LETTERS
PUBLISHED ONLINE: 2 NOVEMBER 2015 | DOI: 10.1038/NCLIMATE2844

nature climate change

# Abrupt onset and prolongation of aragonite undersaturation events in the Southern Ocean

Claudine Hauri[1,2*†], Tobias Friedrich[1†] and Axel Timmermann[1]

**Ocean acidification may lead to seasonal aragonite undersaturation in surface waters of the Southern Ocean as early as 2030 (ref. 1). These conditions are harmful to key organisms such as pteropods[2], which contribute significantly to the pelagic foodweb and carbon export fluxes in this region[3]. Although the severity of ocean acidification impacts is mainly determined by the duration, intensity and spatial extent of aragonite undersaturation events, little is known about the nature of these events, their evolving attributes and the timing of their onset in the Southern Ocean. Using an ensemble of ten Earth system models, we show that starting around 2030, aragonite undersaturation events will spread rapidly, affecting ∼30% of Southern Ocean surface waters by 2060 and >70% by 2100, including the Patagonian Shelf. On their onset, the duration of these events will increase abruptly from 1 month to 6 months per year in less than 20 years in >75% of the area affected by end-of-century aragonite undersaturation. This is likely to decrease the ability of organisms to adapt to a quickly evolving environment[4]. The rapid equatorward progression of surface aragonite undersaturation can be explained by the uptake of anthropogenic $CO_2$, whereas climate-driven physical or biological changes will play a minor role.**

The Southern Ocean (south of 40° S) ecosystem plays a fundamental role in global biogeochemical cycling through its effect on nutrient distributions and the air–sea balance of $CO_2$ (refs 5,6). Despite its remoteness, this region also hosts valuable krill and toothfish fisheries[7]. Recent trends in the Southern Ocean foodweb[8], which can be linked partly to regional ocean warming and sea-ice retreat[9], prompt the concern that further progression of anthropogenic stressors on sensitive marine organisms can have ripple effects far beyond the Southern Ocean. One key threat to Southern Ocean biota is the rapid progression of ocean acidification[2,10,11], which is caused by the uptake of anthropogenic $CO_2$. Along with a decreasing pH, the uptake of anthropogenic $CO_2$ decreases the $CO_3^{2-}$ concentration and thereby the saturation state ($\Omega$) of the $CaCO_3$ minerals aragonite (arag), calcite (calc) and magnesian calcite. These minerals chemically dissolve once $\Omega$ decreases below the well-established thermodynamic threshold of $\Omega = 1$. Many marine calcifiers are sensitive to a decreasing $\Omega$ in the ocean and develop species-dependent responses already well above this thermodynamical threshold[12]. For example, aragonite-forming organisms such as soft clams and pteropods exhibit a negative net calcification rate at $\Omega_{arag} \sim 1.5$ (ref. 12) and close to 1 (ref. 2), respectively. In the following, we will refer to these species-dependent thresholds as biological thresholds.

Here, we use monthly output from ten Earth system models from the Coupled Model Intercomparison Project, Phase 5 (CMIP5, see Methods) to study the history and future development of Southern Ocean low-$\Omega_{arag}$ and -$\Omega_{calc}$ events. These CMIP5 models are the most advanced, global, state-of-the-art climate–carbon-cycle models. By using a multi-model ensemble we expect to add robustness to our analysis as potential shortcomings of individual models are of less consequence for the overall results[13]. Our analysis focuses mostly on aragonite undersaturation events, as aragonite is the most soluble $CaCO_3$ mineral and because $\Omega_{arag} = 1$ closely lines up with the biological threshold of pteropods. Aragonite undersaturation in sea water can occur sporadically and naturally as a result of background variability; superimposed on this natural variability is the long-term ocean acidification trend[14,15]. The Southern Ocean is at particular risk of becoming undersaturated with respect to aragonite in the near future[1,11], as thermodynamics and upwelling of $CO_2$-rich deep waters cause a naturally low $\Omega_{arag}$ environment[16]. The duration of these aragonite undersaturation events is an important indicator for the survival chances of organisms sensitive to these conditions, such as pteropods, as it quantifies how long these organisms will be exposed to lower calcification and increased dissolution rates, higher energetic cost, and suppressed metabolism, eventually leading to reduced growth and reproduction[17,18]. Furthermore, the rates at which the intensity and duration of aragonite undersaturation events change are crucial, as they may be faster than the evolutionary processes that could eventually lead to adaptation to low-$\Omega_{arag}$ habitats[4]. Compared with previous studies that were based on annual mean values of $\Omega_{arag}$ simulated by ocean-only models[11] or the extrapolation of relatively sparse measurements of Southern Ocean near-surface carbon-cycle parameters[1], monthly output from the ten CMIP5 coupled Earth system models employed in our study enables us to explore the spatial characteristics and temporal evolution of the habitat of pteropods and other sensitive organisms with unprecedented detail and robustness, and in the presence of natural climate variability and greenhouse warming.

Under the high-emissions Representative Concentration Pathway 8.5 (RCP8.5; ref. 19, see Methods), the ensemble mean of ten CMIP5 models documents that the duration and spatial extent of aragonite undersaturation events in the Southern Ocean will change rapidly over the next 40 years (Figs 1 and 2). According to the modelling results, regions in the Bellingshausen and Ross seas already experience sporadic short surface aragonite undersaturation events under present-day conditions (Fig. 1a). Although such short events are masked out in multi-year data products such as the Global Ocean Data Analysis Project (Supplementary Fig. 5a and reference in figure caption) as well as in the comparable ten-year ensemble means (Supplementary Fig. 5b), observations and salinity-based estimates of surface $\Omega_{arag}$ from these regions sometimes attain values near 1, thus supporting the model results[20,21]. The absence of simulated surface aragonite undersaturation events before 1965

[1]International Pacific Research Center (IPRC), SOEST, University of Hawai'i, Honolulu, Hawaii 96822, USA. [2]International Arctic Research Center (IARC), University of Alaska Fairbanks, Fairbanks, Alaska 99775, USA. †These authors contributed equally to this work. *e-mail: chauri@hawaii.edu

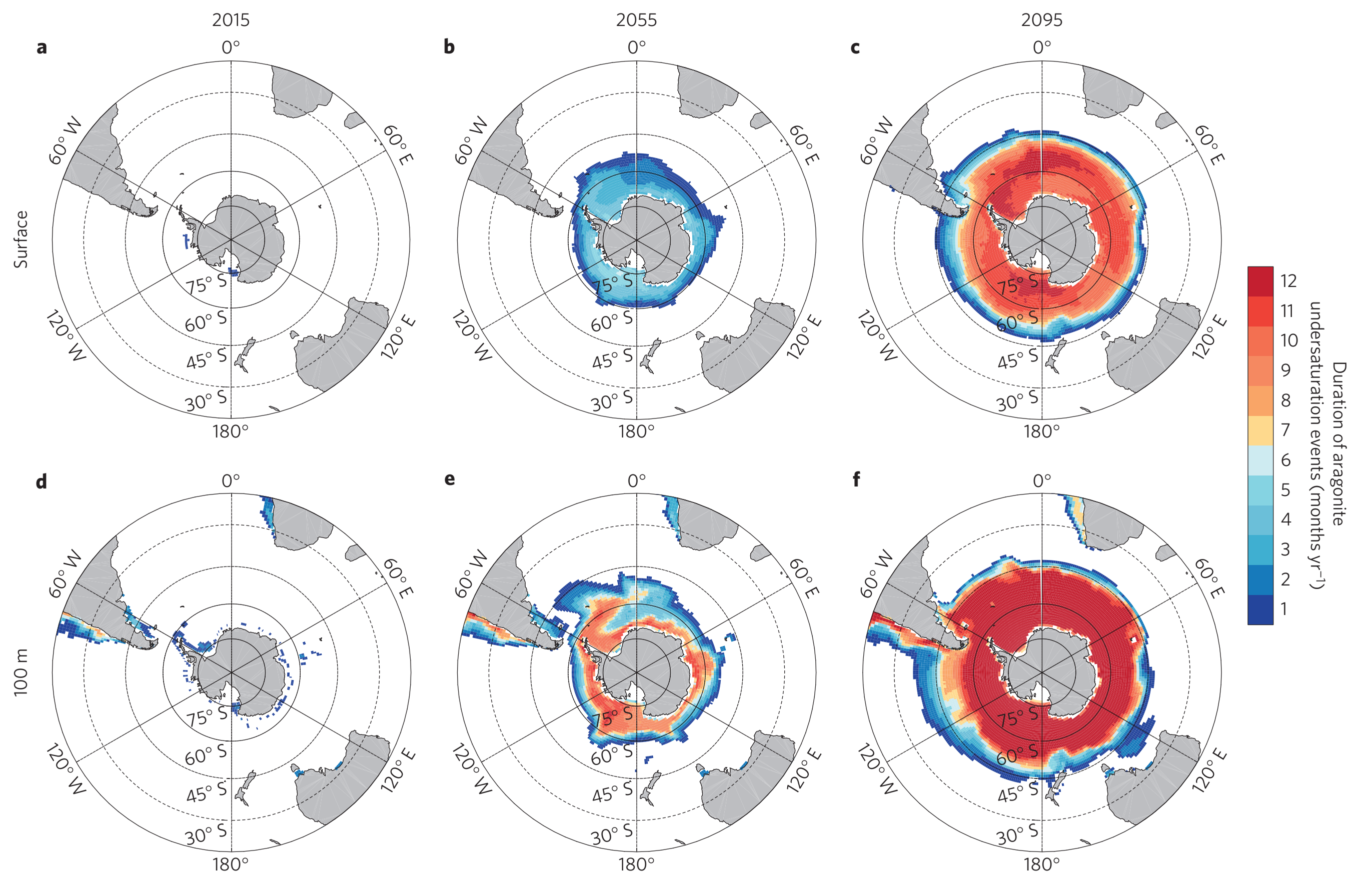


**Figure 1 | Duration of aragonite undersaturation events in months per year. a–f**, Decadal average of ensemble-mean duration of aragonite undersaturation events at the surface at present day (**a**) and around 2055 (**b**) and 2095 (**c**), and at 100 m at present day (**d**) and around 2055 (**e**) and 2095 (**f**). Duration was rounded to the closest integer. See Supplementary Fig. 2 for pre-industrial and additional years.

indicates that their present occurrence may already be a result of the uptake of anthropogenic $CO_2$ (Supplementary Fig. 2a).

The spatial extent of surface aragonite undersaturation events is projected to rapidly increase between 2035 and 2040 across the Southern Ocean, beginning in western Antarctica and quickly expanding towards eastern Antarctica (Supplementary Fig. 2c,d). Following the RCP8.5 $CO_2$ emission scenario, surface aragonite undersaturation events are projected to cover >70% ($\sigma = \pm 6\%$) of the Southern Ocean by the end of the century (Fig. 2a, see Supplementary Fig. 3 for other $\Omega_{arag}$ and $\Omega_{calc}$ thresholds).

The duration of surface aragonite undersaturation events also increases rapidly. By 2055, areas south of 60° S are projected to experience 1–5-month-long aragonite undersaturation events; by the end of the century, these events are projected to last anywhere between 8 and 12 months per year (Fig. 1b,c; see Supplementary Fig. 4 for regional duration of events with other $\Omega_{arag}$ and $\Omega_{calc}$ thresholds). Areas with important fisheries as far north as the southern tip of New Zealand, southern Chile, and the Patagonian Shelf will also experience surface aragonite undersaturation events by 2095. Although these surface aragonite undersaturation events are projected to be relatively short in southern New Zealand, they will last up to ten months in southern Chile and on the Patagonian Shelf according to the CMIP5 Earth system model simulations. There socio-economically important organisms such as molluscs, echinoderms and copepods may be directly affected and harmed by these long-lasting aragonite undersaturation events[12,22]. The vulnerability of copepods may have additional negative effects, as they comprise an essential part of the diet of anchovetas[23], which are an important harvest, and a key species in the diet of marine mammals, seabirds and carnivorous fish in central–southern Chile and on the Patagonian Shelf[24,25].

Pteropods are abundant down to depths of 200 m (ref. 3) and will experience prolonged aragonite undersaturation events even earlier because $\Omega_{arag}$ naturally decreases with depth. The historical model simulation suggests that sporadic, confined and short aragonite undersaturation events at 100 m depth were a natural phenomenon in the Ross and Weddell seas around 1900 (Supplementary Fig. 2e), most likely due to the upwelling of $CO_2$-rich Circumpolar Deep Water[26]. Similarly, sporadic coastal upwelling of corrosive waters in the Humboldt and Benguela current systems and onto the Patagonian Shelf may have also caused brief natural aragonite undersaturation episodes at 100 m depth during times when the atmospheric $CO_2$ was still close to pre-industrial levels (Fig. 2a, inset). These events lasted between 1 and 2 months per year around Antarctica, in the Benguela Current System and on the Patagonian Shelf, and up to 6 months per year in southern Chile. However, as a result of the ocean's uptake of anthropogenic $CO_2$, these subsurface aragonite undersaturation events are simulated to occur in areas all around Antarctica under present-day conditions, and are projected to become perennial features by 2055 (Fig. 1e). By the end of the twenty-first century, the models also show year-round aragonite undersaturation at 100 m depth in large parts on the Patagonian Shelf and along the Chilean coast (Fig. 1f).

The duration of aragonite undersaturation events changes abruptly (Fig. 2b), with implications for the adaptation potential of organisms with longer lifespans, such as pteropods that live between 1 and 3 years[27]. In more than 75% of the area that will be aragonite undersaturated by the end of the century, the transition time (time between the first occurrence of aragonite undersaturation events

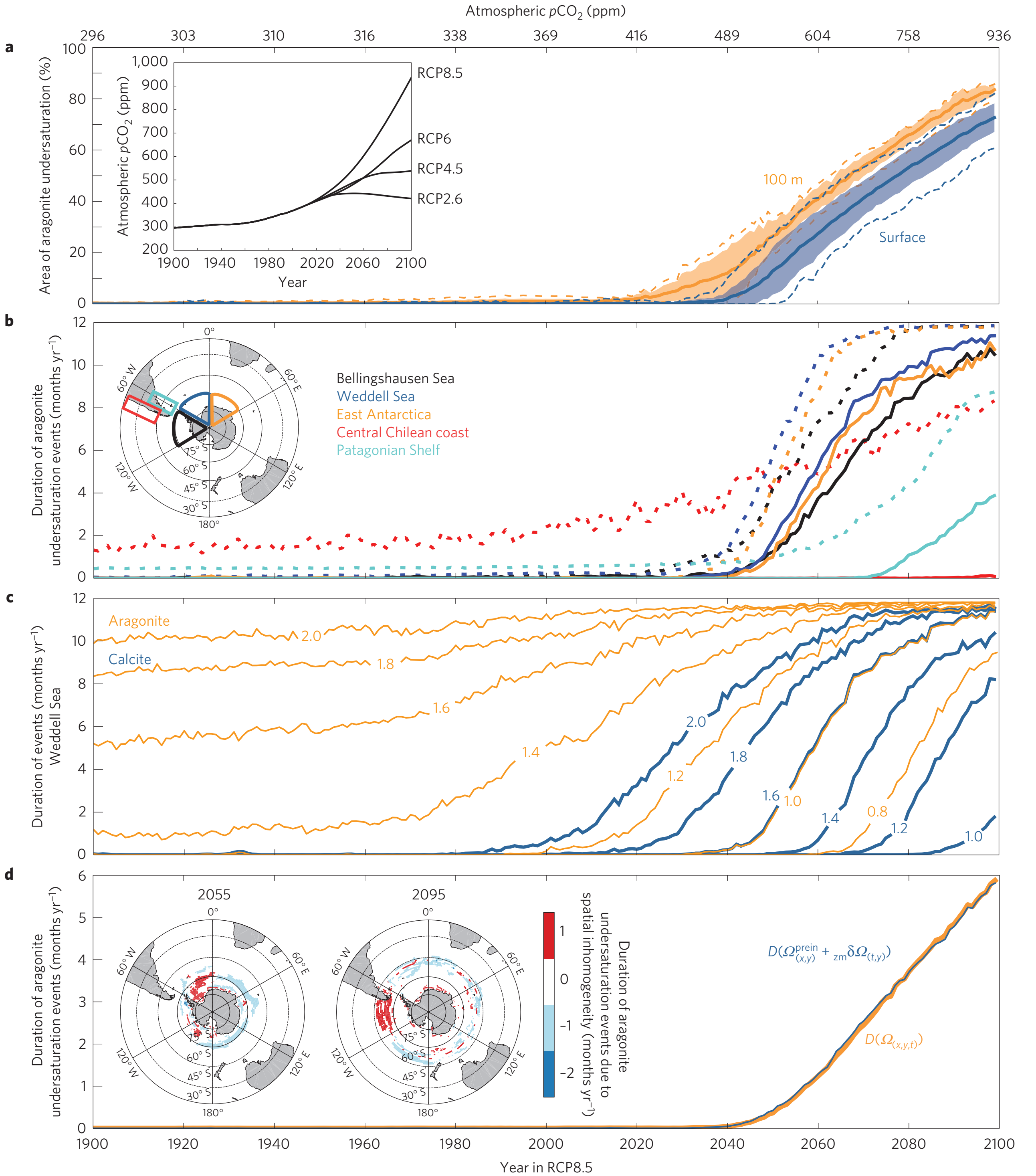


**Figure 2 | Spatial extent and regional progression of aragonite undersaturation events. a**, Area-weighted mean of the spatial extent of areas in the Southern Ocean (south of 40° S), undersaturated with regard to aragonite at the surface (blue) and at 100 m depth (orange), as a function of time following historical and the high-emissions Representative Concentration Pathway 8.5 (RCP8.5 (ref. 19), bottom *x* axis) and atmospheric $CO_2$ concentration (top *x* axis). Solid lines, ensemble mean; shaded areas, $\pm 1\sigma$; dashed lines, maximum and minimum of ensemble. Inset shows atmospheric $CO_2$ concentration as a function of time for RCP2.6, RCP4.5, RCP6 and RCP8.5. **b**, Area-weighted ensemble-mean duration of aragonite undersaturation events at the surface (solid lines) and at 100 m depth (dashed lines) for the Weddell Sea (blue, south of 60° S, 0°–60° W), Bellingshausen Sea (black, south of 60° S, 60°–120° W), East Antarctica (orange, south of 60° S, 0°–60° E), central Chilean coast (red, 15°–45° S, 68°–78° W) and Patagonian Shelf (light blue, 35°–55° S, 54°–68° W). Areas are illustrated with their corresponding colours in the map inset. **c**, Area-weighted ensemble-mean duration of low-aragonite and -calcite saturation state events in the Weddell Sea. **d**, Area-weighted ensemble-mean duration of aragonite undersaturation events ($D(\Omega_{(x,y,t)})$, orange) versus the area-weighted duration of aragonite undersaturation events due to the meridional trend ($D(\Omega^{prein}_{(x,y)} + {}_{zm}\delta\Omega_{(t,y)})$, blue, where zm represents zonal mean) across the Southern Ocean. Duration of aragonite undersaturation events due to spatial inhomogeneity ($D^{in} = D(\Omega_{(x,y,t)}) - D(\Omega^{prein}_{(x,y)} + {}_{zm}\delta\Omega_{(t,y)})$) around 2055 and 2095 are shown in the left and right inset, respectively.

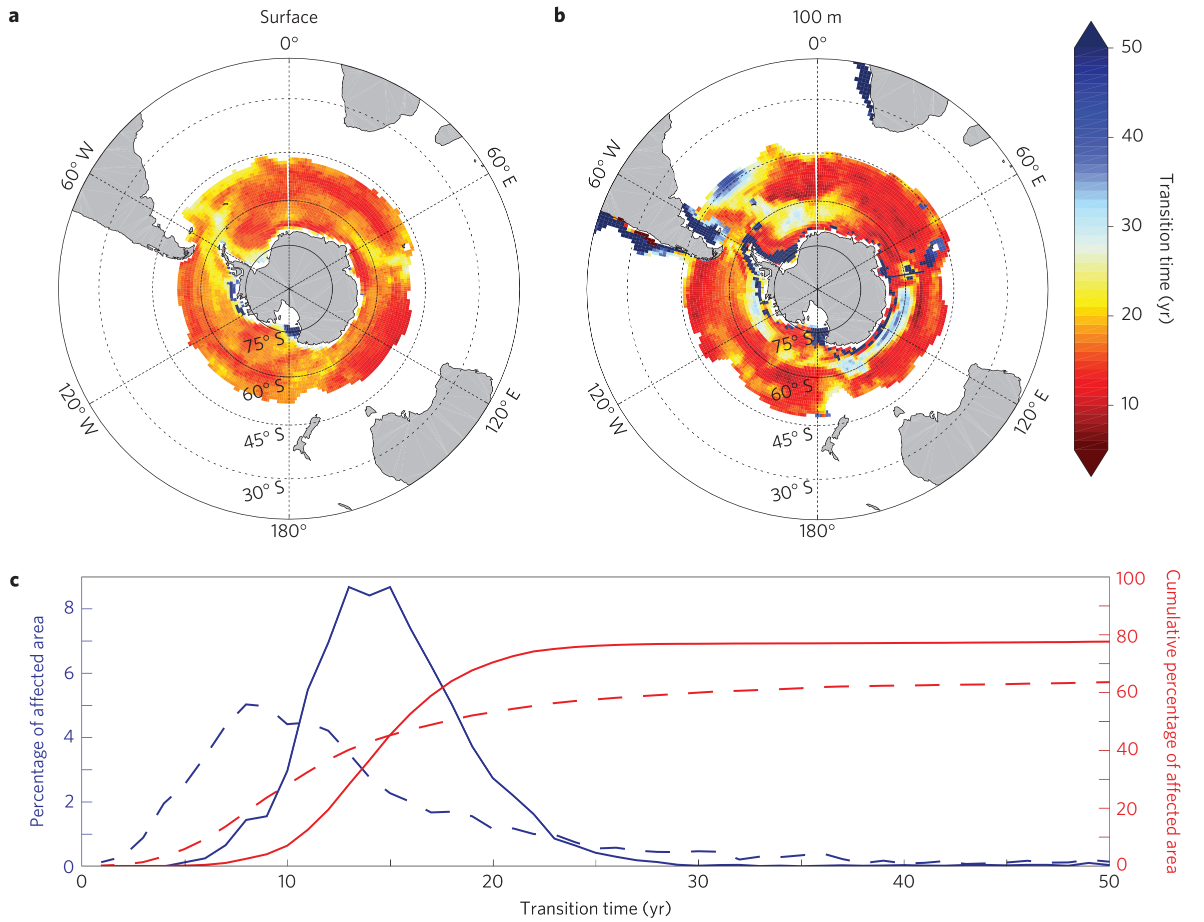


**Figure 3 | Transition time from short (1 month per year) to long (6 months per year) aragonite undersaturation events. a,b**, Ensemble mean of transition time at the surface (**a**) and at 100 m depth (**b**). **c**, Relative (blue) and cumulative (red) percentage of area in the Southern Ocean (south of 40° S) affected by end-of-century aragonite undersaturation at the surface (bold line) and at 100 m depth (dashed line) as a function of the transition time. Transition time is defined as how many years will pass from the first occurrence of an aragonite undersaturation event until it lasts 6 months per year, and is based on a 10-year running mean of the ensemble mean.

until events are 6 months or longer) is less than 20 years (Fig. 3). This short transition time and abrupt change in exposure duration to unfavourable conditions may be too fast for sensitive organisms such as pteropods to adapt, as these changes will occur within just a few generations. Furthermore, the transition time is shorter for organisms with a lower biological threshold than the transition time for organisms with a higher biological threshold, suggesting that calcifiers with a lower biological threshold may have less time to adapt to long aragonite undersaturation events (duration ≥6 months) once $\Omega$ drops below their biological threshold (Fig. 2c and Supplementary Fig. 4).

The spatial characteristics of projected twenty-first century surface aragonite undersaturation event dynamics in the Southern Ocean are largely determined by the present-day mean meridional gradient of $CO_3^{2-}$ concentration (Supplementary Fig. 5) and the superimposed relatively homogeneous anthropogenic $CO_2$ trend (Fig. 2d, see Methods). The largest zonal inhomogeneity, driven possibly by climate-induced changes to biology and ocean circulation, shortens and lengthens the duration of surface aragonite undersaturation events by only one month south of 65° S at around 2055, and in regions further north and close to Antarctica by the end of the century (Fig. 2d).

Whereas the simulated large-scale meridional pattern of $\Omega_{arag}$ is very similar to observational estimates (Supplementary Fig. 5), CMIP5 models may not adequately represent small-scale features, especially close to Antarctica. For example, the winter mixed-layer depth is generally too shallow in CMIP5 simulations[28], which may prevent deep $CO_2$-rich water from reaching the surface, leading potentially to an overestimation of $\Omega_{arag}$ values. Regional processes such as glacial meltwater input from Antarctica are not represented adequately in the Earth system models used here, which would directly decrease $\Omega_{arag}$ in coastal areas. Furthermore, potential negative or positive physiological responses from organisms to ocean acidification, such as altered calcification rates, $N_2$ fixation and net primary production, are not parameterized in the Earth system models used in CMIP5, but could cause significant changes to the carbonate chemistry and biogeochemical cycles[29]. Nevertheless, the ensemble mean represents the observed spatial pattern and temporal trend very well (see Methods and Supplementary Figs 5–7).

Our results suggest that the projected characteristics of Southern Ocean surface aragonite undersaturation events are almost entirely determined by anthropogenic $CO_2$ emissions and in this way conform to results of a recent study[30]. By comparing the different $CO_2$ emission scenarios (Fig. 2a, inset), it becomes apparent that only an immediate and substantial decrease in greenhouse gas emissions (RCP2.6) would keep surface aragonite undersaturation events sporadic, short (<1 month) and localized (Fig. 2, $CO_2$ axis). In contrast, atmospheric $CO_2$ concentrations of more than 500 ppm, as reached in the RCP4.5 scenario at the end of the century, are sufficient to cause surface aragonite undersaturation in ~20% of the Southern Ocean (Fig. 2a), with events lasting up to 5 months at 100 m depth (Fig. 2b). The rapid progression of ocean acidification is accompanied by additional climate-induced physical and chemical changes in the ocean[9,31]. The interaction of ocean acidification with these other changing environmental variables may also impact other key organisms, such as diatoms, that are otherwise not vulnerable to ocean acidification alone[32]. Our results show that the threat of ocean acidification in the sensitive Southern Ocean ecosystem may be more imminent than previously thought, and may spread quickly to the southern tips of New Zealand, South America and South Africa, with potentially far-reaching consequences to fisheries, local economies and livelihoods.

## Methods

Methods and any associated references are available in the online version of the paper.

Received 23 April 2015; accepted 15 September 2015; published online 2 November 2015

## References

1. McNeil, B. I. & Matear, R. J. Southern Ocean acidification: A tipping point at 450-ppm atmospheric $CO_2$. *Proc. Natl Acad. Sci. USA* **105**, 18860–18864 (2008).
2. Bednaršek, N. *et al*. Extensive dissolution of live pteropods in the Southern Ocean. *Nature Geosci.* **5**, 881–885 (2012).
3. Hunt, B. P. V. *et al*. Pteropods in Southern Ocean ecosystems. *Prog. Oceanogr.* **78**, 193–221 (2008).
4. Sunday, J. M. *et al*. Evolution in an acidifying ocean. *Trends Ecol. Evol.* **29**, 117–125 (2014).
5. Sarmiento, J. L., Gruber, N., Brzezinski, M. A. & Dunne, J. P. High-latitude controls of thermocline nutrients and low latitude biological productivity. *Nature* **427**, 56–60 (2004).
6. Gruber, N. *et al*. Oceanic sources, sinks, and transport of atmospheric $CO_2$. *Glob. Biogeochem. Cycles* **23**, 1–21 (2009).
7. Brooks, C. M. Competing values on the Antarctic high seas: CCAMLR and the challenge of marine-protected areas. *Polar J.* **3**, 277–300 (2013).
8. Schofield, O. *et al*. How do polar marine ecosystems respond to rapid climate change? *Science* **328**, 1520–1523 (2010).
9. Bromwich, D. H. *et al*. Central West Antarctica among the most rapidly warming regions on Earth. *Nature Geosci.* **6**, 139–145 (2012).
10. Kawaguchi, S. *et al*. Risk maps for Antarctic krill under projected Southern Ocean acidification. *Nature Clim. Change* **3**, 843–847 (2013).
11. Orr, J. C. *et al*. Anthropogenic ocean acidification over the twenty-first century and its impact on calcifying organisms. *Nature* **437**, 681–686 (2005).
12. Ries, J. B., Cohen, A. L. & McCorkle, D. C. Marine calcifiers exhibit mixed responses to $CO_2$-induced ocean acidification. *Geology* **37**, 1131–1134 (2009).
13. Tebaldi, C. & Knutti, R. The use of the multi-model ensemble in probabilistic climate projections. *Phil. Trans. R. Soc. A* **365**, 2053–2075 (2007).
14. Friedrich, T. *et al*. Detecting regional anthropogenic trends in ocean acidification against natural variability. *Nature Clim. Change* **2**, 167–171 (2012).
15. Hauri, C., Gruber, N., McDonnell, A. M. P. & Vogt, M. The intensity, duration, and severity of low aragonite saturation state events on the California continental shelf. *Geophys. Res. Lett.* **40**, 1–5 (2013).
16. Orr, J. C. in *Ocean Acidification* Vol. 2 (eds Gattuso, J.-P. & Hansson, L.) 41–66 (Oxford Univ. Press, 2011).
17. Wood, H. L., Spicer, J. I. & Widdicombe, S. Ocean acidification may increase calcification rates, but at a cost. *Proc. Biol. Sci.* **275**, 1767–1773 (2008).
18. Seibel, B. A., Maas, A. E. & Dierssen, H. M. Energetic plasticity underlies a variable response to ocean acidification in the pteropod, *Limacina helicina antarctica*. *PLoS ONE* **7**, e30464 (2012).
19. IPCC *Climate Change 2013: The Physical Science Basis* (eds Stocker, T. F. *et al*.) (Cambridge Univ. Press, 2013).
20. Hauri, C. *et al*. Inorganic carbon dynamics along the Western Antarctic Peninsula from 1998 until 2013. *Biogeosci. Discuss.* **12**, 6929–6969 (2015).
21. McNeil, B. I., Sweeney, C. & Gibson, J. A. E. Short note: Natural seasonal variability of aragonite saturation state within two Antarctic coastal ocean sites. *Antarct. Sci.* **23**, 411–412 (2011).
22. Lewis, C. N., Brown, K. A., Edwards, L. A., Cooper, G. & Findlay, H. S. Sensitivity to ocean acidification parallels natural $pCO_2$ gradients experienced by Arctic copepods under winter sea ice. *Proc. Natl Acad. Sci. USA* **110**, E4960–E4967 (2013).
23. Espinoza, P. & Bertrand, A. Revisiting Peruvian anchovy (*Engraulis ringens*) trophodynamics provides a new vision of the Humboldt Current system. *Prog. Oceanogr.* **79**, 215–227 (2008).
24. Montecino, V. & Lange, C. B. The Humboldt current system: Ecosystem components and processes, fisheries, and sediment studies. *Prog. Oceanogr.* **83**, 65–79 (2009).
25. Bisbal, G. A. The Southeast South American shelf large marine ecosystem. *Mar. Policy* **19**, 21–38 (1995).
26. Carrillo, C. J., Smith, R. C. & Karl, D. M. Processes regulating oxygen and carbon dioxide in surface waters west of the Antarctic Peninsula. *Mar. Chem.* **84**, 161–179 (2004).
27. Bednaršek, N., Tarling, G. A., Fielding, S. & Bakker, D. C. E. Population dynamics and biogeochemical significance of *Limacina helicina antarctica* in the Scotia Sea (Southern Ocean). *Deep-Sea Res. II* **59–60**, 105–116 (2012).
28. Meijers, A. J. S. The Southern Ocean in the coupled model intercomparison project phase 5. *Phil. Trans. R. Soc. A* **372**, 20130296 (2014).
29. Doney, S. C., Fabry, V. J., Feely, R. A. & Kleypas, J. A. Ocean acidification: The other $CO_2$ problem. *Ann. Rev. Mar. Sci.* **1**, 169–192 (2009).
30. Hauck, J. *et al*. On the Southern Ocean $CO_2$ uptake and the role of the biological carbon pump in the 21st century. *Glob. Biogeochem. Cycles* http://dx.doi.org/10.1002/2015GB005140 (2015).
31. Montes-Hugo, M. *et al*. Recent changes in phytoplankton communities associated with rapid regional climate change along the western Antarctic Peninsula. *Science* **323**, 1470–1473 (2009).
32. Hoppe, C. J. M., Holtz, L.-M., Trimborn, S. & Rost, B. Ocean acidification decreases the light-use efficiency in an Antarctic diatom under dynamic but not constant light. *New Phytol.* http://dx.doi.org/10.1111/nph.13334 (2015).

## Acknowledgements

We acknowledge support from the National Science Foundation Ocean Acidification Program (OCE-1314209). This is IPRC publication no. 1152 and SOEST publication no. 9508.

## Author contributions

T.F. prepared and analysed the model output. C.H. analysed the model output and wrote the paper. All authors were involved in the study design, discussed the results and commented on the manuscript.

## Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to C.H.

## Competing financial interests

The authors declare no competing financial interests.

## Methods

**CMIP5 models and simulations.** Our analysis was based on the output of historical and Representative Concentration Pathway 8.5 (RCP8.5; ref. 19) simulations, conducted with ten Earth system models from the Coupled Model Intercomparison Project, Phase 5 (CMIP5), including CESM1-BGC, CanESM2, GFDL-ESM2G, GFDL-ESM2M, HadGEM2-ES, HadGEM2-CC, IPSL-CM5A-LR, IPSL-CM5A-MR, IPSL-CM5B-LR and MPI-ESM-LR (see Supplementary Table 1 for model details and references). All models are fully coupled climate–biogeochemical models that simulate interannual variability and future climate change. All models include an annual cycle and were forced with atmospheric $CO_2$ concentrations following RCP8.5 (ref. 19). Monthly outputs for the surface aragonite saturation state ($\Omega_{arag}$) from 1860 to 2100 for each model were downloaded from the official CMIP5 data portal (https://pcmdi9.llnl.gov/search/cmip5). Individual CMIP5 model output was regridded to a $1° \times 1°$ horizontal resolution and subsequently averaged to derive an ensemble mean.

**Model evaluation.** Comparison of the model ensemble mean of $\Omega_{arag}$ for the period 1991–2000 with $\Omega_{arag}$ from the Global Ocean Data Analysis Project shows that the observed Southern Hemisphere large-scale pattern of meridionally decreasing $\Omega_{arag}$ is well represented by the ensemble mean (Supplementary Fig. 5 and references in figure caption). The spatial correlation coefficients between observed and simulated Southern Ocean $\Omega_{arag}$ (south of 40° S) range between 0.81 and 0.91 for the different models (Supplementary Fig. 6). All models simulate the amplitude of variations well, shown by the normalized standard deviation between 0.78 and 0.9 in the Taylor diagram. Cruise data from the Pacific sector of the Southern Ocean[33] suggest that the model mean underestimates $\Omega_{arag}$ slightly ($\sim$0.1 units) in this region (Supplementary Fig. 7a). The modelled trend of $\Omega_{arag}$ was also compared with available time series from other regions, including the Bermuda Atlantic Timeseries Station (BATS); the European Station for Timeseries in the Ocean, close to the Canary Islands (ESTOC); and the Hawaii Ocean Timeseries (HOT), which indicates that the ensemble mean follows the observed decreasing trend of $\Omega_{arag}$ closely (Supplementary Fig. 7 and references therein).

**Duration of aragonite undersaturation events.** The duration of aragonite undersaturation events is defined as a consecutive sequence of months with surface waters undersaturated with respect to $\Omega_{arag}$ ($\Omega_{arag} < 1$). Similarly, the duration of aragonite undersaturation events at 100 m depth is defined as a consecutive sequence of months with depth of the aragonite saturation horizon shallower than 100 m. A ten-year running mean duration of surface aragonite undersaturation events per year and model was calculated to avoid the influence of internal model variability. The duration of aragonite undersaturation events was then averaged across each ensemble member and rounded to the closest integer. The duration for other $\Omega_{arag}$ thresholds and for calcite was obtained accordingly.

**Model agreement on first occurrence of aragonite undersaturation events.** A ten-year running mean of the multi-model ensemble mean suggests that surface aragonite undersaturation events first occurred in the Bellingshausen and Ross seas around 1965 (Supplementary Fig. 1a). Even though there are no observations from the 1960s to evaluate these results, recent data show $\Omega_{arag}$ around 1 in these regions[20,21]. However, IPSL-CM5A-MR and IPSL-CM5A-LR already simulate sporadic aragonite undersaturation events in coastal areas off Antarctica and CanESM2 off the coast of central Chile at the beginning of the twentieth century (Supplementary Fig. 1b), suggesting a large uncertainty in the timing of the first anthropogenic aragonite undersaturation events within the multi-model ensemble. All models agree that coastal waters around Antarctica will experience surface undersaturation before 2060 (Supplementary Fig. 1c).

**Spatial inhomogeneity.** To elucidate whether the temporal progression of the duration and spatial spread of aragonite undersaturation events is driven mainly by the natural latitudinal $[CO_3{}^{2-}]$ distribution and superimposed anthropogenic $CO_2$ trend, or whether small-scale regional ocean circulation features associated with zonal inhomogeneities also have an influence, a fictitious data set of $\Omega_{arag}$ ($\Omega^{fic}$) for each model using the following equation: $\Omega^{fic}_{(x,y,t)} = \Omega^{prein}_{(x,y)} + {}_{zm}\delta\Omega_{(t,y)}$ was generated. $\Omega^{prein}$ represents a ten-year climatological mean of pre-industrial $\Omega_{arag}$ conditions (1861–1870) and ${}_{zm}\delta\Omega$ is the zonal mean temporal trend of $\Omega_{arag}$ for the period from 1900 to 2099 (Fig. 2d). The duration of aragonite undersaturation events per model and as an ensemble mean were then calculated on the basis of $\Omega^{fic}$. Finally, the duration of aragonite undersaturation events due to spatial inhomogeniety was defined as $D^{in} = D(\Omega_{(x,y,z)}) - D(\Omega^{prein}_{(x,y)} + {}_{zm}\delta\Omega_{(t,y)})$.

## References

33. Midorikawa, T. *et al*. Decreasing pH trend estimated from 35-year time series of carbonate parameters in the Pacific sector of the Southern Ocean in summer. *Deep-Sea Res. I* **61**, 131–139 (2012).