trend in $\Delta\text{APO}_{\text{Climate}}$ (1.16 ± 0.18 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)¹⁰, available only for

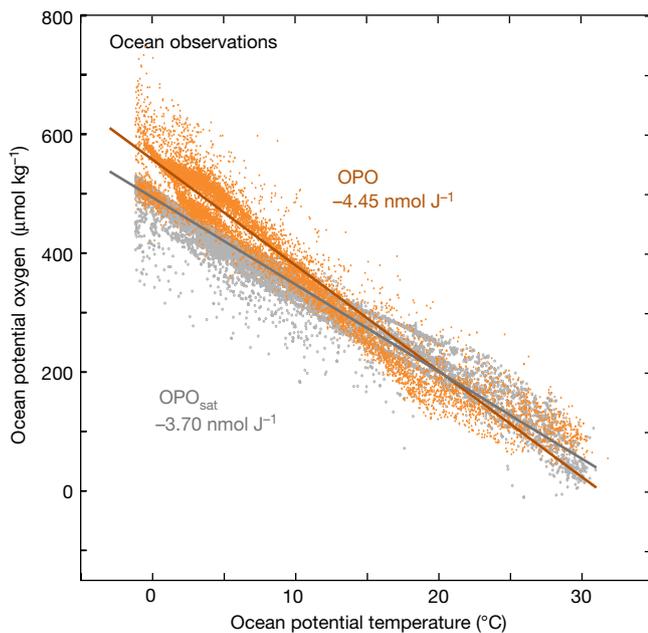


Fig. 4 | Observed link between potential oxygen and ocean heat. OPO concentrations in situ (OPO, yellow) and at saturation based on O_2 and CO_2 solubility (OPO_{sat} , grey) as a function of ocean temperature in the GLODAPv2 database³².

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

The APO data provide a much-needed independent confirmation of the recent upward revisions in estimates of ocean warming^{5,9}. A higher value of ΔOHC compatible with both $APO_{Climate}$ 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 mm yr^{-1} (Methods), in full agreement with satellite constraints on thermal expansion, corrected for the freshwater contribution ($1.50 \pm 0.40 \text{ mm yr}^{-1}$)^{8,23}.

A higher ΔOHC will also affect the equilibrium climate sensitivity, recently estimated at between $+1.5 \text{ K}$ and $+4.5 \text{ K}$ if CO_2 is doubled¹. 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)^{1,24}, 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_2 emissions by 25% to stay within the $2 \text{ }^\circ\text{C}$ global warming target (see Methods).

We find that the APO–heat coupling ($APO_{Climate}$) is most robust on decadal and longer timescales. Strong cancellation of biological O_2 and CO_2 fluxes is not expected on all temporal scales²⁵. On seasonal timescales, air–sea O_2 fluxes driven by marine photosynthesis are around eight times larger than those of CO_2 owing to slow equilibration of CO_2 (ref. ²⁵). More complex coupling is also possible on interannual timescales²⁶, such as the weaker lagged air–sea CO_2 flux compared with the O_2 flux during El Niño events²⁷.

Atmospheric O_2 and CO_2 measurements have been applied previously to estimate global land and ocean CO_2 sinks, but relied on estimates of ocean heat content and model-based oceanic O_2 -to-heat ratios to correct for climate-driven O_2 outgassing^{28–30}. 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_2 –heat relationship. Further work to constrain the separate contributions of O_2 and CO_2 to APO is needed to refine estimates of land and ocean carbon sinks using atmospheric O_2 and CO_2 .

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>.

Received: 4 December 2017; Accepted: 29 August 2018;

Published online 31 October 2018.

- IPCC. *Climate Change 2013: The Physical Science Basis* (eds Stocker, T. F. et al.) (Cambridge Univ. Press, Cambridge, 2013).
- Abraham, J. P. et al. A review of global ocean temperature observations: implications for ocean heat content estimates and climate change. *Rev. Geophys.* **51**, 450–483 (2013).
- Riser, S. C. et al. Fifteen years of ocean observations with the global Argo array. *Nat. Clim. Change* **6**, 145–153 (2016).
- Boyer, T. et al. Sensitivity of global upper-ocean heat content estimates to mapping methods, XBT bias corrections, and baseline climatologies. *J. Clim.* **29**, 4817–4842 (2016).
- Cheng, L. et al. XBT science: assessment of instrumental biases and errors. *Bull. Am. Meteorol. Soc.* **97**, 924–933 (2016).
- Keeling, R. F. & Manning, A. C. in *Treatise on Geochemistry* 385–404 (Elsevier, Oxford, 2014).
- Forster, P. M. Inference of climate sensitivity from analysis of Earth’s energy budget. *Annu. Rev. Earth Planet. Sci.* **44**, 85–106 (2016).
- Church, J. A. et al. in *Climate Change 2013: The Physical Science Basis* (eds Stocker, T. F. et al.) 1137–1216 (IPCC, Cambridge Univ. Press, Cambridge, 2013).
- Ishii, M. et al. Accuracy of global upper ocean heat content estimation expected from present observational data sets. *Sci. Online Lett. Atmos.* **13**, 163–167 (2017).
- Johnson, G. C. et al. Ocean heat content. *Am. Meteorol. Soc. Bull.* **98**, S66–S68 (2017).
- Desbruyères, D. G., Purkey, S. G., McDonagh, E. L., Johnson, G. C. & King, B. A. Deep and abyssal ocean warming from 35 years of repeat hydrography. *Geophys. Res. Lett.* **43**, 10356–10365 (2016).
- Cheng, L. et al. Improved estimates of ocean heat content from 1960 to 2015. *Sci. Adv.* **3**, e1601545 (2017).
- Allan, R. P. et al. Changes in global net radiative imbalance 1985–2012. *Geophys. Res. Lett.* **41**, 5588–5597 (2014).
- Palmer, M. D. Reconciling estimates of ocean heating and Earth’s radiation budget. *Curr. Clim. Change Rep.* **3**, 78–86 (2017).
- Loeb, N. G. et al. Observed changes in top-of-the-atmosphere radiation and upper-ocean heating consistent within uncertainty. *Nat. Geosci.* **5**, 110–113 (2012).
- Battle, M. et al. Measurements and models of the atmospheric Ar/N_2 ratio. *Geophys. Res. Lett.* **30**, 1786 (2003).
- Ritz, S. P., Stocker, T. F. & Severinghaus, J. P. Noble gases as proxies of mean ocean temperature: sensitivity studies using a climate model of reduced complexity. *Quat. Sci. Rev.* **30**, 3728–3741 (2011).
- Resplandy, L. et al. Constraints on oceanic meridional heat transport from combined measurements of oxygen and carbon. *Clim. Dyn.* **47**, 3335–3357 (2016); erratum **49**, 4317 (2017).
- Stephens, B. B. et al. Testing global ocean carbon cycle models using measurements of atmospheric O_2 and CO_2 concentration. *Glob. Biogeochem. Cycles* **12**, 213–230 (1998).
- Le Quéré, C. et al. Global carbon budget 2016. *Earth Syst. Sci. Data* **8**, 605–649 (2016).
- DeVries, T. The oceanic anthropogenic CO_2 sink: storage, air–sea fluxes, and transports over the industrial era. *Glob. Biogeochem. Cycles* **28**, 631–647 (2014).
- Wang, R. et al. Influence of anthropogenic aerosol deposition on the relationship between oceanic productivity and warming. *Geophys. Res. Lett.* **42**, 10745–10754 (2015).
- Rietbroek, R., Brunnabend, S.-E., Kutsche, J., Schröter, J. & Dahle, C. Revisiting the contemporary sea-level budget on global and regional scales. *Proc. Natl Acad. Sci. USA* **113**, 1504–1509 (2016).
- IPCC. *Climate Change 2007: Synthesis Report* (eds Core Writing Team, Pachauri, R. K. & Reisinger, A.) (IPCC, Geneva, 2008).
- Keeling, R. F. & Severinghaus, J. P. in *The Carbon Cycle* (eds Wigley, T. M. L. & Schimel, D.) 134–140 (Cambridge Univ. Press, New York, 2000).
- Resplandy, L., Séférian, R. & Bopp, L. Natural variability of CO_2 and O_2 fluxes: what can we learn from centuries-long climate models simulations? *J. Geophys. Res. Oceans* **120**, 384–404 (2015).
- Eddelbar, Y. A. et al. Impacts of ENSO on air–sea oxygen exchange: observations and mechanisms. *Glob. Biogeochem. Cycles* **31**, 2017GB005630 (2017).
- Keeling, R. F. & Garcia, H. E. The change in oceanic O_2 inventory associated with recent global warming. *Proc. Natl Acad. Sci. USA* **99**, 7848–7853 (2002).
- Bopp, L., Le Quéré, C., Heimann, M., Manning, A. C. & Monfray, P. Climate-induced oceanic oxygen fluxes: implications for the contemporary carbon budget. *Glob. Biogeochem. Cycles* **16**, 1022 (2002).
- Keeling, C. D., Piper, S. C., Whorf, T. P. & Keeling, R. F. Evolution of natural and anthropogenic fluxes of atmospheric CO_2 from 1957 to 2003. *Tellus B Chem. Phys. Meteorol.* **63**, 1–22 (2011).

31. Levitus, S. et al. World ocean heat content and thermosteric sea level change (0–2000 m), 1955–2010. *Geophys. Res. Lett.* **39**, L10603 (2012).
32. Olsen, A. et al. The Global Ocean Data Analysis Project version 2 (GLODAPv2)—an internally consistent data product for the world ocean. *Earth Syst. Sci. Data* **8**, 297–323 (2016).

Acknowledgements We thank M. Winton for useful suggestions. L.R. acknowledges support from the Climate Program Office of the National Oceanic and Atmospheric Administration (NOAA), grant NA13OAR4310219, and from the Princeton Environmental Institute. The National Center for Atmospheric Research (NCAR) is sponsored by the National Science Foundation (NSF). We also thank the people who maintain the APO time series at Scripps and the groups developing the models CESM, GFDL, IPSL and UViC, used in this study. The Scripps O₂ program has been supported by a series of grants from the US NSF and the NOAA, most recently 1304270 and NA15OAR4320071. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the authors and do not necessarily reflect the views of the NSF and NOAA. We thank the staff at the Cape Grim Baseline Air Pollution Station of the Canadian Greenhouse Gas program for collection of air samples.

Reviewer information *Nature* thanks L. Cheng, F. Primeau and the other anonymous reviewer(s) for their contribution to the peer review of this work.

Author contributions L.R. directed the analysis of the datasets and models used here and shared responsibility for writing the manuscript; R.F.K. shared responsibility for writing the manuscript; R.W. performed 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

Extended data is available for this paper at <https://doi.org/10.1038/s41586-018-0651-8>.

Reprints and permissions information is available at <http://www.nature.com/reprints>.

Correspondence and requests for materials should be addressed to L.R.

Publisher's note: Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

METHODS

Observed changes in APO. A change in atmospheric potential oxygen concentration (in per meg) is defined following¹⁹:

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

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 O_2/N_2 ratios (in per meg); X_{CO_2} is the 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 O_2/CO_2 ratio of terrestrial ecosystems³³; and X_{O_2} ($= 0.2094$) is the reference value of atmospheric mole fraction of O_2 necessary to convert X_{CO_2} from p.p.m. to per meg units.

$\Delta\text{APO}_{\text{OBS}}$ is computed from in situ atmospheric changes in CO_2 concentrations and O_2/N_2 ratios¹⁹ measured at stations of the Scripps Institution of Oceanography network (available online at <http://scrippsco2.ucsd.edu>)⁶. 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° N, 117° W), Alert (82.5° N, 62.5° W) and Cape Grim (40.5° S, 144.5° E) and weighted by the stations' latitudinal distribution³⁴. Station annual means are based on bimonthly data fit to a four-harmonic seasonal cycle and a stiff long-term trend⁶. 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 1σ interval (± 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 CO_2 emissions from oil, coal, gas, flaring and cement production ($\Delta\text{CO}_{2(i)}$ in moles)²⁰ weighted by their O_2/C combustion ratios, R_i (ref. ⁶):

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

where M_{air} is the number of moles of dry air in the atmosphere (convert moles of CO_2 to p.p.m.).

The uncertainty on $\Delta\text{APO}_{\text{FF}}$ includes uncertainties in CO_2 emissions ($\Delta\text{CO}_{2(i)}$)³⁵ and in combustion ratios (R_i in Extended Data Table 3)³⁶. Uncertainties on $\Delta\text{CO}_{2(i)}$ are not independent in time and were estimated using an autoregressive model³⁷ (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 CO_2 uptake (ΔCO_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 ΔCant_0 is the flux driven by the rise in CO_2 assuming steady ocean circulation (ΔCant_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 CO_2 ($\Delta\text{Cant}'$ is positive, that is, warming reduces the uptake of C_{ant}). $\Delta\text{APO}_{\text{Cant}}$ can be expressed as the weighted sum of the two terms ΔCant_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 ΔCant_0 and $\Delta\text{Cant}'$ are in moles. Note that $\Delta\text{CO}_{2\text{Climate}}$ is accounted for in $\Delta\text{APO}_{\text{Climate}}$.

ΔCant_0 is taken from a recent ocean inversion scheme with assimilation of observed potential temperature, salinity, radiocarbon and CFC-11 (ref. ²¹), 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 $+0.2$ 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 ΔCant_0 and $\Delta\text{Cant}'$. We allow for uncertainty in ΔCant_0 following ref. ²¹, 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 inter-annual variability in the ocean sink of a 0.2 Pg C yr^{-1} . We also allow an additional 1% uncertainty (less than $0.03 \text{ Pg C yr}^{-1}$) in ΔCant_0 resulting from imperfectly known atmospheric CO_2 history³⁸, 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 model³⁹, and CO_2 data from ref. ⁴⁰ and the Scripps 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^{41–43}. 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 years⁴⁴. 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 O_2 , CO_2 and APO air–sea fluxes is evaluated with the IPSL ocean model NEMO-PISCES v2 (ref. ⁴⁵), 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)²². 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 1σ 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 O_2 outgassing of $19.0 \text{ Tmol yr}^{-1}$ for the 1980–2007 period (16 Tmol yr^{-1} for the entire 1960–2007 simulation period) and an oceanic CO_2 uptake of 8.3 Tmol yr^{-1} for the 1980–2007 period (6.8 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 $+0.27$ per meg yr^{-1} over 27 years of simulation (1980–2007), which we extrapolate to our 1991–2016 period. Increased O_2 outgassing accounts for an increase in APO of $+0.51$ per meg yr^{-1} , and CO_2 uptake accounts for a change in APO of -0.24 per meg yr^{-1} ($\text{APO}_{\text{AtmD}(\text{O}_2)}$ and $\text{APO}_{\text{AtmD}(\text{CO}_2)}$ 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. ²²).

To our knowledge this is the first estimate of the effect of anthropogenic aerosol deposition on both O_2 and CO_2 air–sea fluxes at the global scale. Note however that ref. ⁶ used anthropogenic aerosol N inventories and scaling arguments to estimate an ocean 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 ± 0.15 per meg yr^{-1} for 1991–2016.

Hydrography-based estimates of ocean heat uptake. We used four global-ocean estimates of ΔOHC , based on hydrographic measurements, in Fig. 1. Ocean warming rates from the surface to 2,000 m are from ref. ¹⁰ (PMEL), ref. ⁹ (MRI; <https://climate.mri-jma.go.jp/pub/ocean/ts/v7.2/>), an updated version of ref. ³¹ (NCEI; www.nodc.noaa.gov/OC5/3M_HEAT_CONTENT/basin_avt_data.html) and ref. ¹² (CHEN; http://159.226.119.60/cheng/images_files/TOA_OHC_errorbar_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. ¹¹ being based on the global ship-based sections program (GO-SHIP; <http://www.go-ship.org>)⁴⁶.

Ocean observations of ocean potential oxygen. We used in situ ocean observations from GLODAPv2 (ref. ³²) combined with an anthropogenic carbon estimate²¹ interpolated at the location of each sample to compute 78,456 values (GLODAPv2 quality control = 0; marginal seas and coastal waters were removed) of 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 carbon⁴⁷. 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 salinity⁴⁸; 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 alkalinity⁴⁹.

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 nmol J^{-1} ; $r^2 = 0.95$), similar to the link found between OPO_{sat} and potential temperature (-3.7 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 nmol J^{-1}) and the release of N_2 (0.7 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 3.7×10^{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 $1.0 \text{ per meg per } 10^{22} \text{ J}$, which is partly counterbalanced by the outgassing of N_2 that decreases APO by $0.2 \text{ per meg per } 10^{22} \text{ J}$ (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)^{50,51}, the Institut Pierre-Simon Laplace Coupled Model 5 version IPSL-CM5A-LR (IPSL here)⁵², the Community Earth System Model large ensemble CESM-LE (CESM here)⁵³ and the UVic model version 2.9 (UVic here)⁵⁴. Evaluation of these models and their biogeochemical components can be found in previous studies^{51,55–57}. GFDL, IPSL and UVic participated in the Coupled Model Intercomparison Project Phase 5 (CMIP5)⁵⁸.

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 values⁵⁹.

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

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

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

where ΔF_{O_2} , ΔF_{CO_2} and ΔF_{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 respectively⁶⁰. 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 salinity⁶¹.

The link between long-term changes in $APO_{Climate}$ and ocean heat content—that is, $\Delta APO_{Climate}$ -to- ΔOHC ratios—were computed for each model using the 180 years of simulations (1920–2100). Resulting $\Delta APO_{Climate}$ -to- ΔOHC ratios vary between 0.83 and 0.99 per meg per 10^{22} 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- ΔOHC ratios using multiple 26-year-long segments from the 180-year simulations. We obtained 616 $\Delta APO_{Climate}$ -to- ΔOHC 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. 33). An oxidative ratio lower than 1.1 would yield a weaker $\Delta APO_{Climate}$ -to- ΔOHC slope and hence a slightly higher estimate of ΔOHC 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- ΔOHC ratios (interannual variations and O_2/C ratio) combine to yield $\pm 0.01 \text{ per meg per } 10^{22} \text{ J}$ for the CESM and GFDL models, $\pm 0.02 \text{ per meg per } 10^{22} \text{ J}$ for the UVic model, and $\pm 0.05 \text{ per meg per } 10^{22} \text{ J}$ for the IPSL model (1σ). 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 ΔOHC 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 mm yr^{-1} and 1.74 mm yr^{-1} . Following ref. 62, this calculation assumes that 45% of the warming occurs below 700 m, and that the steric rise is $1 \text{ mm per } 0.60 \times 10^{22} \text{ J}$ above 700 m, and $1 \text{ mm per } 1.15 \times 10^{22} \text{ J}$ below 700 m (that is, a global steric rise of $1 \text{ mm per } 0.84 \times 10^{22} \text{ J}$). Assuming that 48% of the warming occurs below 700 m (ref. 10) would yield a global steric rise of $1 \text{ mm per } 0.86 \times 10^{22} \text{ J}$ 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 Group⁶³.

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. 1). 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. 1):

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

where N is the global heat imbalance, which mostly consists of the ocean heat uptake; F is the radiative forcing (in W m^{-2}); ΔT is the increase in surface temperature (in K) above a natural steady state; and α is the climate feedback parameter (in $\text{W m}^{-2} \text{ K}^{-1}$), which is inversely proportional to the effective equilibrium climate sensitivity¹. All terms in equation (3) are time integrated over the period of interest.

The IPCC Fifth Assessment Report gives a ΔOHC 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 ΔOHC that is compatible with both APO and hydrographic constraints. By applying equation (3)¹ to surface temperature data over the period 1991–2016 (HadCrut4 version 4.5, ref. 64, 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 K back to 2.0 K . An increase of the lower bound from 1.5 K to 2.0 K corresponds to a need to reduce maximum emissions by 25% to stay within the 2°C global warming target (because of the almost linear

relationship between warming and cumulative emissions; see Fig. SPM.10 in ref. ¹). This corresponds to a reduction in maximum allowable cumulative CO₂ emissions from 4,760 Gt CO₂ to 3,570 Gt CO₂.

We tested the sensitivity of the climate sensitivity by using three alternate temperature datasets (NASA GISS Surface Temperature Analysis GISTEMP⁶⁵, available at <https://data.giss.nasa.gov/gistemp/>; the NOAA/OAR/ESRL global surface temperature data⁶⁶ 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/; 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.

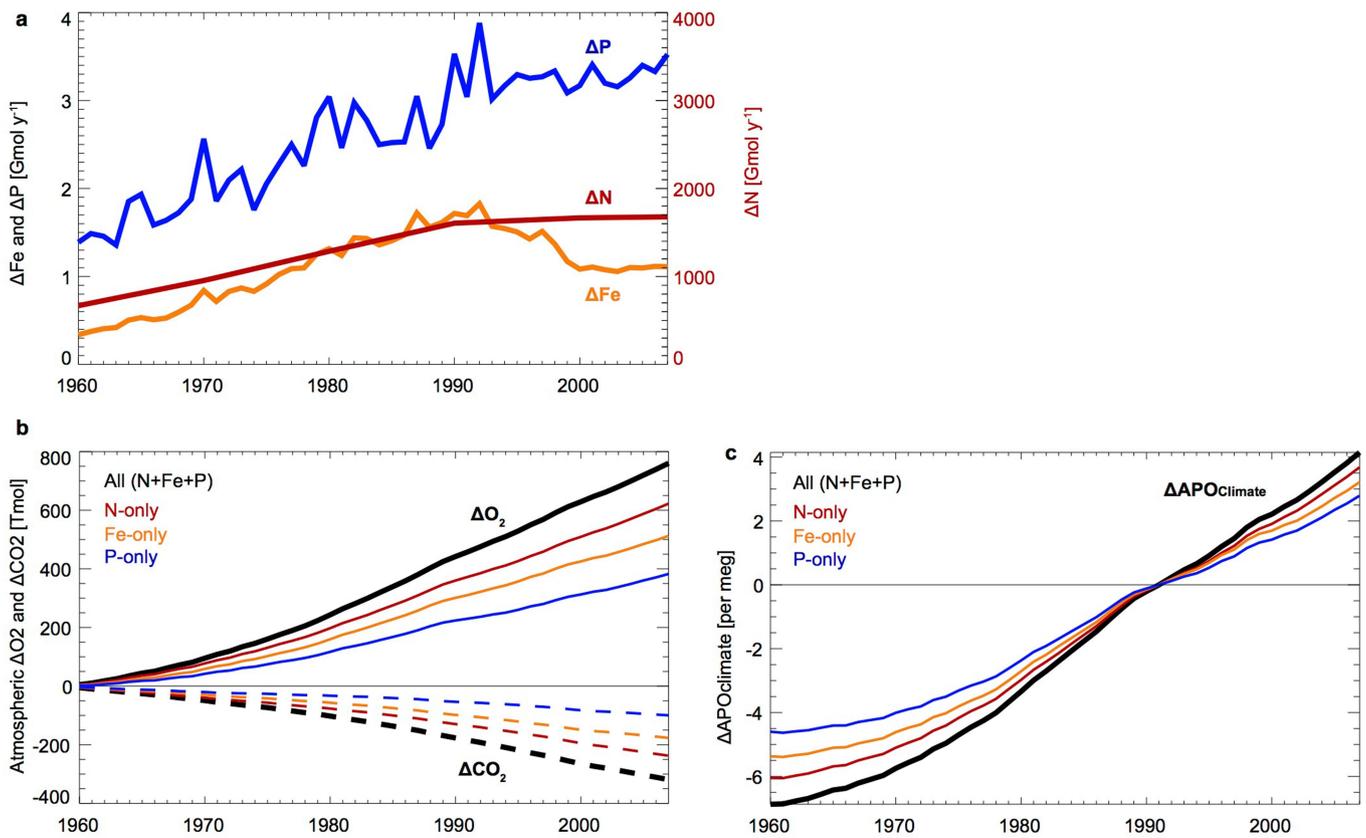
Link to global ocean deoxygenation. Our application of O₂ atmospheric measurements to constrain long-term ocean warming can be compared with earlier work that considers warming-driven oceanic O₂ outgassing. Multiplying our warming rate of $1.33 \pm 0.20 \times 10^{22} \text{ J yr}^{-1}$ by the O₂-to-heat 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 data⁶⁷ (about $40 \text{ Tmol O}_2 \text{ yr}^{-1}$), ocean data above 1,000 m ($55\text{--}65 \text{ Tmol O}_2 \text{ yr}^{-1}$, refs. ^{68,69}) and global ocean data⁷⁰ ($96 \pm 42 \text{ Tmol O}_2 \text{ yr}^{-1}$). This calculation suggests that ocean CO₂ uptake is reduced by warming at a ratio of around 0.70 nmol of CO₂ per joule (the difference between the O₂-to-heat ratio of 3.70 nmol J^{-1} and the OPO-to-heat ratio of 4.45 nmol J^{-1}).

Code availability. ESM codes are available online for IPSL-CM5A-LR (cmc.ipsl.fr/ipsl-climate-models/), GFDL-ESM2M (mdl-mom5.herokuapp.com/web/docs/project/quickstart/), UVic (climate.uvic.ca/model/) and CESM (www.cesm.ucar.edu/models/).

Data availability

Scripps APO data are available at <http://scrippso2.ucsd.edu/apo-data>. APO_{Climate} data, contributions to APO_{OBS} 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).

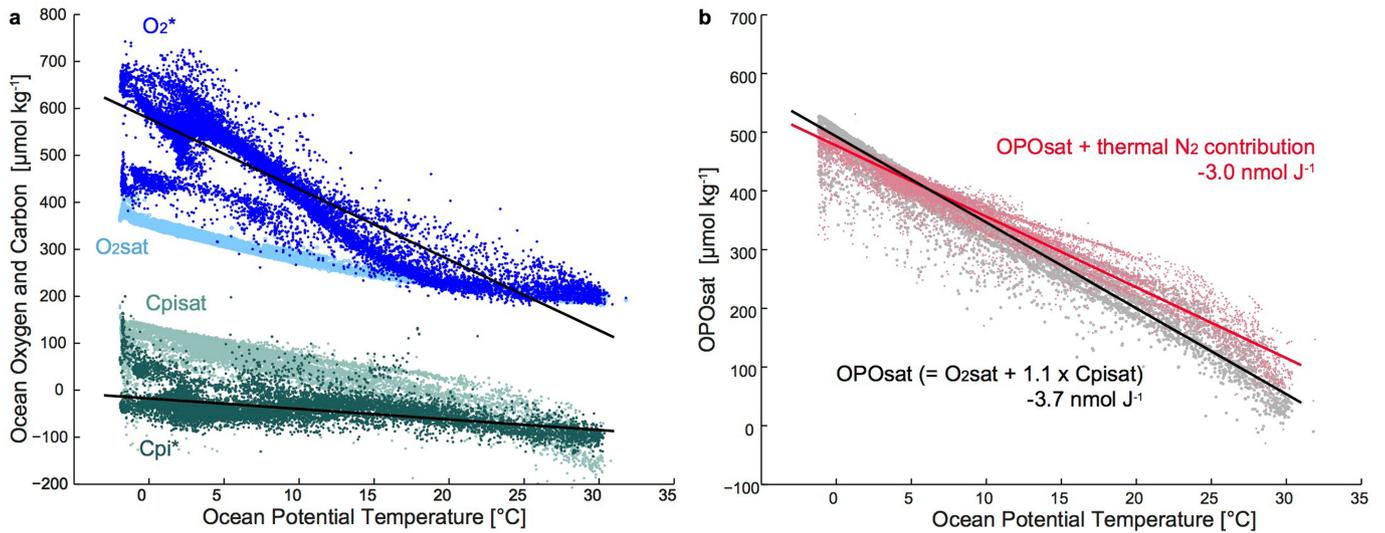
33. Severinghaus, J. P. *Studies of the Terrestrial O₂ and Carbon Cycles in Sand Dune Gases and in Biosphere*. PhD Thesis, Columbia Univ. (1995).
34. Hamme, R. C. & Keeling, R. F. Ocean ventilation as a driver of interannual variability in atmospheric potential oxygen. *Tellus B Chem. Phys. Meteorol.* **60**, 706–717 (2008).
35. Andres, R. J., Boden, T. A. & Higdon, D. A new evaluation of the uncertainty associated with CDIAC estimates of fossil fuel carbon dioxide emission. *Tellus B Chem. Phys. Meteorol.* **66**, 23616 (2014).
36. Keeling, R. F., Manning, A. C., Paplawsky, W. J. & Cox, A. C. On the long-term stability of reference gases for atmospheric O₂/N₂ and CO₂ measurements. *Tellus B Chem. Phys. Meteorol.* **59**, 3–14 (2007).
37. Ballantyne, A. P. et al. Audit of the global carbon budget: estimate errors and their impact on uptake uncertainty. *Biogeosciences* **12**, 2565–2584 (2015).
38. Bronselaer, B., Winton, M., Russell, J., Sabine, C. L. & Khattiwala, S. Agreement of CMIP5 simulated and observed ocean anthropogenic CO₂ uptake. *Geophys. Res. Lett.* **44**, 12298–12305 (2017).
39. Oeschger, H., Siegenthaler, U., Schotterer, U. & Gugelmann, A. A box diffusion model to study the carbon dioxide exchange in nature. *Tellus* **27**, 168–192 (1975).
40. MacFarling Meure, C. et al. Law Dome CO₂, CH₄ and N₂O ice core records extended to 2000 years BP. *Geophys. Res. Lett.* **33**, L14810 (2006).
41. Wang, D., Gouhier, T. C., Menge, B. A. & Ganguly, A. R. Intensification and spatial homogenization of coastal upwelling under climate change. *Nature* **518**, 390–394 (2015).
42. Ito, T., Nenes, A., Johnson, M. S., Meskhidze, N. & Deutsch, C. Acceleration of oxygen decline in the tropical Pacific over the past decades by aerosol pollutants. *Nat. Geosci.* **9**, 443–447 (2016).
43. Jickells, T. D. et al. A reevaluation of the magnitude and impacts of anthropogenic atmospheric nitrogen inputs on the ocean. *Glob. Biogeochem. Cycles* **31**, 289–305 (2017).
44. Somes, C. J., Landolfi, A., Koeve, W. & Oschlies, A. Limited impact of atmospheric nitrogen deposition on marine productivity due to biogeochemical feedbacks in a global ocean model. *Geophys. Res. Lett.* **43**, 4500–4509 (2016).
45. Aumont, O., Ethé, C., Tagliabue, A., Bopp, L. & Gehlen, M. PISCES-v2: an ocean biogeochemical model for carbon and ecosystem studies. *Geosci. Model Dev.* **8**, 2465–2513 (2015).
46. Talley, L. D. et al. Changes in ocean heat, carbon content, and ventilation: a review of the first decade of GO-SHIP global repeat hydrography. *Annu. Rev. Marine Sci.* **8**, 185–215 (2016).
47. Sarmiento, J. L. & Gruber, N. Sinks for anthropogenic carbon. *Phys. Today* **55**, 30–36 (2002).
48. Garcia, H. E. & Gordon, L. I. Oxygen solubility in seawater: better fitting equations. *Limnol. Oceanogr.* **37**, 1307–1312 (1992).
49. Gruber, N., Sarmiento, J. L. & Stocker, T. F. An improved method for detecting anthropogenic CO₂ in the oceans. *Glob. Biogeochem. Cycles* **10**, 809–837 (1996).
50. Dunne, J. P. et al. GFDL's ESM2 global coupled climate-carbon Earth system models. Part I: physical formulation and baseline simulation characteristics. *J. Clim.* **25**, 6646–6665 (2012).
51. Dunne, J. P. et al. GFDL's ESM2 global coupled climate-carbon Earth system models. Part II: carbon system formulation and baseline simulation characteristics. *J. Clim.* **26**, 2247–2267 (2013).
52. Séférian, R., Iudicone, D., Bopp, L., Roy, T. & Madec, G. Water mass analysis of effect of climate change on air-sea CO₂ fluxes: the Southern Ocean. *J. Clim.* **25**, 3894–3908 (2012).
53. Kay, J. E. et al. The Community Earth System Model (CESM) large ensemble project: a community resource for studying climate change in the presence of internal climate variability. *Bull. Am. Meteorol. Soc.* **96**, 1333–1349 (2015).
54. Keller, D. P., Oschlies, A. & Eby, M. A new marine ecosystem model for the University of Victoria Earth System Climate Model. *Geosci. Model Dev.* **5**, 1195–1220 (2012).
55. Bopp, L. et al. Multiple stressors of ocean ecosystems in the 21st century: projections with CMIP5 models. *Biogeosciences* **10**, 6225–6245 (2013).
56. Keller, D. P., Kriest, I., Koeve, W. & Oschlies, A. Southern Ocean biological impacts on global ocean oxygen. *Geophys. Res. Lett.* **43**, 6469–6477 (2016).
57. Long, M. C., Deutsch, C. & Ito, T. Finding forced trends in oceanic oxygen. *Glob. Biogeochem. Cycles* **30**, 381–397 (2016).
58. Taylor, K. E., Stouffer, R. J. & Meehl, G. A. An overview of CMIP5 and the experiment design. *Bull. Am. Meteorol. Soc.* **93**, 485–498 (2012).
59. Moore, J. K., Lindsay, K., Doney, S. C., Long, M. C. & Misumi, K. Marine ecosystem dynamics and biogeochemical cycling in the Community Earth System Model [CESM1(BGC)]: comparison of the 1990s with the 2090s under the RCP4.5 and RCP8.5 scenarios. *J. Clim.* **26**, 9291–9312 (2013).
60. Rödenbeck, C., Le Quéré, C., Heimann, M. & Keeling, R. F. Interannual variability in oceanic biogeochemical processes inferred by inversion of atmospheric O₂/N₂ and CO₂ data. *Tellus B Chem. Phys. Meteorol.* **60**, 685–705 (2008).
61. Hamme, R. C. Mechanisms controlling the global oceanic distribution of the inert gases argon, nitrogen and neon. *Geophys. Res. Lett.* **29**, 35-1–35-4 (2002).
62. Trenberth, K. E., Fasullo, J. T., von Schuckmann, K. & Cheng, L. Insights into Earth's energy imbalance from multiple sources. *J. Clim.* **29**, 7495–7505 (2016).
63. WCRP Global Sea Level Budget Group. Global sea level budget 1993–present. *Earth Syst. Sci. Data* **10**, 1551–1590 (2018).
64. Morice, C. P., Kennedy, J. J., Rayner, N. A. & Jones, P. D. Quantifying uncertainties in global and regional temperature change using an ensemble of observational estimates: the HadCRUT4 data set. *J. Geophys. Res. Atmos.* **117**, D08101 (2012).
65. Hansen, J., Ruedy, R., Sato, M. & Lo, K. Global surface temperature change. *Rev. Geophys.* **48**, RG4004 (2010).
66. Vose, R. S. et al. NOAA's merged land-ocean surface temperature analysis. *Bull. Am. Meteorol. Soc.* **93**, 1677–1685 (2012).
67. Keeling, R. F., Körtzinger, A. & Gruber, N. Ocean deoxygenation in a warming world. *Annu. Rev. Mar. Sci.* **2**, 199–229 (2010).
70. Schmidtko, S., Stramma, L. & Visbeck, M. Decline in global oceanic oxygen content during the past five decades. *Nature* **542**, 335–339 (2017).
68. Helm, K. P., Bindoff, N. L. & Church, J. A. Observed decreases in oxygen content of the global ocean. *Geophys. Res. Lett.* **38**, L23602 (2011).
69. Ito, T., Minobe, S., Long, M. C. & Deutsch, C. Upper ocean O₂ trends: 1958–2015. *Geophys. Res. Lett.* **44**, 4214–4223 (2017).



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

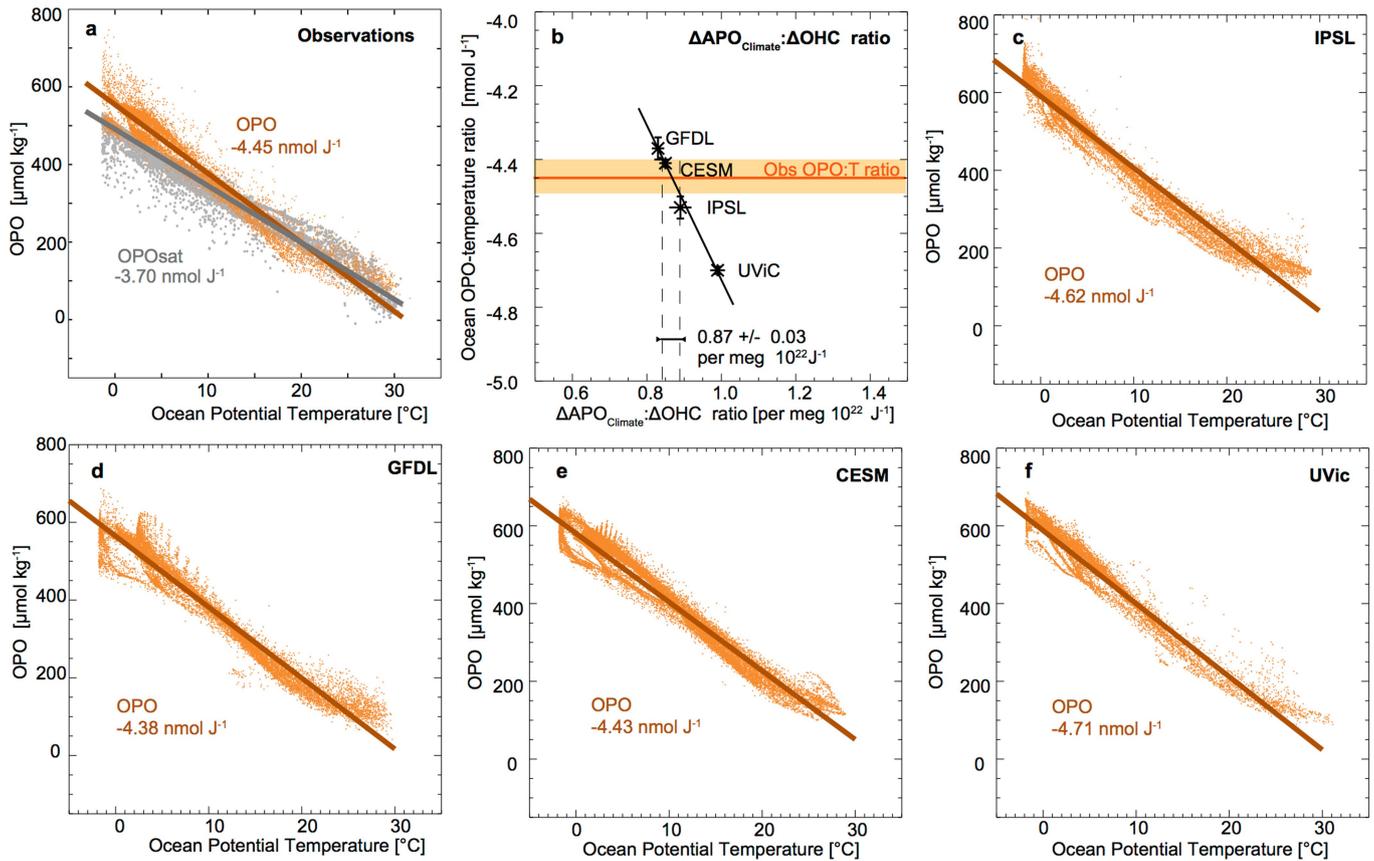
a, Anomaly, relative to 1850 levels, in deposition of atmospheric anthropogenic aerosols (N, P and Fe) at the air–sea interface between 1960 and 2007, derived from model simulations with and without aerosols²².

b, Impact of aerosol eutrophication on atmospheric O_2 (solid lines) and 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 O_2^* , $\text{O}_{2\text{sat}}$, C_{pi}^* and C_{pisat} as a function of potential temperature in the Glodapv2 database³². **b**, OPO_{sat} ($= \text{O}_{2\text{sat}} + 1.1 \times \text{C}_{\text{pisat}}$, in grey) and the expected effects of OPO_{sat} and the thermal exchanges of N_2 ($= \text{O}_{2\text{sat}} + 1.1 \times \text{C}_{\text{pisat}} - X_{\text{O}_2} / X_{\text{N}_2} [\text{N}_2 - \text{mean}(\text{N}_2)]$), in red). For clarity only 16×10^3 points randomly picked out of the 78,456 data points available

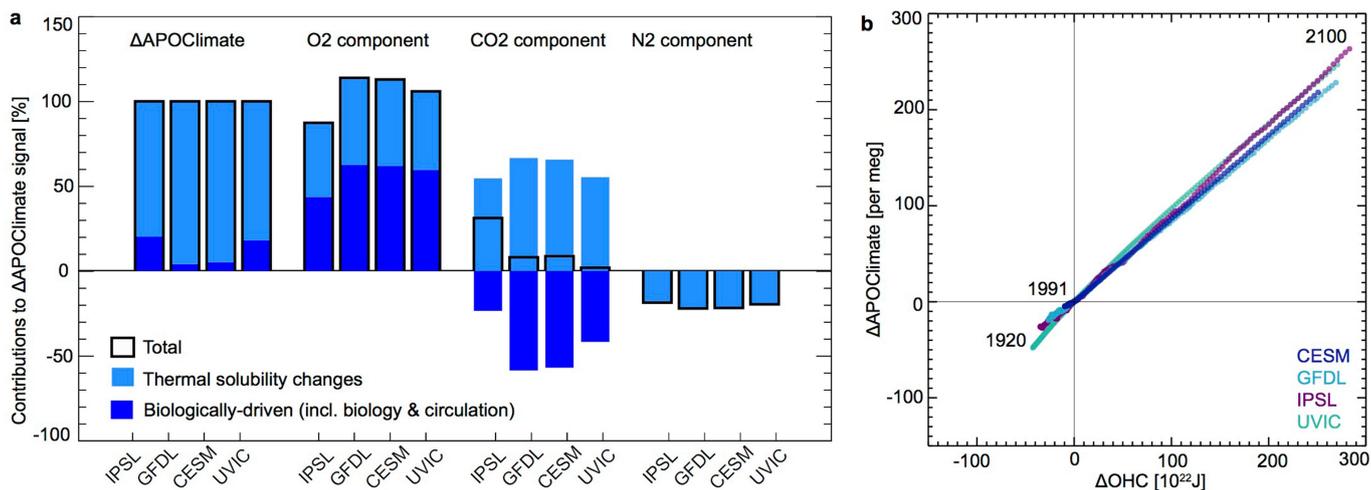
are shown for each variable. Note that very low values of O_2^* (around $450 \mu\text{mol kg}^{-1}$) at low temperature (less than 10°C) correspond to data collected in the Arctic Ocean, where phosphate concentrations (used for O_2^* calculation) are comparatively lower than in other cold ocean regions. Low O_2^* values in the Arctic explain the relatively low values of OPO shown in Extended Data Fig. 3a at temperatures below 10°C .



Extended Data Fig. 3 | Link between OPO, $\text{APO}_{\text{Climate}}$ and ocean heat.

a, c–f, OPO concentrations (yellow) and OPO concentrations at saturation based on O_2 and CO_2 solubility (OPO_{sat} , grey) as a function of ocean temperature in the GLODAPv2 database³² (**a**) and four Earth-system models (IPSL, GFDL, CESM and UVic; **c–f**). Slopes give the OPO-to-temperature ratios in nmol J^{-1} . **b,** The link between $\Delta\text{APO}_{\text{Climate}}$ and

changes in ocean heat content (that is, $\Delta\text{APO}_{\text{Climate}}$ -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^{-1} (vertical dashed lines). To avoid visual saturation, only 16,000 points, picked randomly, are shown for OPO.



Extended Data Fig. 4 | Changes in $\text{APO}_{\text{Climate}}$ ($\Delta\text{APO}_{\text{Climate}}$) and ocean heat content (ΔOHC) 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_2 , CO_2 and N_2 components—that is, how much of $\Delta\text{APO}_{\text{Climate}}$ is explained by changes in O_2 , CO_2 and N_2 air–sea fluxes due to ocean saturation changes and biologically driven changes. **b**, Model $\Delta\text{APO}_{\text{Climate}}$ -to- ΔOHC ratios over the 180 years of simulation (referenced to year 1991) in per meg per 10^{22} J units are: 0.85 ± 0.01 (CESM), 0.83 ± 0.01 (GFDL), 0.89 ± 0.03 (IPSL) and 0.99 ± 0.02 (UVic).

Extended Data Table 1 | Sources of the hydrographic databased estimates of global changes in ocean heat content (ΔOHC) 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

	1991-2016	1993-2016	2007-2016
	$\Delta\text{OHC trend } (\pm 1-\sigma)$	$\Delta\text{OHC trend } (\pm 1-\sigma)$	$\Delta\text{OHC trend } (\pm 1-\sigma)$
$\text{APO}_{\text{Climate}}$	1.33 ± 0.20	-	-
PMEL	-	1.35 ± 0.10	1.16 ± 0.20
MRI	1.00 ± 0.11	1.03 ± 0.12	1.23 ± 0.22
NCEI	0.89 ± 0.08	0.90 ± 0.09	1.28 ± 0.16
CHEN	1.07 ± 0.07	1.10 ± 0.08	1.09 ± 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- σ uncertainty	References
$\Delta\text{APO}_{\text{OBS}}$				
Corrosion			± 0.3 per meg yr ⁻¹	
Leakage			± 0.2 per meg yr ⁻¹	
Desorption			± 0.1 per meg yr ⁻¹	
Thermal fractionation			± 2 per meg (± 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		± 0.03	
Oil	1.44	Ref. 36	± 0.03	Ref. 36
Gas	1.95		± 0.04	
Cement	0.0		± 0.00	
Flaring	1.98		± 0.07	
Emissions ΔCO_2				
Coal			$\pm 7.0\%$	
Oil	Time varying	Ref. 20	$\pm 5.5\%$	Ref. 35
Gas			$\pm 6.5\%$	
Cement			$\pm 12\%$	
Flaring			$\pm 12\%$	
$\Delta\text{APO}_{\text{Cant}}$				
ΔCant_0	Time varying (~ 2 to 3 PgC yr ⁻¹)	Ref. 21	1- σ of 10 experiments (< 0.3 PgC yr ⁻¹)	Ref. 21
			+ 1% uncertainty (< 0.03 PgC yr ⁻¹) (atmospheric CO ₂ history)	<i>this study</i>
$\Delta\text{Cant}'$	0.05 PgC yr ⁻¹ (0.12 per meg yr ⁻¹)	<i>this study</i>	± 0.05 PgC yr ⁻¹ (± 0.12 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 1σ 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 O₂, CO₂ and APO due to anthropogenic aerosol deposition

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

Trends in APO due to atmospheric deposition ($\Delta\text{APO}_{\text{AtmD}}$) are decomposed into contributions from the O₂ flux only (APO_{AtmD(O₂)}) and the CO₂ flux only (APO_{AtmD(CO₂)}). Results are from model simulations²². Anomalies in air–sea flux are positive towards the atmosphere. The total trend used in this study is in bold type.