# Irreversibility of Marine Climate Change Impacts under Carbon Dioxide Removal

Xinru Li<sup>1</sup>, Kirsten Zickfeld<sup>2</sup>, Sabine Mathesius<sup>3</sup>, Karen E. Kohfeld<sup>2</sup>, and John Brian Robin Mathews<sup>4</sup>

<sup>1</sup>The University of British Columbia <sup>2</sup>Simon Fraser University <sup>3</sup>GEOMAR Helmholtz Centre for Ocean Research Kiel <sup>4</sup>Memorial University of Newfoundland

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#### Abstract

Artificial carbon dioxide removal (CDR) from the atmosphere has been proposed as a measure for mitigating climate change and restoring the climate system to a target state after exceedance ("overshoot"). This research investigates to what extent overshoot and subsequent recovery of a given cumulative CO2 emissions level by CDR leaves a legacy in the marine environment using an Earth system model. We use RCP2.6 and its extension to year 2300 as the reference scenario and design a set of cumulative emissions and temperature overshoot scenarios based on other RCPs. Our results suggest that the overshoot and subsequent return to a reference cumulative emissions level would leave substantial impacts on the marine environment. Although the changes in sea surface temperature, pH and dissolved oxygen are largely reversible, global mean values and spatial patterns of these variables differ significantly from those in the reference scenario when the reference cumulative emissions are attained.

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- <sup>4</sup> <sup>1</sup> Department of Geography, Simon Fraser University, Burnaby, B.C., Canada<sup>1</sup>
- <sup>5</sup> <sup>2</sup> School of Resource & Environmental Management, Simon Fraser University, Burnaby, B.C.,
- 6 Canada
- 7 <sup>3</sup> Université Paris-Saclay, Saint-Aubin, France
- 8 Corresponding author: Xinru Li (<u>xinru.li@alumni.ubc.ca</u>)

### 9 Key Points:

- Changes in sea surface temperature, pH and dissolved oxygen concentration in overshoot scenarios are largely reversible.
- Changes in average ocean values of these variables are not reversible centuries after the overshoot is removed.
- Spatial changes in these variables exhibit substantial differences between overshoot and non-overshoot scenarios in some regions.

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<sup>&</sup>lt;sup>1</sup> Xinru Li is now in the Department of Geography, University of British Columbia, Vancouver, B.C., Canada.

# 17 Abstract

- 18 Artificial carbon dioxide removal (CDR) from the atmosphere has been proposed as a measure
- 19 for mitigating climate change and restoring the climate system to a target state after exceedance
- 20 ("overshoot"). This research investigates to what extent overshoot and subsequent recovery of a
- given cumulative  $CO_2$  emissions level by CDR leaves a legacy in the marine environment using
- an Earth system model. We use RCP2.6 and its extension to year 2300 as the reference scenario
- and design a set of cumulative emissions and temperature overshoot scenarios based on other
   RCPs. Our results suggest that the overshoot and subsequent return to a reference cumulative
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- changes in sea surface temperature, pH and dissolved oxygen are largely reversible, global mean
- values and spatial patterns of these variables differ significantly from those in the reference
- 28 scenario when the reference cumulative emissions are attained.

# 29 Plain Language Summary

- 30 Commitments by countries to reduce emissions of carbon dioxide (CO<sub>2</sub>) fall short of what is
- 31 needed to limit global warming to below 2°C, a goal of the Paris Agreement, creating a risk that
- 32 the 2°C limit may be exceeded. Restoring global temperature to a target level after exceedance
- 33 ("overshoot") requires artificial carbon dioxide removal (CDR) from the atmosphere. Earlier
- 34 studies showed that changes in surface air temperature can be reversed by CDR, but climate
- 35 variables that take longer to respond to changes in atmospheric CO<sub>2</sub> are slower to reverse. In this
- 36 research we investigate to what extent the impacts of overshoot on the marine environment can
- be reversed with CDR. We show that the ocean responds slowly to a decrease in atmospheric
- CO<sub>2</sub> concentration by CDR, particularly for scenarios with large levels of overshoot. The
   overshoot results in substantial impacts on the marine environment for centuries with potentially
- 40 detrimental effects for marine ecosystems.

# 41 **1 Introduction**

In 2015 the Paris Climate Change Conference adopted the Paris Agreement that calls for ''holding the increase in the global average temperature to well below 2°C above pre-industrial levels'' (UNFCCC, 2015). Countries' current pledges for CO<sub>2</sub> emissions reductions are inconsistent with this goal (Rogelj et al., 2016; UNFCCC, 2016). Thus, unless greenhouse gas emissions decrease rapidly, limiting warming to well below 2°C without temporarily exceeding that level is unlikely.

48 CO<sub>2</sub>, the principal anthropogenic greenhouse gas, has a long atmospheric lifetime as its 49 concentration remains elevated for centuries to millennia following cessation of anthropogenic 50 emissions (Eby et al., 2009; Solomon et al., 2009). Several studies showed that the reduction of 51 CO<sub>2</sub> emissions to net zero can stabilize global warming, but not reverse it (Matthews & Caldeira, 52 2008; Lowe et al., 2009; Frölicher & Joos, 2010; Gillett et al., 2011; Zickfeld et al., 2013; Ehlert 53 & Zickfeld, 2017). Global mean temperature can be restored to a target level on geological 54 (>10,000 years) timescales, implying that global warming is irreversible on human timescales (decades to a century) (Eby et al., 2009; Solomon et al., 2009). Thus, carbon dioxide removal 55 56 (CDR) from the atmosphere, also referred to as "negative CO<sub>2</sub> emissions", has been proposed for 57 mitigating CO<sub>2</sub>-induced climate changes and restoring the climate system to a state that avoids 58 "dangerous" impacts (UNFCCC Article 2, 1992), if climate actions fail to reduce emissions sufficiently fast to comply with the "well below 2°C" climate limit. 59

60 Previous studies indicated that changes in surface air temperature due to anthropogenic

- 61  $CO_2$  emissions can be reversed through net-negative  $CO_2$  emissions (Cao & Caldeira, 2010;
- MacDougall, 2013; Jones et al., 2016; Tokarska & Zickfeld, 2015), while climate variables with long response timescales, for example ocean thermal expansion, exhibit a delay in their
- long response timescales, for example ocean thermal expansion, exhibit a delay in the
   responses to net-negative CO<sub>2</sub> emissions (Boucher et al., 2012; Bouttes et al., 2013b;
- MacDougall, 2013; Mathesius et al., 2015; Tokarska & Zickfeld, 2015; Ehlert & Zickfeld, 2018).
- 66 Mathesius et al. (2015) showed that delayed emissions reduction followed by implementation of
- 67 negative emissions at a likely unfeasible rate cannot entirely restore global mean ocean
- 68 temperature, dissolved oxygen and pH to their levels in a low-emission scenario. The lagged
- 69 behavior in ocean conditions implies that CDR could be ineffective at reversing climate change
- 70 impacts on the ocean, due to the ocean's long (millennial) response timescale.

71 This research aims to further investigate the reversibility of changes in ocean conditions 72 after the implementation of net-negative CO<sub>2</sub> emissions using an Earth System model of 73 intermediate complexity. We place the analysis in the context of emission scenarios that 74 temporarily exceed 2°C global warming relative to pre-industrial conditions and later stabilize 75 warming below 2°C. We compare the ocean response in such "overshoot" scenarios to that in a 76 reference scenario that limits warming to below 2°C with limited overshoot when the same 77 amount of cumulative CO<sub>2</sub> emissions is reached. We examine the change in ocean conditions 78 under different levels of surface air temperature and cumulative CO<sub>2</sub> emissions overshoot and 79 subsequent return to the levels in a reference low-emission scenario.

# 80 2 Model and Simulations

81 This study employs the University of Victoria Earth System Climate Model (UVic 82 ESCM), an Earth System Model of Intermediate Complexity (EMIC). This model consists of an 83 energy moisture balance model of the atmosphere with dynamical wind feedback coupled to a 84 three-dimensional ocean general circulation model and a dynamic/thermodynamic sea ice model 85 (Weaver et al., 2001). The physical climate model is coupled to a land surface model, a dynamic 86 terrestrial vegetation model, ocean inorganic and organic carbon cycle models and a sediment 87 model (Eby et al., 2009) (see Text S1 for further details).

88 To force the model a set of future  $CO_2$  emission scenarios (Figure 1a) is designed based 89 on the Representative Concentration Pathways (RCPs) and their extensions (EXPs) until year 2300 (Meinshausen et al., 2011). We devise a "reference" scenario that follows RCP2.6 and its 90 91 extension until year 2300, which is consistent with limiting global mean temperature increase to 92 2°C above pre-industrial levels in 80% of CMIP5 models (Collins et al., 2013). A set of 93 "overshoot" scenarios is designed based on the other RCPs (RCP4.5, RCP6 and RCP8.5), which 94 are modified in such a way that their cumulative CO<sub>2</sub> emissions temporarily exceed and then 95 converge to those of RCP/EXP2.6 in 2300 (Figures 1a and S1; see Text S2 for further details). 96 Land use change-related CO<sub>2</sub> emissions are prescribed to follow RCP2.6 and are the same for all 97 scenarios.

98 The overshoot scenarios are named according to the maximum net-negative CO<sub>2</sub> 99 emissions rate applied in each overshoot scenario: 5 GtC/yr, 9 GtC/yr and 15 GtC/yr in RCP4.5-100 CDR5, RCP6-CDR9 and RCP8.5-CDR15 respectively. Though negative emissions of 15 GtC/yr 101 are at the upper end of the range considered feasible (The National Academies Press, 2019), this 102 rate is used here to explore the implications of a large range of cumulative emissions overshoot. 103 Negative CO<sub>2</sub> emissions are applied to the model without specifying any particular CDR technology. The removed  $CO_2$  is assumed to be taken out of the ocean-atmosphere system

105 permanently (e.g., by storage underground).

106 The model was spun up for 10,000 years under year-850 conditions, following the

107 protocol for the EMIC model intercomparison project conducted in support of the Fifth

108 Assessment Report of IPCC (Eby, 2013). The long model spin-up practically eliminated any drift.

- 109 The model was then initialized from year-850 equilibrium state and forced with observed
- atmospheric  $CO_2$  concentration, radiative forcing from non- $CO_2$  greenhouse gases, sulphate
- aerosols, land-use changes, and natural forcings (orbital, solar and volcanic) to the year 2006.
- 112 This historical simulation provided the initial condition for a range of future simulations (2006-112 2000) foread with the CO emission according described above. In addition to CO
- 113 3000) forced with the  $CO_2$  emission scenarios described above. In addition to  $CO_2$  emissions, 114 radiative forcing from non- $CO_2$  greenhouse gases was prescribed to follow RCP2.6 and its
- extension to 2300 and was held fixed at year-2300 levels thereafter. Aerosol radiative forcing
- (direct effect) and land cover change followed RCP2.6 until 2100 and were held constant
- 117 thereafter. Natural forcings were specified as follows: orbital forcing was held fixed at year-2005
- 118 levels; solar irradiance was set to repeat the last solar cycle (1996-2008); and volcanic forcing
- 119 was set to zero. Non-CO<sub>2</sub> forcings are the same in all scenarios.

# 120 **3 Results**

121 3.1 Global Average Response

122 We analyze differences in climate variables between the reference scenario RCP2.6 and the overshoot scenarios, with a focus on year 2300 when the same cumulative CO<sub>2</sub> emissions are 123 124 achieved in all scenarios (Figure S1). The atmospheric CO<sub>2</sub> concentration in all scenarios 125 increases with net positive anthropogenic CO<sub>2</sub> emissions and starts to decline when CO<sub>2</sub> uptake 126 by natural carbon sinks exceeds net  $CO_2$  emissions. The  $CO_2$  concentration in the overshoot 127 scenarios is reduced to a level lower than that in RCP2.6 by 20 ppmv (RCP4.5-CDR5) to 53 128 ppmv (RCP8.5-CDR15) in 2300 (Figure 1b). This considerable difference in atmospheric CO<sub>2</sub> 129 concentration between RCP2.6 and the overshoot scenarios is due to inertia in the ocean carbon 130 cycle response (Figure S2), which lags the decline in atmospheric  $CO_2$ . The initial rise in 131 atmospheric CO<sub>2</sub> causes surface air temperature (SAT) to increase. Implementation of net-132 negative emissions leads to a decline in SAT in the overshoot scenarios, with SAT being warmer 133 than in RCP2.6 by 0.1°C (RCP4.5-CDR5) to 0.3°C (RCP8.5-CDR15) in 2300 (Figure 1c). This 134 result indicates that emission pathways with larger overshoot result in slightly different SAT 135 despite the same amount of cumulative CO<sub>2</sub> emissions.

136 Globally averaged sea surface temperature (SST) increases with the rising  $CO_2$ 137 concentration and starts to decline 15 years (RCP8.5-CDR15) to 50 years (RCP4.5-CDR5) after 138 the decline in atmospheric CO<sub>2</sub>. In 2300, SST in the overshoot scenarios exceeds that in RCP2.6 139 by 0.1°C (RCP4.5-CDR5) to 0.3°C (RCP8.5-CDR15) (Figure 1d). Global average ocean 140 temperature (OT) increases during the whole simulation in RCP2.6 (Figure 1h). In the overshoot 141 scenarios, OT peaks and declines following implementation of net negative emissions, and then 142 increases again. The decline of OT lags the reduction in atmospheric CO<sub>2</sub> concentration by about 143 120 years (RCP8.5-CDR15) to 180 years (RCP4.5-CDR5). OT in the overshoot scenarios is 144 warmer than in RCP2.6 by 0.2°C (RCP4.5-CDR5) to 0.6°C (RCP8.5-CDR15) in 2300 (Figure 145 1h). Sea level rise due to ocean thermal expansion (i.e. thermosteric sea level rise) shows similar

lagged responses as OT, with larger thermosteric sea level rise in the overshoot scenarios relative
to RCP2.6 by 12 cm (RCP4.5-CDR5) to 38 cm (RCP8.5-CDR15) in 2300 (Figure S3).

148 Changes in dissolved oxygen concentration (DO) are determined by changes in ocean 149 temperature, which affect oxygen solubility in sea water, ocean ventilation and biological 150 processes, which affect the production and consumption of DO. Sea surface DO declines with 151 rising SST and recovers after implementation of net negative CO<sub>2</sub> emissions, reaching a level 152 close to 2.0% below pre-industrial in 2300 in all scenarios (Figure 1e). Global average DO in 153 RCP2.6 decreases over the whole simulation (Figure 1i). In the overshoot scenarios, global 154 average ocean DO continues to decline for 120 years (RCP8.5-CDR15) to 190 years (RCP4.5-155 CDR5) after the peak in atmospheric  $CO_2$  concentration, and then temporarily increases before 156 declining again (Figure 1i). In 2300, the decrease in global average ocean DO ranges from 7.9 % 157 (RCP4.5-CDR5) to 8.9 % (RCP8.5-CDR15) below preindustrial, compared to 6.7% in RCP2.6 158 (Figure 1i). After year 2300, global average ocean DO in the overshoot scenarios decreases again 159 until the end of the simulation, with the DO decline rate varying between scenarios (Figure 1i). 160 Interestingly, the rate of global average DO decline is lowest in the scenario with the largest 161 overshoot (RCP8.5-CDR15) (Figure 1i), possibly caused by more vigorous ocean circulation 162 (Figure S4, S5). 163 Sea surface pH drops with rising atmospheric  $CO_2$  and recovers when the  $CO_2$ 164 concentration declines (Figure 1b, 1f). In 2300, the decrease in sea surface pH below

preindustrial in the overshoot scenarios is smaller than that in RCP2.6 by 0.01 units (RCP4.5CDR5) to 0.03 units (RCP8.5-CDR15) (Figure 1f), which reflects the lower atmospheric CO2
concentration in the overshoot scenarios discussed previously. Global average ocean pH in
RCP2.6 decreases slightly over the whole simulation (Figure 1g). In the overshoot scenarios,

global average ocean pH reaches a minimum between years 2190 (RCP8.5-CDR15) and 2210
 (RCP4.5-CDR5) and then recovers. In 2300, the decrease in global average ocean pH ranges

from 0.10 units (RCP4.5-CDR5) to 0.12 units (RCP8.5-CDR15) below preindustrial, compared

to 0.08 units in RCP2.6 (Figure 1g). The slower recovery of entire ocean pH compared to sea

surface pH in 2300 can be attributed to slow upward mixing of excess  $CO_2$  and enhanced ocean

174 stratification, which further slows the release of  $CO_2$  to the atmosphere and the reversal of pH

175 decline in the ocean interior.

176



177 178 Figure 1. Time series of global mean variables for the reference (RCP2.6) and overshoot scenarios. a, net 179 anthropogenic  $CO_2$  emissions rate (GtC·yr<sup>-1</sup>) (defined as  $CO_2$  emissions from fossil fuels and land-use 180 change minus CO<sub>2</sub> removals). RCP scenarios are shown for comparison (dashed lines). **b**, atmospheric 181  $CO_2$  concentration (ppmv). c, surface air temperature change (°C). d, sea surface temperature change (°C). 182 e, sea surface dissolved oxygen concentration change (%). f, sea surface pH change (total pH scale). h, 183 global average ocean temperature change (°C). i, global average ocean dissolved oxygen concentration 184 change (%). g, global average ocean pH change. Changes in panels c-g are relative to year 1800. Note that 185 panel **a** has a different horizontal scale than the other panels.

186 3.2 Regional Ocean Impacts

At the time the cumulative CO<sub>2</sub> emissions overshoot is removed (year 2300), the spatial distribution of sea surface variables (temperature, dissolved oxygen concentration and pH) exhibits large differences between the overshoot scenarios and the reference scenario RCP2.6, despite similar global mean sea surface values.

In year 2300, most sea surface areas are warmer relative to preindustrial except the North
 Atlantic south of Greenland and the Weddell Sea where SST is cooler (Figure S6a-d). Large sea
 surface areas are warmer in the overshoot scenarios than in RCP2.6 in 2300, particularly in the

194 Southern Ocean between 40°S and 60°S (Figure 2a-c). The larger warming in this region is

- associated with stronger Southern Hemisphere westerlies (driven by a stronger surface air
- temperature gradient between 30°S and 60°S; Figure S7), leading to a more vigorous meridional
- 197 overturning circulation and enhanced subduction of surface waters around 60°S (Fyfe et al.,
- 198 2007). In contrast, SST is cooler in the North Atlantic south of Greenland in 2300 relative to
- 199 RCP2.6 (Figure 2a-c). The cooler SST is driven by a weakening of the Atlantic meridional
- 200 overturning circulation (AMOC) (Figure S8), which results in reduced northward heat transport 201 in the LIVie Model (Searke et al. 2004)
- 201 in the UVic Model (Saenko et al., 2004).
- Large areas of the interior ocean are warmer relative to preindustrial in 2300 (Figure S6eh), with larger warming in the overshoot scenarios than in RCP2.6, particularly at 200-2,000 m
- 204 depth in the latitudinal band of 60°S to 60°N (Figure 2d-f). Heat enters the interior ocean in areas
- 205 of North Atlantic Deep Water (NADW) formation and around 60°S where surface waters are
- subducted to intermediate depth and then spread meridionally along isopycnals. Because of the
- 207 larger interior ocean warming in the overshoot scenarios and long timescale of ocean mixing, it
- takes longer for the excess heat to be released. Faster warming of the sea surface relative to the
- 209 interior and weaker AMOC contribute to ocean stratification that slows the reversal of ocean
- warming further. A larger temperature increase thus persists below the subsurface to a depth of
- about 2,000 m relative to RCP2.6 when the cumulative emissions overshoot is removed.

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Figure 2. Spatial distribution of ocean temperature anomalies relative to the reference scenario RCP2.6 in 214 2300. a-c, latitude-longitude distribution of sea surface temperature anomaly (°C). d-f, zonal mean 215 anomaly of ocean temperature (°C). a&d, RCP4.5-CDR5 minus RCP2.6. b&e, RCP6-CDR9 minus 216 RCP2.6. c&f, RCP8.5-CDR15 minus RCP2.6.

217 In year 2300, large sea surface areas show a decrease in DO concentration relative to 218 preindustrial except the North Atlantic south of Greenland and the polar regions in both 219 hemispheres, which exhibit DO increase (Figure S9a-d). In the North Atlantic, the sea surface 220 DO concentration in the overshoot scenarios is larger than in RCP2.6 in 2300 (Figure 3a-c),

221 mainly driven by the lower SST in that region (Figure 2a-c). The DO concentration is also larger in the Southern Ocean south of 60°S, particularly in the Weddell Sea (Figure 3a-c). The decline
in sea ice area south of 60°S relative to preindustrial is larger in the overshoot scenarios than in
RCP2.6 in 2300 (Figure S10). The larger sea ice decline could lead to stronger air-sea oxygen
exchange and thus a higher DO concentration south of 60°S. The sea surface DO concentration
decreases between 50°S and 60°S relative to RCP2.6 (Figure 3a-c), largely caused by the larger
SST increase in this region (Figure 2a-c).

228 Changes in DO concentration relative to preindustrial are larger in the overshoot 229 scenarios in vast parts of the interior ocean in 2300 (Figure S9e-h), with both negative and 230 positive DO anomalies relative to RCP2.6 (Figure 3d-f). The DO concentration is larger than in 231 RCP2.6 between 60°S and 60°N at 200-1,400 m depth (particularly in the latitudinal bands of 232  $40^{\circ}$ N to  $60^{\circ}$ N and  $40^{\circ}$ S to  $60^{\circ}$ S). The DO increase between  $40^{\circ}$ N and  $60^{\circ}$ N largely occurs in the 233 North Pacific (Figure S11), where the meridional overturning circulation strengthens and 234 ventilation is enhanced in the overshoot scenarios relative to RCP2.6 (Figure S12, S13). The 235 enhanced overturning in the North Pacific has previously been shown to occur in response to 236 weakened AMOC in the UVic ESCM (Saenko et al., 2004; Zickfeld et al., 2008). Changes in 237 biological processes in the North Pacific are inconsistent with the positive DO anomaly at mid 238 depth, suggesting that the dominant driver of this anomaly is increased ventilation. Net primary 239 production increases in the eastern North Pacific leading to increased detrital export and 240 remineralization (Figure S14-17), which are expected to cause a decrease in DO concentration 241 and an increase in nutrient concentration at depth, both of which are opposite to the response 242 simulated by the model (Figure3d-f, S17). The enhanced ventilation, on the other hand, 243 transports more oxygen to depth and more nutrients into the surface layer, consistent with model 244 response (Figure S18). The DO increase at intermediate depths between  $40^{\circ}$ S and  $60^{\circ}$ S is 245 associated with the increased meridional overturning circulation in the Southern Ocean (Figure 246 S19). The Southern Hemisphere westerlies strengthen relative to RCP2.6 in 2300 (Figure S7), 247 leading to stronger subduction of surface waters and subsequent northward transport along 248 isopycnals. This process transports oxygen from the sea surface to intermediate depths, thereby 249 increasing the DO concentration of intermediate waters north of 60°S relative to RCP2.6 in 2300. 250 The DO concentration declines at 1,500-3,000 m depth around 60°N relative to RCP2.6 251 in 2300 (Figure 3d-f) due to a weaker AMOC (Figure S8b-e), which results in reduced 252 ventilation of deep waters in regions where NADW is formed. The DO concentration also decreases at depth south of 60°S, potentially driven by the reduced Antarctic Bottom Water 253 254 formation and inflow of NADW with reduced DO concentration that results from reduced deep-255 water formation in the North Atlantic relative to RCP2.6 in 2300 (Figure S8, 19). Enhanced

- remineralization of interior ocean detritus could also contribute to the DO decrease (FigureS15,16).
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259 260 Figure 3. Spatial distribution of ocean dissolved oxygen (DO) concentration anomaly relative to the 261 reference scenario RCP 2.6 in 2300. a-c, latitude-longitude distribution of sea surface DO concentration 262 anomaly (µmol/L). d-f, zonal mean anomaly of ocean DO concentration (µmol/L). a&d, RCP4.5-CDR5 263 minus RCP2.6. b&e, RCP6-CDR9 minus RCP2.6. c&f, RCP8.5-CDR15 minus RCP2.6.

264 Ocean pH declines in large areas both at the sea surface and at depth relative to 265 preindustrial in 2300, particularly in the Arctic (Figure S20). At the sea surface, the pH decrease 266 in the overshoot scenarios is smaller than in RCP2.6 (consistent with the slightly lower 267 atmospheric CO<sub>2</sub> concentration), except in the Arctic where a larger pH decrease is evident in the 268 overshoot scenarios (Figure 4a-c). In the interior ocean, the pH decrease relative to RCP2.6 is

largely located at 300-2,000 m depth in the latitudinal band of 45°S to 45°N and in the Arctic in
2300 (Figure 4d-f).

271 Ocean pH declines with increasing carbon uptake by the ocean. Regional patterns of 272 changes in ocean pH correspond to spatial patterns of changes in dissolved inorganic carbon 273 (DIC) concentration (Figure 4d-f, S21). In 2300, DIC concentration remains higher in the overshoot scenarios than in RCP2.6 between 45°S and 45°N and in the Arctic (Figure S21, S22). 274 275 The higher DIC concentration is associated with the slow upward mixing of excess carbon and 276 enhanced stratification relative to RCP2.6, the same processes that delay the reversal of OT and 277 DO changes. Lower pH in the Arctic relative to RCP2.6 in 2300 is attributed to the difference in 278 sea ice area. During the period of rising surface air temperature, Arctic sea ice area declines more 279 strongly in the overshoot scenarios than in RCP2.6 (Figure S23), allowing for larger CO<sub>2</sub> uptake. 280 Arctic sea ice recovers after surface air temperature starts to decline, which hampers the release of CO<sub>2</sub> back to the atmosphere. Even though the sea ice area is smaller in the overshoot scenarios 281 than in RCP2.6 in 2300 (Figure S24), the larger DIC anomaly persists. 282

283 The pH decrease lowers the aragonite saturation state, which influences the calcification 284 rates of marine calcifying organisms. The aragonite saturation horizon shoals substantially relative to preindustrial, especially in the Arctic, due to soaring atmospheric CO<sub>2</sub> (Figure 4d-f). 285 286 This shoaling is larger in the overshoot scenarios than in RCP2.6, with greater shoaling in 287 scenarios with higher cumulative emissions overshoot. When the cumulative emissions 288 overshoot is removed (year 2300), the aragonite saturation horizon in the overshoot scenarios is 289 largely restored to the level in RCP2.6 except in the Arctic where it shoals to about 100 m 290 (RCP8.5-CDR15) to 900 m (RCP4.5-CDR5) below the surface, compared to 1400 m in RCP2.6 291 (Figure 4d-f). The difference in shoaling of the aragonite saturation horizon results from the 292 strong pH decrease relative to RCP2.6 in the Arctic in 2300.

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294 295 Figure 4. Spatial distribution of ocean pH anomaly relative to the reference scenario RCP2.6 in 2300. a-c, 296 latitude-longitude distribution of sea surface pH anomaly. **d-f**, zonal mean anomaly of ocean pH. **a&d**, 297 RCP4.5-CDR5 minus RCP2.6. b&e, RCP6-CDR9 minus RCP2.6. c&f, RCP8.5-CDR15 minus RCP2.6. 298 The black and yellow dashed lines in panels d-f represent the aragonite saturation horizon under pre-299 industrial conditions and in RCP2.6 in 2300, respectively. The purple dashed lines represent the aragonite



#### 301 4 Discussion and Conclusions

302 Our results suggest that net negative CO<sub>2</sub> emissions allow for a fast reversal of 303 atmospheric CO<sub>2</sub> concentration, global mean surface air temperature and sea surface conditions 304 in scenarios where a given cumulative CO<sub>2</sub> emissions and hence temperature limit is first 305 exceeded. These global average results, however, hide substantial regional differences: for 306 instance, SSTs are about 2°C warmer in the Antarctic Circumpolar Current, and more than 2°C 307 colder in the North Atlantic in the scenario with the largest degree of overshoot relative to the 308 reference scenario RCP2.6. Furthermore, large differences exist in average ocean values of 309 temperature, dissolved oxygen concentration and pH, with larger differences for scenarios with 310 higher levels of overshoot. There are also significant differences in the interior ocean distribution 311 of these variables. In 2300, ocean temperature is up to 2°C warmer in the subsurface tropical 312 ocean and pH up to 0.2 units lower in the Arctic relative to RCP2.6. These results suggest that 313 negative  $CO_2$  emissions are ineffective at reversing changes in the marine environment on 314 human timescales (decades to a century), particularly following high levels of cumulative  $CO_2$ 315 emissions overshoot.

316 The fast reversal of atmospheric  $CO_2$  and SAT are in agreement with previous studies 317 exploring the Earth system response to negative CO<sub>2</sub> emissions (Cao & Caldeira, 2010; Tokarska 318 & Zickfeld, 2015; Jones et al., 2016). The reversal of changes in sea surface variables on decadal 319 timescales and in global ocean variables on multi-centennial timescales in response to net 320 negative anthropogenic emissions are consistent with the findings of Mathesius et al. (2015) and 321 can be expected to be robust across models, as they are in line with our understanding of the 322 different timescales that govern uptake of heat and  $CO_2$  by the ocean (Archer et al., 1997). The 323 spatial distribution of the differences of ocean temperature, pH and DO concentration in RCP8.5-324 CDR15 relative to RCP2.6 is broadly in agreement with that shown in Mathesius et al. (2015), 325 with regional differences due to different responses in ocean circulation. A salient feature of the 326 UVic ESCM employed for this study is the onset of convection in the North Pacific following 327 weakening of the meridional overturning circulation in the Atlantic. As Saenko et al. (2004) 328 argues, this effect is physically plausible and is consistent with climate reconstructions.

329 The slow upward mixing of heat and gases (e.g.  $CO_2$ ,  $O_2$ ), weakened overturning 330 circulation and increased stratification are important factors that hamper reversal of conditions in 331 the ocean's interior to the state in RCP2.6. Due to the coarse resolution of the model employed in 332 this study, diapycnal mixing and deep-water formation are heavily parameterized, leading to 333 potential biases in the recovery timescale. Another possible source of bias is the lack of a 334 dynamic land ice module in the UVic ESCM v2.9. The climate system response of melting land 335 ice and continental ice sheets that produce freshwater input causing weakening of global 336 overturning circulation is not included in our analysis. Previous studies suggest that land ice 337 would continue to melt for centuries to millennia (Lemke et al., 2007), slowing the recovery of 338 the overturning circulation. The overestimated AMOC recovery timescale possibly leads to 339 overestimation of the degree of reversibility of changes in ocean conditions. The emission 340 scenarios used in this study are idealized and may not be consistent with emissions from 341 different gases. Radiative forcing from non-CO<sub>2</sub> greenhouse gases and aerosols follows RCP 2.6 342 in all scenarios, irrespective of the CO<sub>2</sub> emission rate in individual scenarios. As many gases are 343 co-emitted with CO<sub>2</sub> (Rogelj et al., 2014), non-CO<sub>2</sub> forcing would likely be higher in the 344 overshoot scenarios, implying larger ocean heat uptake during the overshoot phase and delayed

345 recovery of the interior ocean temperature and DO concentration.

346 The larger changes in marine conditions in the overshoot scenarios relative to RCP2.6 in 347 2300 imply potentially detrimental effects on marine ecosystems. The warmer ocean could 348 fundamentally affect biological processes, as most biological rates are temperature dependent 349 (Eppley, 1972). Most marine species are sensitive to thermal stress (Gattuso et al., 2015). The 350 increasing temperature in the Tropics could shift species distributions poleward, thereby 351 reducing biodiversity in that region (Thomas et al., 2012). While DO increase in the tropical 352 Indian Ocean and the Southern Pacific could alleviate hypoxia in these regions, the DO decrease 353 in some coastal regions of the tropical Atlantic and Southern Ocean (Figure S11) could 354 exacerbate hypoxia leading to mortality and habitat compression (Mislan et al., 2017). The 355 reduced ocean pH leads to shoaling of the aragonite saturation horizon, particularly in the Arctic, 356 which enhances the vulnerability of organisms that build shells or other structures of aragonite,

357 such as deep-sea corals (Cao et al., 2014; AMAP Assessment, 2013).

358 We conclude that cumulative  $CO_2$  emissions overshoot and subsequent return to a climate 359 target state would leave a substantial legacy in the marine environment for centuries, particularly

- 360 for scenarios with large levels of overshoot. Thus, our findings support the view that early  $CO_2$
- 361 emissions involve lower risks for the marine environment than delayed emissions reduction
- followed by CDR (Seneviratne et al., 2018), as CDR cannot entirely restore the climate system to
- a state reached following a low emission scenario.

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495

Figure 1 in the manuscript.



Figure 2 in the manuscript.



90

60

30

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-30

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-90

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-180

latitude (°)

Figure 3 in the manuscript.



Figure 4 in the manuscript.







 $\Delta$  sea surface pH

