

Supplemental Table S1 - List of CMIP6 experiments conducted with CanESM5-CanOE. The list of experiments conducted with CanESM5 is much more extensive and is not detailed here. Note that all realizations are numbered r*1p2f1, i.e., the p1 model was retired before any CanESM5-CanOE experiments were run.

MIP	Experiment	# of realizations
CMIP	piControl	500 y
CMIP	historical	3
CMIP	esm-piControl	500 y
CMIP	esm-hist	3
CMIP	1pctCO2	1
ScenarioMIP	ssp126	3
ScenarioMIP	ssp245	3
ScenarioMIP	ssp370	3
ScenarioMIP	ssp585	3
C4MIP	1pctCO2-bgc	1
C4MIP	1pctCO2-rad	1
C4MIP	esm-ssp585	3
CDRMIP	esm-pi-cdr-pulse	3
CDRMIP	esm-pi-CO2-pulse	3
OMIP	omip1	1

Supplemental Table S2 - Alkalinity sources and sinks associated with nitrogen cycle processes in moles of alkalinity equivalent per mole of N. Positive value indicates alkalinity source.

Process	Alkalinity source/sink
Phytoplankton NH ₄ uptake	-1
Phytoplankton NO ₃ uptake	+1
Organic N remineralization	+1
N ₂ fixation	+1
Nitrification (NH ₄ oxidation to NO ₃)	-2
Denitrification	+1
Annamox	0

Supplemental Table S3 - Other CMIP6 models used in the cross-model analyses. Model, centre, and variable names are official CMIP6 names.

Model	Centre	Country	Variables used
ACCESS-ESM1-5	CSIRO	Australia	dfe
CESM2	NCAR	US	dfe, dissic, no3, so, thetao, talk
CESM2-WACCM	NCAR	US	dissic, so, thetao, talk
CNRM-ESM2-1	CNRM-CERFACS	France	dfe, epc100, dissic, no3, o2, so, thetao, talk
GFDL-CM4	NOAA-GFDL	US	dfe, dissic, o2, so, thetao, talk
GFDL-ESM4	NOAA-GFDL	US	dfe, epc100, dissic, no3, o2, so, thetao, talk
IPSL-CM6A-LR	IPSL	France	dissic, o2, so, thetao, talk
MIROC-ES2L	MIROC	Japan	dfe, dissic, o2, so, thetao, talk
MPI-ESM1-2-HAM	HAMMOZ-Consortium	Germany	dfe, no3, o2, so, thetao
MPI-ESM1-2-LR	MPI-M	Germany	dfe, dissic, epc100, no3, o2, so, thetao
MPI-ESM1-2-HR	MPI-M	Germany	dissic, no3, o2, so, thetao, talk
MRI-ESM2-0	MRI	Japan	dissic, o2, so, thetao
NorESM2-LM	NCC	Norway	dfe, dissic, epc100, no3, o2
NorESM2-MM	NCC	Norway	dissic, no3, o2, so, thetao
UKESM1-0-LL	MOHC	UK	dfe, epc100, dissic, no3, o2, so, thetao, talk

Supplemental Table S4 - Latitude and longitude boundaries used for spatial averaging of regions (Figures 10, 14, 16, 17). In Figure 10, Western Subarctic Pacific and Eastern Subarctic Pacific are combined as “North Pacific” (latitude boundaries as below).

Region	S bound	N bound	W bound	E bound
North Atlantic	45N	65N	50W	10W
Western Subarctic Pacific	40N	52N	160E	160W
Eastern Subarctic Pacific	40N	52N	160W	130W
Southern Ocean		30S		
Tropical Pacific	2S	2N	180	76W
Tropical Atlantic	5S	5N	30W	0

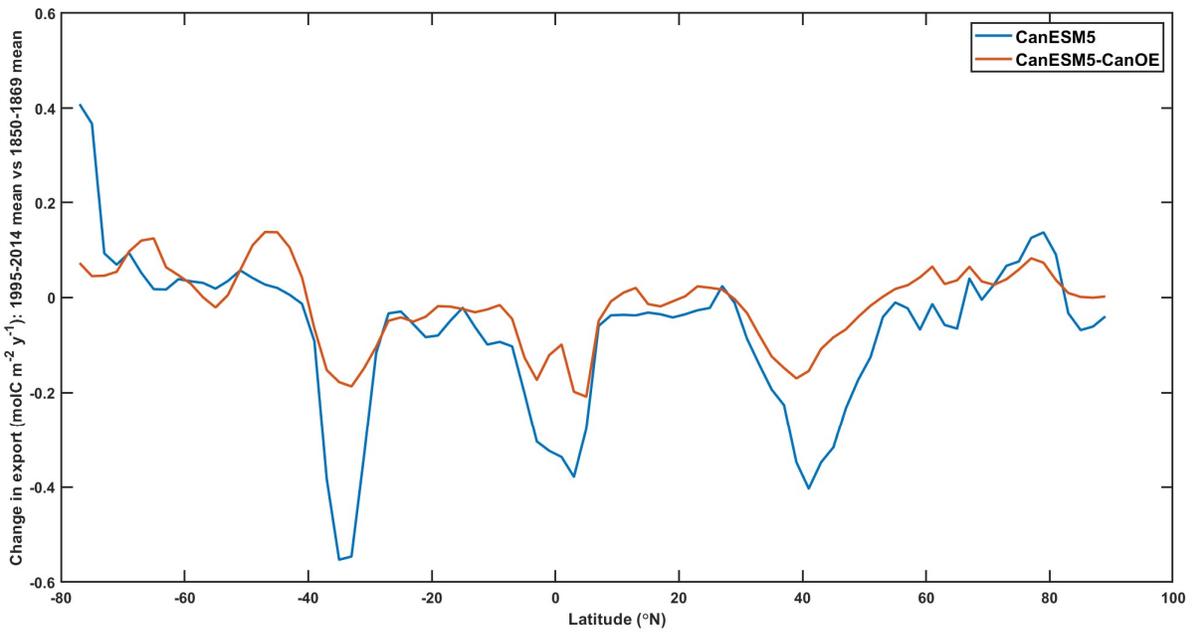


Figure S1 -Change in zonal mean export production in CanESM5 and CanESM5-CanOE over historical experiment (difference of 20 year means for first and last 20 y).

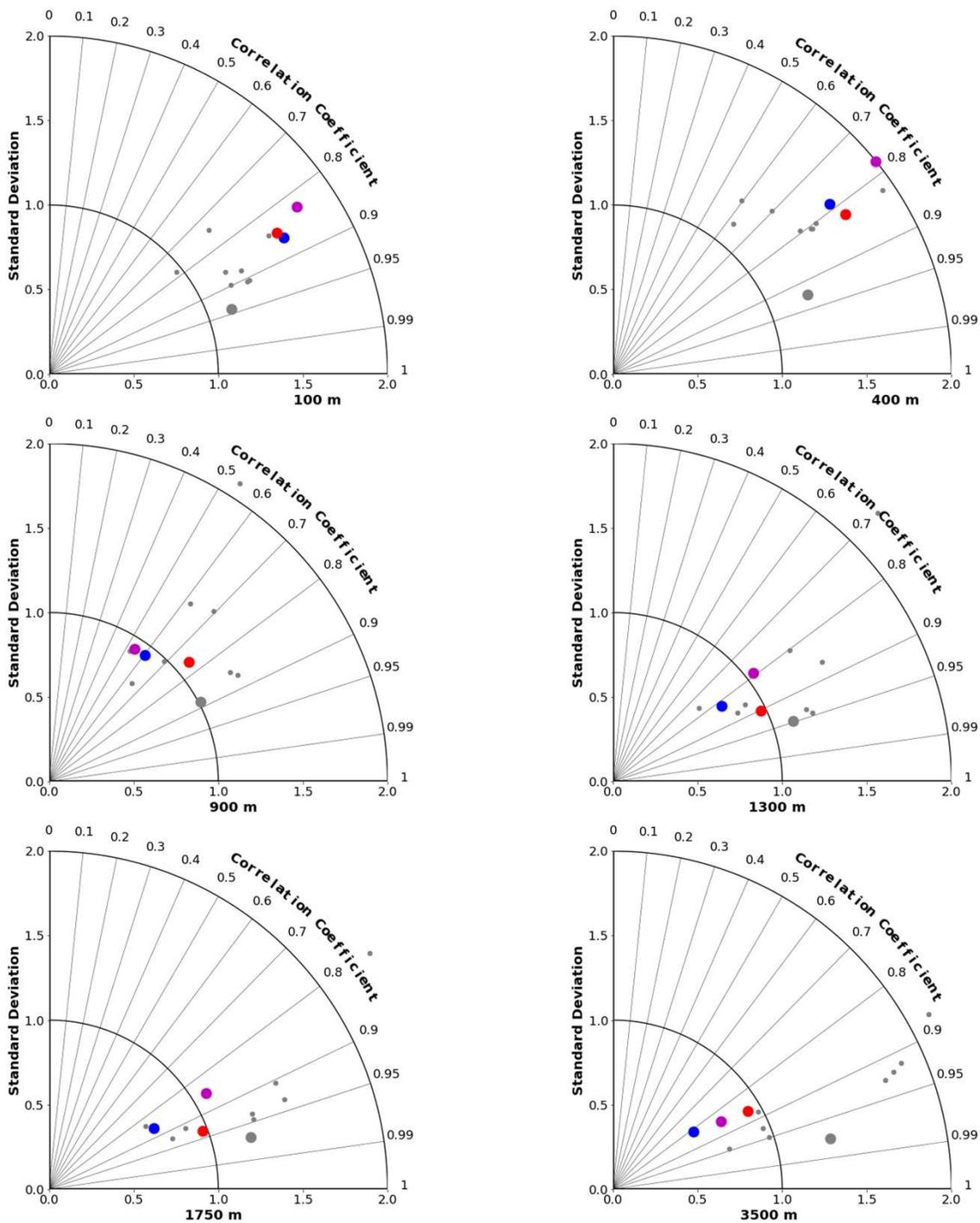
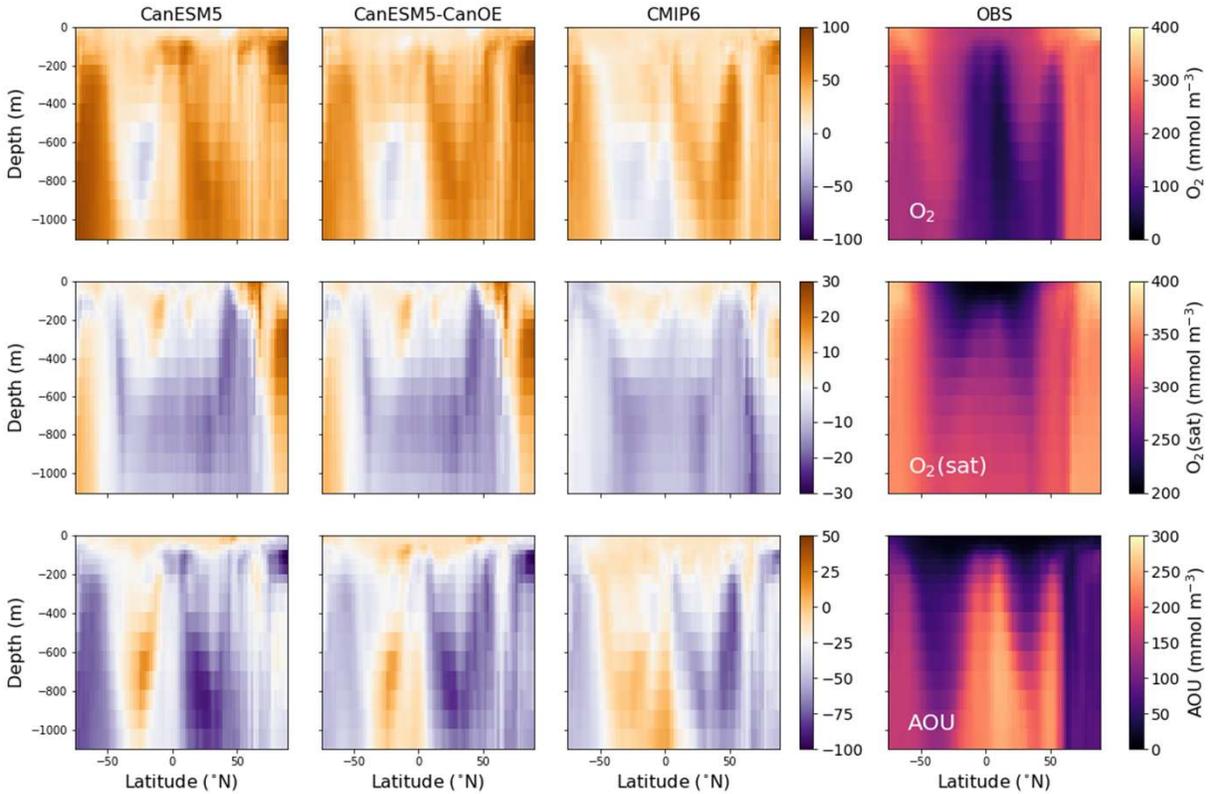
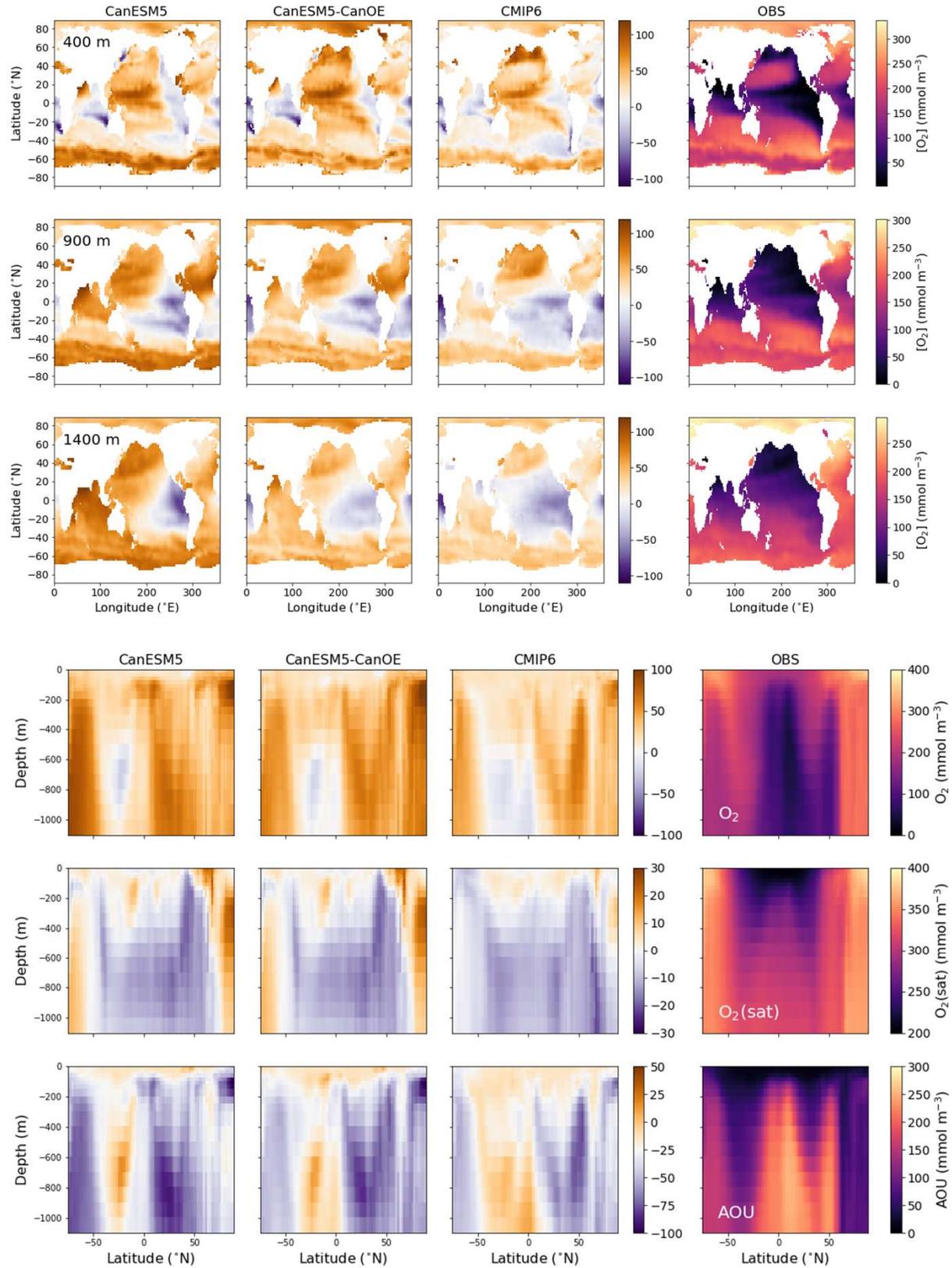
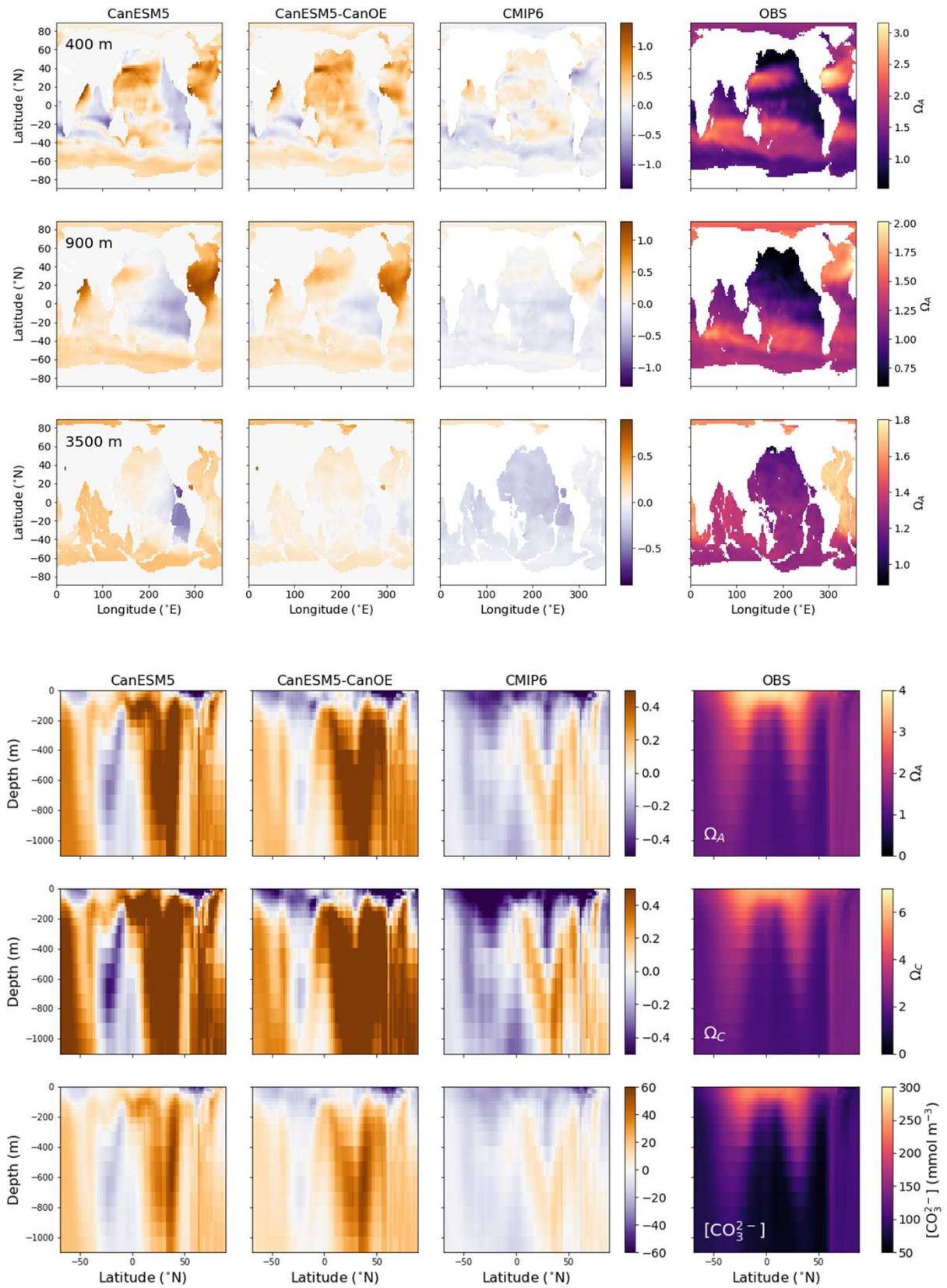


Figure S2 - Taylor diagrams for total alkalinity on various depth horizons. Colour codes and z levels as in Figures 4, 8, and 9.



Supplemental Figure S3 - Alternate versions of Figures 2, 3, 6 and 7 that show model deviations from observational data products.



Supplemental Figure S3 - Alternate versions of Figures 2, 3, 6 and 7 that show model deviations from observational data products.

Aeolian iron dissolution

Aeolian iron was introduced assuming a fractional solubility of 1.4% in the surface layer and adopting the PISCES parameterization for subsurface dissolution. According to Aumont et al (2015) this assumes a sinking speed of 2 m/d and a dissolution rate of 0.0001 d^{-1} , which translates to an e-folding length scale of 20000 m and is consistent with the NEMO 3.4 PISCES code, except for the length scale, which is set to a much smaller 1000 m.

We can think of this parameterization as the divergence dF/dz of a flux that declines exponentially with depth:

$$Fz = F0 * \exp(-k/w * z)$$

where k is a first-order dissolution rate and w is the sinking speed (e.g., Christian et al., 1997).

$$dF/dz = -k/w * Fz$$

is the input of dissolved iron at depth z .

Because the subsurface layers are much thicker than the surface layer, the total amount of iron introduced results in an effective fractional solubility of aeolian iron that is much larger than the specified 1.4%. The whole water column dissolution is 6.4%, of which 22% is introduced in the surface layer. Note that if the somewhat arbitrary substitution of an e-folding length scale of 1000 m for 20000 m were not done, the difference would be much larger: a net fractional solubility of 25%, of which only 5.5% would go into the surface layer. (All of these numbers are grid-specific and are for the ORCA1 levels.)

Because in our model the rate of scavenging loss is very high for concentrations above 0.6 nM, much of this additional aeolian iron will be removed without ever entering the euphotic zone. While the net dissolution of the total aeolian iron input is much higher than the specified fractional solubility of 1.4%, it is important to note that in terms of biological response it is clearly not equivalent to a model that supplies aeolian Fe only to the surface layer with a fractional solubility of 6.4%. It is probably closer to a model with 1.4% and no subsurface dissolution, although exactly where it falls on this spectrum will vary with region and season.

Aumont, O., C. Ethé, A. Tagliabue, L. Bopp, and M. Gehlen, 2015. PISCES-v2: an ocean biogeochemical model for carbon and ecosystem studies. *Geoscientific Model Development* 8: 2465–2513.

Christian, J.R., M.R. Lewis, and D.M. Karl, 1997. Vertical fluxes of carbon, nitrogen and phosphorus in the North Pacific Subtropical Gyre near Hawaii. *Journal of Geophysical Research* 102: 15667-15677.

Dissolved iron model comparison to observations

As there is no gridded global data product for dissolved iron (dFe) we present here some comparisons with individual bottle samples from the GEOTRACES Intermediate Data Product 2017 (Schlitzer et al., 2018), the MBARI data compilation (www3.mbari.org/chemsensor/Data/), and the Pacific data set compiled by PICES WG22 (meetings.pices.int/members/working-groups/disbanded/wg22). The MBARI data include both profile (the "Global Iron Data") and surface transect ("MBARI SOLAS") data (the SOLAS data also include some profiles but are primarily underway surface measurements). The GEOTRACES data are primarily profile data (393 profiles total, only ~10% of 7519 data are <50 m). Most of the MBARI data are also in the PICES data set; these were preprocessed to remove redundant data. Data from the underway surface transects were excluded to avoid overweighting these regions due to autocorrelation, leaving a total of 3575 data points. Concentrations > 2.5 nM, which comprise 1.3% of GEOTRACES data and 3.3% of PICES/MBARI data, were excluded from the model/data comparisons below.

Evaluating the CanOE model beyond what has been discussed in the main text identified several key points, most of which have already been made to some degree. CanESM5-CanOE compares favourably with other CMIP6 models but is quite biased towards a 'nutrient-type' rather than 'scavenged-type' profile (Figures S4b, c, e-g). CanOE rates of Fe scavenging are very high above 0.6 nM dFe and very low below this concentration, resulting in an almost constant deep water concentration of 0.6 nM. The collected data (of which there are >10000 vs only a few hundred in 1997) show that the basic hypothesis articulated by Johnson et al. (1997) still holds: deep-water concentrations are generally close to 0.6 nM, although much higher concentrations are sometimes observed at depths of thousands of metres (Figure S4b, see e.g. Resing et al., 2014). High concentrations are also sometimes observed in near-surface waters (Figure S4b) and CanOE is among the models least able to reproduce these. CanOE consistently overestimates the lowest observed concentrations and underestimates the higher ones (Figure S4d). One might argue that other models do little better in a statistical sense. However, in several models the deviations from the 1:1 line are less systematic than in CanOE; the residuals may be larger but they are more homogeneous. Therefore purely statistical comparisons as in Figures S4e-g can be misleading. These biases can clearly be attributed to the rather simplistic scavenging model employed, and will be addressed in future versions.

Figure S4a – Global distribution of dissolved iron (dFe) measurements in the GEOTRACES (black/green) and PICES/MBARI (blue) data compilations. GEOTRACES Pacific data north of 25°S are indicated in green. Blue dots outside the Pacific are MBARI data.

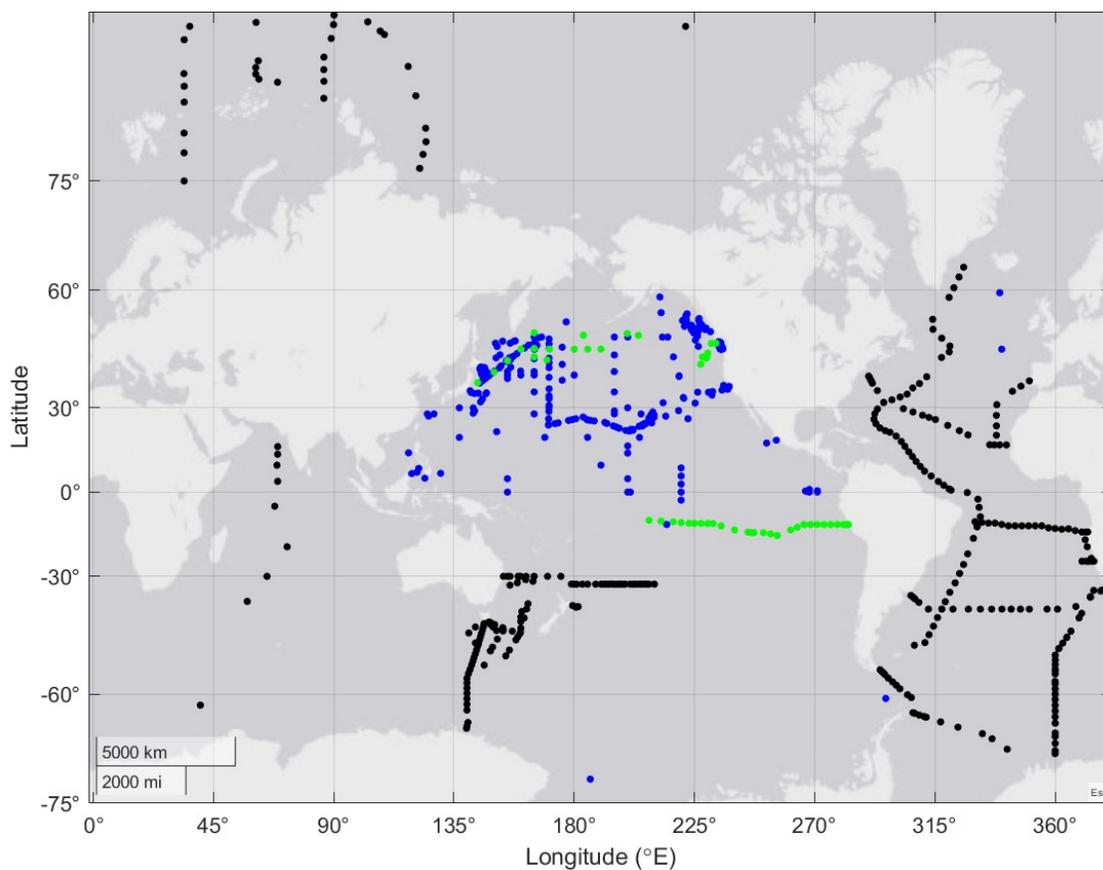


Figure S4b - All observations of dissolved iron (dFe) concentration (<2 nM) plotted against sampling depth. GEOTRACES data are in black and PICES/MBARI data are blue. Model data are global mean profiles of annual mean data as in Figure 13 in the main text.

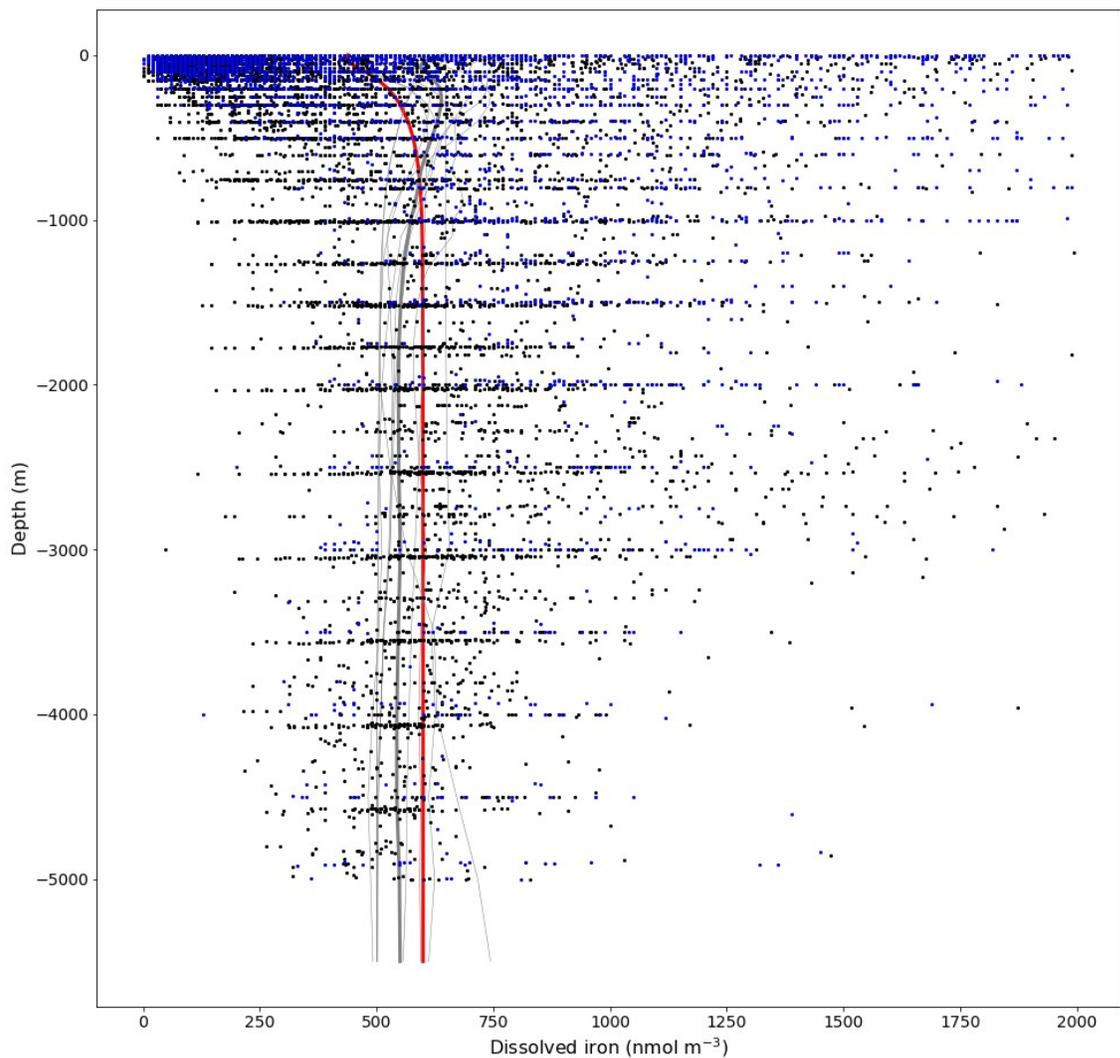


Figure S4c - Mean of observations of dissolved iron (dFe) concentration (<2 nM) for various depth strata, including both GEOTRACES and PICES/MBARI data. Means of all data available in that depth range; spatial coverage is very incomplete and inconsistent among the depth strata. Averaging layers are 50 m (0-1000 m), 100 m (1000-3000 m) or 300 m (>3000 m) thick. Model data are global mean profiles of annual mean data as in Figure 13 in the main text.

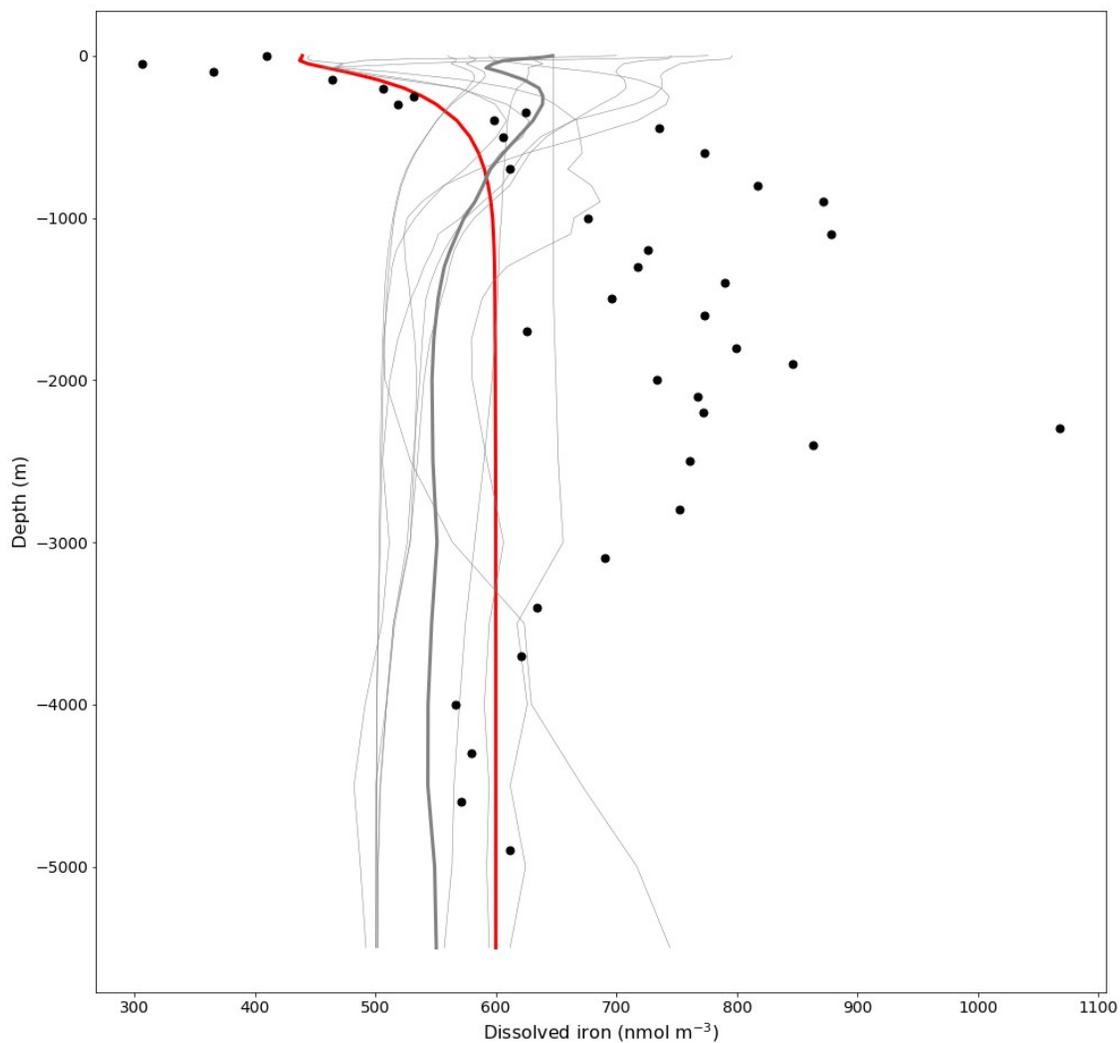


Figure S4d - Modelled and observed mean dissolved iron (dFe) concentrations (excluding those >2.5 nM) in the upper 50 m at locations where a depth profile was taken (N=1122). Observed data are means of all measurements made at depths <50 m within a given profile. Model data are climatological surface values for the month in which the observed data were collected (red: CanESM5-CanOE; black CNRM-ESM2-1; blue: GFDL-CM4; magenta: MPI-ESM1-2-LR). Selection of models is somewhat arbitrary but includes the models that show the highest overall skill according to the metrics shown in Figures S4e-g.

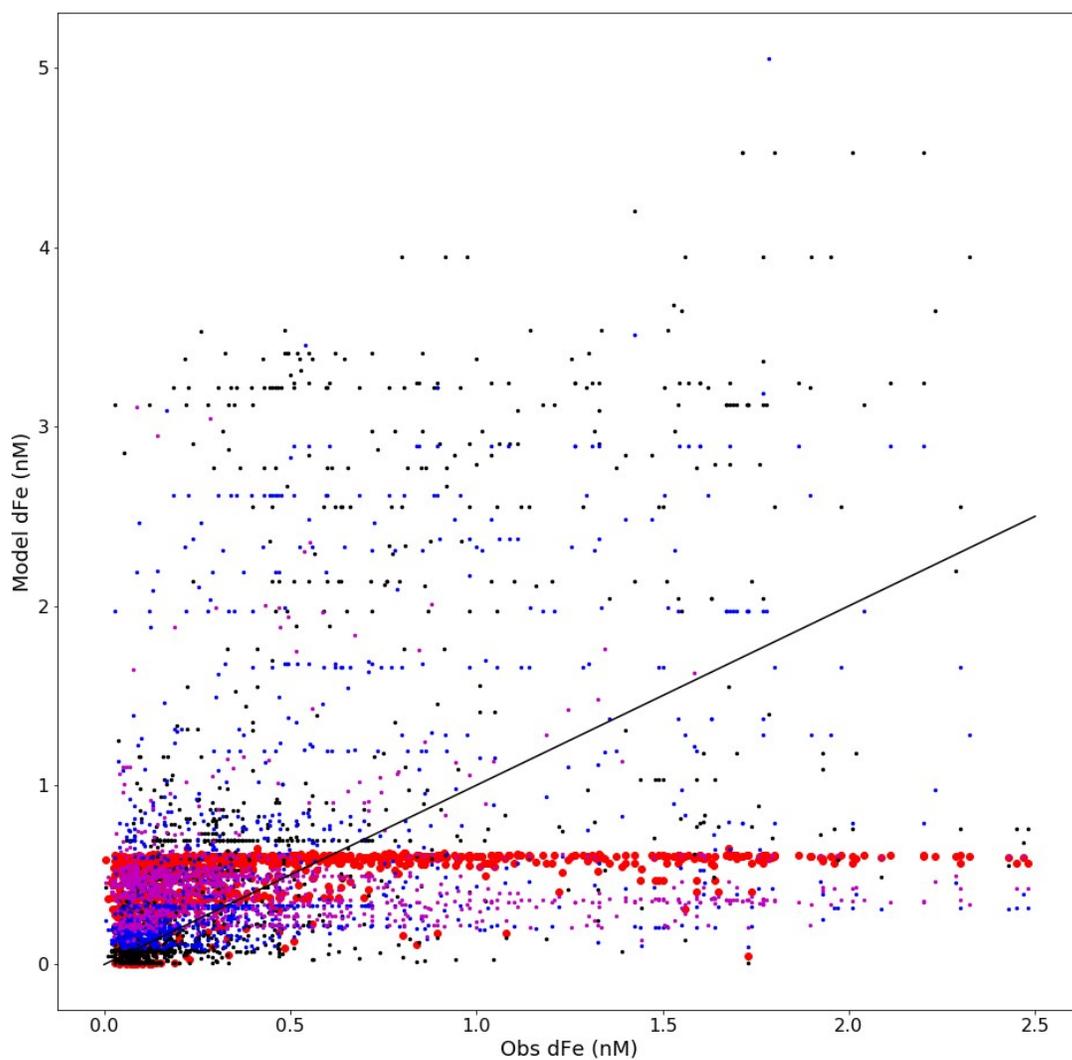


Figure S4e - Root mean square error and correlation coefficient for surface dissolved iron data (upper 50 m mean with maximum of 2.5 nM, as in Figure S4d) for CMIP6 models for which seasonal data were available.

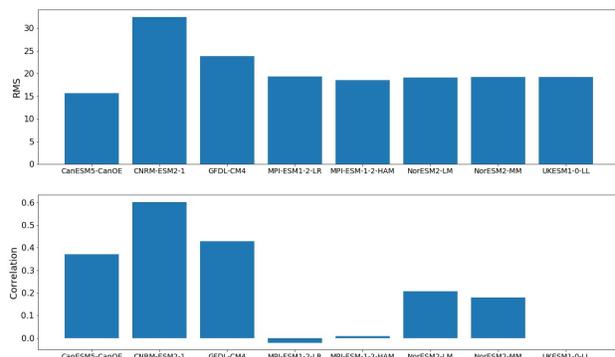


Figure S4f - As S4e but for the Pacific only (north of 25°S).

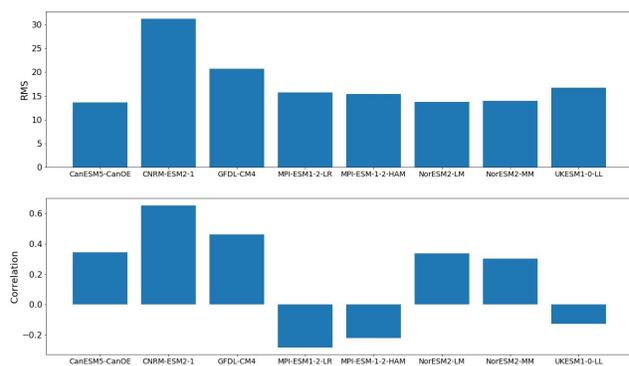
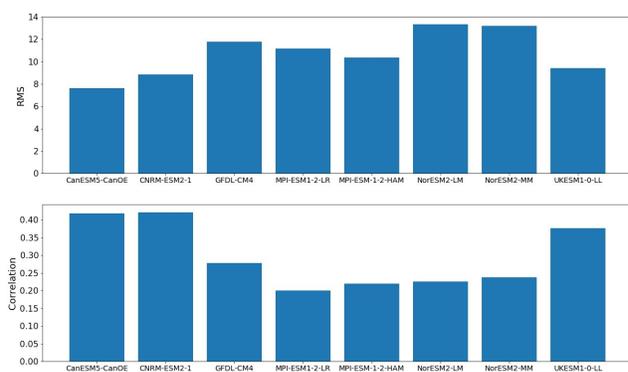


Figure S4g - As S4e but excluding the Pacific north of 25°S.



References

Resing, J.A., P.N. Sedwick, C.R. German, W.J. Jenkins, J.W. Moffett, B.M. Sohst and A. Tagliabue, 2015. Basin-scale transport of hydrothermal dissolved metals across the South Pacific Ocean. *Nature* 523 : 200–203.

Schlitzer, R., et al., 2018. The GEOTRACES Intermediate Data Product 2017, *Chemical Geology*, 493, 210-223

The data file for the PICES WG22 data compilation requests that data users cite the original publications. As all of the data are used except for the SOLAS underway data, all of the original references are listed here:

Boyle, E.A., B.A. Bergquist, R.A. Kayser and N. Mahowald, 2005. Iron, manganese and lead at Hawaii Ocean Time-Series station ALOHA: Temporal variability and an intermediate water hydrothermal plume. *Geochimica et Cosmochimica Acta*, 69: 933-952.

Brown, M. T., W. M. Landing, and C. I. Measures, 2005. Dissolved and particulate Fe in the western and central North Pacific: Results from the 2002 IOC cruise. *Geochem, Geophys. Geosyst.*, 6, Q10001, doi: 10.1029/2004GC000893.

Bruland, K., Orians, K. and Cowen, P. ,1994. Reactive trace metals in the stratified central North Pacific. *Geochem. Cosmochim. Acta*. 58, 3171-3182.

Coale, K. H., Fitzwater, S. E., Gordon, R. M., Johnson, K. S. and Barber, R. T., 1996a. Control of community growth and export production by upwelled iron the equatorial Pacific Ocean. *Nature*, 379:621-624.

Ezoe M , Ishita T , Kinugasa M , Lai X, Norisuye K, Sohrin Y, 2004, Distributions of dissolved and acid-dissolvable bioactive trace metals in the North Pacific Ocean. *Geochemical Journal*, 38(6), 535-550.

Fitzwater, S.E., K.H. Coale, R.M. Gordon, K.S. Johnson, M.E. Ondrusek, 1996, Iron deficiency and phytoplankton growth in the equatorial Pacific, *Deep-Sea Res. II*, 43, 995-1015

Fujishima, Y., K. Ueda, M. Maruo, E. Nakayama, C. Tokutome, H. Hasegawa M. Matsui and Y. Sohrin. Distribution of Trace Bioelements in the Subarctic North Pacific Ocean and the Bering Sea (the R/V Hakuho Maru Cruise KH-97-2), *Journal of Oceanography*, Vol. 57, pp.261 to 273, 2001

Gordon, R. M., Johnson, K. S. and Coale, K. H., 1998. The behavior of iron and other trace elements during the IronEx I and PlumEx experiments in the equatorial Pacific. *Deep-Sea Res. II* 45: 995-1041.

Gordon, R.M. , J.H. Martin, G.A. Knauer, 1982, Iron in north-east Pacific waters, *Nature*, 299, 611-612

Johnson - MLML World Iron Data Set: Appendix A to Johnson, K. S., Gordon, R. M. and Coale, K. H., 1997, What controls dissolved iron concentrations in the world ocean? *Mar. Chem.* 57: 137-161.

Johnson, K.S., Elrod, V., Fitzwater, S., Plant, J.N., Chavez, F.P., Tanner, S.J., Gordon, M., Westphal, D.L., Perry, K.D., Wu, J., and Karl, D.M., 2003. Surface ocean-lower atmosphere interactions in the Northeast Pacific Ocean Gyre: Aerosols, iron, and the ecosystem response, *GBC*(17), 10.1029/2002GB002004

Johnson, K.S., F.P. Chavez, V.A. Elrod, S.E. Fitzwater, J.T. Pennington, K.R. Buck, and P.M. Walz, 2001. The annual cycle of iron and the biological response in central California coastal waters. *Geophys. Res. Letters* 28: 1247-1250.

Johnson, WK, Miller, LA, Sutherland, ND, Wong, CS, 2005. Iron transport by mesoscale Haida eddies in the Gulf of Alaska, *Deep-Sea Res. II*, 52: 933-953.

Kitayama, S., K. Kuma, E. Manabe, K. Sugie, H. Takata, Y.

Isoda, K. Toda, S. Saitoh, S. Takagi, Y. Kamei, and K. Sakaoka, 2009. Controls on iron distributions in the

deep water column of the North Pacific Ocean: iron(?) hydroxide solubility and humic-type fluorescent dissolved organic matter. *J. Geophys. Res.*, 114: C08019, doi:10.1029/2008JC004754

Kondo, Y., 2007. Dynamics of the organic Fe complexing ligands and phytoplankton in the Pacific Ocean. The University of Tokyo. Ph.D. Thesis, 256pp.

Kondo, Y., S. Takeda and K. Furuya, 2012. Distinct trends in dissolved Fe speciation between shallow and deep waters in the Pacific Ocean. *Marine Chemistry*, 134–135, 18–28

Kuma, K., J. Nishioka, K. Matsunaga, 1996, Controls on iron (III) hydroxide solubility in seawater: The influence of pH and natural organic chelators, *Limnol. Oceanogr.*, 41, 396-407

Kuma, K., A. Katsumoto, H. Kawakami, F. Takatori, K. Matsunaga, 1998, Spatial variability of Fe(III) hydroxide solubility in the water column of the northern North Pacific Ocean, *Deep-Sea Res. I*, 45, 91-113

Kuma, K., Isoda, Y. and Nakabayashi, S., 2003, Control on dissolved iron concentrations in deep waters in the western North Pacific: Iron(III) hydroxide solubility. *J. Geophys. Res.*, 108 (C9): 3289, doi:10.1029/2002JC001481 (2003).

Landing, W.M., K.W. Bruland, 1987, The contrasting biogeochemistry of iron and manganese in the Pacific Ocean, *Geochim. et Cosmo. Acta*, 51, 29-43

Mackey, D.J., J.E. O'Sullivan, R.J. Watson, 2002, Iron in the western Pacific: A riverine or hydrothermal source for iron in the Equatorial Undercurrent?, *Deep-Sea Res. I*, 49, 877-893

Martin, J.H. and R.M. Gordon, 1988, Northeast Pacific iron distributions in relation to phytoplankton productivity, *Deep-Sea Res. I*, 35, 177-196

Martin, J.H., R.M. Gordon, S. Fitzwater, W.W. Broenkow, 1989, VERTEX: phytoplankton/iron studies in the Gulf of Alaska, *Deep-Sea Res.*, 36, 649-680

Nakabayashi, S., K. Kuma, K. Sasaoka, S. Saitoh, M. Mochizuki, N. Shiga, and M. Kusakabe, 2002. Variation in iron(III) solubility and iron concentration in the northwestern North Pacific Ocean. *Limnol. Oceanogr.* 47: 885-892

Nakabayashi, S., M. Kusakabe, K. Kuma, and I. Kudo, 2001. Vertical distributions of Iron(III) hydroxide solubility and dissolved iron in the northwestern North Pacific Ocean. *Geophys. Res. Letters* 28: 4611-4614.

Nishioka, J., S. Takeda, I. Kudo, D. Tsumune, T. Yoshimura, K. Kuma, A. Tsuda, 2003, Size-fractionated iron distributions and iron-limitation processes in the subarctic NW Pacific, *GRL*, 30, NO. 14, 1730, doi:10.1029/2002GL016853.

Nishioka, J., T. Ono, H. Saito, K. Sakaoka, and T. Yoshimura (2011), Oceanic iron supply mechanisms which support the spring diatom bloom in the Oyasio region, western subarctic Pacific, *J. Geophys. Res.*, 116, C02021, doi:10.1029/2010JC006321

Nishioka, J., T. Ono, H. Saito, T. Nakatsuka, S. Takeda, T. Yoshimura, K. Suzuki, K. Kuma, S. Nakabayashi, D. Tsumune, H. Mitsudera, W. K. Johnson, A. Tsuda, (2007) Iron input into the western subarctic Pacific, importance of iron export from the Sea of Okhotsk, *Journal of Geophysical Research*, 112, C10012, doi:10.1029/2006JC004055.

Nishioka, J., Takeda, S., Wong, C., Johnson, W. 2001. Size-fractionated iron concentrations in the northeast Pacific Ocean: Distribution of soluble and small colloidal iron. *Mar. Chem.* 74, 157-179

Obata H., 1997. Development of an automated analytical method of iron in seawater and studies on the behavior of iron in the ocean. Kyoto University. Ph.D. Thesis, 109pp.

Obata, H., H. Karatani and E. Nakayama, 1993. Automated determination of iron in seawater by chelating resin concentration and chemiluminescence detection. *Analytical Chemistry*, 65: 1524-1528

Obata, H., H. Karatani, M. Matsui and E. Nakayama, 1997. Fundamental studies for chemical speciation of iron in seawater with an improved analytical method. *Marine Chemistry*, 56: 97-106

Roy, E., 2009. The detection and biogeochemistry of trace metals in natural waters. Ph.D. Thesis, University of Maine.

Rue, E.L. and K.W. Bruland, 1995. Complexation of iron(III) by natural organic ligands in the central North Pacific as determined by a new competitive ligand equilibration/adsorptive cathodic stripping voltammetric method. *Mar. Chem.* 50: 117-138.

Takata, H., Kuma, K., Saitoh, Y., Chikira, M., Saitoh, S., Isoda, Y., Takagi, S., Sakaoka, K., 2006. Comparing the vertical distribution of iron in the eastern and western North Pacific Ocean. *Geophys. Res. Lett.*, 33, L02613, doi:10.1029/2005GL024538, 2006.

Takata, H., K. Kuma, S. Iwade, Y. Yamajyoh, A. Yamaguchi, S. Takagi, K. Sakaoka, Y. Yamashita, E. Tanoue, T. Midorikawa, K. Kimura, J. Nishioka, 2004, Spatial variability of iron in the surface water of the northwestern North Pacific Ocean, *Mar. Chem.*, 86, 139-157

Takeda, S., H. Obata, 1995, Response of equatorial Pacific phytoplankton to subnanomolar Fe enrichment, *Mar. Chem.*, 50, 219-227

Tsuda, A., S. Takeda, H. Saito, J. Nishioka, Y. Nojiri, I. Kudo and others, 2003, A mesoscale iron enrichment in the western Subarctic Pacific induces a large centric diatom bloom, *Science*, 300, 958-961

Wu, J., E. Boyle, W. Sunda, and L.-S. Wen, 2001. Soluble and colloidal iron in the oligotrophic North Atlantic and North Pacific. *Science* 293: 847-849.