

# Stratospheric ozone depletion and tropospheric ozone increases drive Southern Ocean interior warming

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23 **Atmospheric ozone has undergone distinct changes in the stratosphere and troposphere**  
24 **during the second half of the twentieth century, with depletion in the stratosphere and an**  
25 **increase in the troposphere. Until now, the effect of these changes on ocean heat uptake has**  
26 **been unclear. Here we show that both stratospheric and tropospheric ozone changes have**  
27 **contributed to Southern Ocean interior warming, with the latter being more important.**  
28 **The ozone changes between 1955 and 2000 induced about 30% of the net simulated ocean**  
29 **heat content increase in the upper 2000 m of the Southern Ocean, with around 60%**  
30 **attributed to tropospheric increases and 40% to stratospheric depletion. Moreover, these**  
31 **two warming contributions show distinct physical mechanisms: Tropospheric ozone**  
32 **increases cause a subsurface warming in the Southern Ocean primarily via the deepening**  
33 **of isopycnals, while stratospheric ozone depletion via spiciness changes along isopycnals.**  
34 **Our results highlight that tropospheric ozone is more than an air pollutant and, as a**  
35 **greenhouse gas, has been pivotal to the Southern Ocean warming.**

36

37 Atmospheric ozone has experienced distinct changes in the stratosphere and troposphere during  
38 the second half of the twentieth century. Notable ozone depletion has occurred in the  
39 stratosphere, most strikingly as the ozone hole over Antarctica, which has been attributed  
40 primarily to anthropogenic emissions of ozone-depleting substances<sup>1,2,3,4</sup>. In contrast, ozone  
41 increases in the troposphere have been observed (Extended Data Fig. 1) as a result of  
42 anthropogenic emissions of ozone precursors such as methane, non-methane volatile organic  
43 compounds, carbon monoxide and nitrogen oxides<sup>5,6,7,8</sup>. These atmospheric ozone changes have  
44 profound impacts on Earth's climate system. For example, stratospheric ozone depletion has  
45 significantly altered the tropospheric circulation by displacing the Southern Hemisphere westerly

46 winds poleward during austral summer<sup>9,10,11,12</sup>, though these Southern Hemisphere circulation  
47 trends paused around 2000 and are expected to reverse the sign owing to reduced emissions of  
48 ozone depleting substances following the signing of the Montreal Protocol and its  
49 Amendments<sup>13,14,15</sup>. By contrast, ozone impacts on oceans, especially those due to tropospheric  
50 ozone changes, are relatively less well explored.

51

52 The Fifth Assessment Report of the United Nations Intergovernmental Panel on Climate Change  
53 indicates that ozone constitutes the third-most important contribution to greenhouse gas forcing  
54 since pre-industrial times after carbon dioxide and methane<sup>16</sup>. Stratospheric and tropospheric  
55 ozone changes substantially modulate Earth's radiation balance<sup>17</sup>, and thus could also affect  
56 global ocean heat uptake. The role of the Southern Ocean is critical in the context of climate  
57 change as it is one of the most important regions for taking up excess heat in a warming  
58 climate<sup>18,19</sup>, and is markedly affected by Southern Hemisphere westerly winds<sup>20,21,22,23</sup>. During  
59 the past several decades, the Southern Ocean has shown a rapid subsurface warming<sup>24,25</sup>, only a  
60 small part of which, however, has been attributed to stratospheric ozone depletion<sup>26,27,28</sup>. Given  
61 the concurrent (but opposite) ozone changes in both the stratosphere and troposphere, one gap  
62 remains in our current knowledge of ozone-driven Southern Ocean warming: The impact of the  
63 increase in tropospheric ozone. Here, we employ historical simulations and accompanying ozone  
64 single-forcing experiments with a broad set of climate models from the Coupled Model  
65 Intercomparison Projects Phase 5/6 (CMIP5/6) to probe the mechanisms and impacts of  
66 stratospheric and tropospheric ozone changes on Southern Ocean interior warming during the  
67 second half of the twentieth century.

68

69 **Results**

70 We first examine the ozone single-forcing experiments from the CMIP5 models in which the  
71 models were only forced with historical integrations of atmospheric ozone concentrations instead  
72 of all historical forcings (see Methods). These ozone single-forcing experiments demonstrate the  
73 effects of both stratospheric and tropospheric ozone changes together. Between 1955 and 2000,  
74 ozone depletion generates a strong stratospheric cooling trend in the Southern Hemisphere high  
75 latitudes (Extended Data Fig. 2), which leads to a poleward intensification of Southern  
76 Hemisphere westerly winds in the troposphere reminiscent of “annular mode–like” responses<sup>29</sup>  
77 (Fig. 1a). Along with the response in the atmosphere, ozone changes also produce a pronounced  
78 subsurface warming in the Southern Ocean. Within 40–50°S, the warming rate is larger than 0.01  
79 K/decade in the upper 1000 m (Fig. 1c). When we integrate ocean heat content (OHC) over the  
80 upper 2000 m between 30°S and 60°S where ocean warming mainly occurs, we find a significant  
81 increase of OHC, with a trend of  $5.63 \pm 2.36$  ZJ/decade (1 ZJ =  $10^{21}$  joule; multi-model mean  $\pm$  1  
82 standard deviation among models, see Methods) between 1955 and 2000 (Fig. 2a). Our results  
83 from these CMIP5 model simulations thus suggest a substantial Southern Ocean subsurface  
84 warming in response to stratospheric and tropospheric ozone changes.

85

86 We have also examined the recent ozone single-forcing experiments with the new generation of  
87 CMIP6 models. Unlike those with CMIP5 models, in these experiments CMIP6 models are  
88 forced with historical changes solely in stratospheric ozone concentration (see Methods). Hence  
89 the CMIP6 ozone experiments show solely the effect of stratospheric ozone change. Compared  
90 with the results from the CMIP5 experiments, the CMIP6 ozone experiments show similar  
91 stratospheric cooling in southern high latitudes and poleward intensified Southern Hemisphere

92 westerly winds, which indicates a major role of stratospheric ozone depletion in the atmospheric  
93 response during 1955-2000 (Fig. 1b). However, in the Southern Ocean, we find a much weaker  
94 subsurface warming in the CMIP6 stratospheric ozone only experiments with a pattern consistent  
95 with previous studies<sup>27,30</sup>. Between 40°S and 50°S, the warming rate is much smaller than 0.01  
96 K/decade in the upper 2000 m (Fig. 1d). The upper 2000-m OHC between 30°S and 60°S  
97 exhibits a marginal increase between 1955 and 2000, with a trend of  $0.45 \pm 1.22$  ZJ/decade  
98 (multi-model mean  $\pm$  1 standard deviation among models; Fig. 2a). We further find no  
99 statistically significant difference in transient climate sensitivity between the CMIP5 and CMIP6  
100 models (see Methods) but the Southern Ocean OHC trend in the CMIP5 simulations is one order  
101 of magnitude larger than that in the CMIP6 simulations, indicating that the difference in model  
102 climate sensitivity cannot serve as the major cause of such distinct warming trends in the  
103 Southern Ocean. On the other hand, the comparison between CMIP5 and CMIP6 model  
104 simulations implies that the tropospheric ozone increase is a key driver of Southern Ocean  
105 interior warming. Nevertheless, it is worth noting that this comparison cannot allow for a  
106 conclusive quantification of the impact nor shed light on the mechanism of tropospheric ozone  
107 increases on Southern Ocean warming, since the differences in prescribed historical ozone  
108 datasets between CMIP5 and CMIP6 models (Extended Data Fig. 1) and model responses to  
109 ozone forcing would need to be considered.

110

### 111 **Quantifying the ozone impacts on Southern Ocean warming**

112 To quantify the impact of tropospheric ozone change on Southern Ocean interior warming and  
113 investigate the mechanism, we employ two ensembles of ozone single-forcing simulations  
114 performed with the same climate model, CanESM5. This model simulates a Southern Ocean

115 warming generally in alignment with the ensemble mean result of CMIP6 stratospheric ozone  
116 only experiments (Fig. 2a). The first CanESM5 ensemble is forced with historical changes in  
117 both stratospheric and tropospheric ozone, equivalent to the CMIP5 simulations described above  
118 but adopting the CMIP6 simulation protocol<sup>31,32</sup>. The second ensemble simulation is equivalent  
119 to the CMIP6 ozone experiments described above in which the model is forced with historical  
120 integrations of solely stratospheric ozone changes (see Methods). The difference between the  
121 two ensemble simulations therefore isolates the effect of tropospheric ozone change. Relative to  
122 preindustrial times, we find that both ensemble experiments from CanESM5 simulate a  
123 stratospheric cooling in the southern high latitudes and a significant poleward intensification of  
124 Southern Hemisphere westerlies in the troposphere between 1955 and 2000 (Fig. 3a,b). On the  
125 other hand, tropospheric ozone increases lead to a warming in the troposphere and a cooling in  
126 the stratosphere<sup>33</sup> (Extended Data Fig. 3), together with a significant upward intensification of  
127 Southern Hemisphere westerly winds in the upper levels and a poleward intensification of  
128 westerly winds towards surface (Fig. 3c). These tropospheric-ozone-produced atmosphere  
129 temperature and circulation changes are comparable to those induced by stratospheric ozone  
130 depletion towards the surface layers, suggesting that tropospheric ozone changes could  
131 potentially have considerable impacts on the oceans underneath.

132

133 We further probe the temperature response in the Southern Ocean in the two CanESM5 ensemble  
134 simulations. We find a region of pronounced warming extending downward and equatorward to  
135 the north of 60°S as a response to the combined stratospheric and tropospheric ozone changes.  
136 Between 40°S and 50°S, this tongue of warming waters reaches 1200 m with a warming rate  
137 exceeding 0.01 K/decade (Fig. 3d). Part of this subsurface warming is induced by stratospheric

138 ozone depletion, which is, however, mostly limited to the upper 600 m (Fig. 3e). On the other  
139 hand, the vertical extension of the tongue of warming waters depends essentially on tropospheric  
140 ozone forcing. The increase of tropospheric ozone creates such a deep warming in the Southern  
141 Ocean that warming larger than 0.01 K/decade is found to penetrate as deep as 1000 m within  
142 40-50°S (Fig. 3f). To the north of the tongue of warming waters, there is a tongue of cooling  
143 waters in the upper levels of the Southern Ocean, which results principally from stratospheric  
144 ozone depletion and secondarily from tropospheric ozone increases (Fig. 3e,f). A similar cooling  
145 feature is found at high latitudes south of 55°S (Fig. 3d,f). It is worth noting that the warming  
146 pattern due to tropospheric ozone increases is different from that due to the rising well-mixed  
147 greenhouse gases such as carbon dioxide. The rise of well-mixed greenhouse gases induces  
148 ubiquitous while vertically decaying warming in the upper 2000-m ocean in the Southern  
149 Ocean<sup>20,21,27,34</sup>.

150

151 Here we estimate the OHC variations in the upper 2000 m between 30°S and 60°S in the two sets  
152 of CanESM5 simulations. We find that atmospheric ozone changes induce a robust upward OHC  
153 trend of  $4.58 \pm 3.35$  ZJ/decade (ensemble mean  $\pm$  1 standard deviation among ensembles) between  
154 1955 and 2000, of which about two-fifths ( $1.84 \pm 2.50$  ZJ/decade, ensemble mean  $\pm$  1 standard  
155 deviation among ensembles) can be attributed to stratospheric ozone depletion while the other  
156 three-fifths (2.74 ZJ/decade, the difference of the ensemble means between the two suites of  
157 ozone simulations) is driven by tropospheric ozone increases. Our results confirm the importance  
158 of tropospheric ozone to Southern Ocean heat uptake and storage. Importantly, the increases in  
159 tropospheric ozone have been more effective in driving the interior warming over the Southern  
160 Ocean during the second half of the twentieth century compared to stratospheric ozone depletion.

161  
162 Moreover, to set in context the effect of ozone forcing on the historical OHC increase in the  
163 Southern Ocean over the 1955-2000 period, we compare OHC changes in the upper 2000 m  
164 within 30-60°S between the CanESM5 historical simulations that also include the other  
165 greenhouse gas forcings from carbon dioxide, methane and nitrous oxide and the combined  
166 stratosphere-troposphere ozone single-forcing experiment (Fig. 2b). We find that CanESM5  
167 simulates a general long-term increase of OHC in the Southern Ocean, at a rate of  $13.77 \pm 4.29$   
168 ZJ/decade (ensemble mean  $\pm 1$  standard deviation among ensembles) between 1955 and 2000,  
169 which is consistent with the OHC trends inferred from observations (16.25 ZJ/decade) and  
170 CMIP5 models ( $14.60 \pm 5.27$  ZJ/decade, multi-model mean  $\pm 1$  standard deviation among models)  
171 (Fig. 2b). Using the CanESM5 ozone experiment, we further find that about 33.2% of the net  
172 historical OHC increase between 1955 and 2000 is caused by atmospheric (both stratospheric  
173 and tropospheric) ozone changes. This ratio is in line with that suggested by the historical  
174 CMIP5 model ozone experiment ( $38.4 \pm 10.4\%$ , multi-model mean  $\pm 1$  standard deviation among  
175 models).

176  
177 **Physical mechanisms of ozone-driven Southern Ocean warming**  
178 To further understand the mechanisms by which stratospheric and tropospheric ozone changes  
179 drive Southern Ocean interior warming, we decompose the temperature and salinity changes  
180 between 1955 and 2000 at depth levels into the spiciness changes along isopycnals and  
181 heave-related changes owing to the vertical heave of isopycnals<sup>35</sup> (see Methods). The spiciness  
182 reveals alterations in water mass properties as a result of the subduction of surface temperature  
183 and salinity anomalies and changes by interior mixing processes. The heave of isopycnals could

184 be linked to changes in wind-driven ocean circulation and the redistribution of heat and salt in  
185 the interior ocean<sup>36</sup>.

186

187 We first depict the temperature and salinity responses to total atmospheric ozone variations.  
188 During 1955-2000, the zonally averaged spiciness changes on density surfaces exhibit strong  
189 warming and salification trends in the upper ocean toward isopycnal outcrops, especially in the  
190 latitudes between 40°S and 60°S (Fig. 4a,b). These warming and salification trends primarily  
191 result from stratospheric ozone depletion and not from tropospheric ozone increases (Fig. 4c,d).  
192 Between 40°S and 60°S, the Southern Ocean takes heat from the atmosphere but loses freshwater  
193 in response to stratospheric ozone depletion (Fig. 5a,b), both contributing to warming and  
194 salification spiciness trends<sup>36</sup>. The peak of Southern Ocean surface heat uptake is around 55°S  
195 (Extended Data Fig. 4), essentially due to the increase of downward turbulent latent heat flux<sup>37</sup>  
196 over the Indian Ocean sector (Extended Data Fig. 5). Increases in surface shortwave radiation  
197 fluxes also contribute to Southern Ocean heat uptake in these latitudes (Extended Data Fig. 3).  
198 On the other hand, the reduction in surface freshwater flux can be mostly attributed to changes in  
199 precipitation minus evaporation (P-E) to the north of 54°S but is likely related to sea-ice  
200 variations to the south (Extended Data Fig. 6). Between 40°S and 54°S, the P-E reduction results  
201 from both precipitation decreases and evaporation increases and is especially robust over the  
202 Pacific sector (Extended Data Fig. 7).

203

204 Besides the warming and salification trends in the isopycnal outcropping region between 40°S  
205 and 60°S, we also find cooling and freshening spiciness changes to the north of 40°S on density  
206 surfaces between 26.3 and 27.0 kg/m<sup>3</sup> (Fig. 4a,b), within the density ranges of the Subantarctic

207 Mode Water (SAMW) and Antarctic Intermediate Water (AAIW) simulated by climate models<sup>38</sup>.  
208 Particularly, the spiciness changes in the AAIW density range (26.7-27.0 kg/m<sup>3</sup>) can be  
209 attributed mostly to stratospheric ozone depletion (Fig. 4c,d) while those in the SAMW density  
210 range (26.3-26.6 kg/m<sup>3</sup>) mainly to tropospheric ozone increases (Fig. 4e,f). These ozone-induced  
211 cooling signals contribute to the cooling trend found at corresponding locations from  
212 observations and historical simulations during 1955-2000 (Extended Data Fig. 8).

213  
214 After remapping the spiciness changes onto depth levels using the mean depth of each density  
215 surface, we find the major spiciness warming (>0.01 K/decade) trends in response to total  
216 atmospheric ozone variations extending equatorward and downward from the surface layer at  
217 60°S to about 600 m at 40°S (Fig. 6a). The stratospheric ozone depletion is responsible for most  
218 of the ozone-induced warming trends in the upper 500 m (Fig. 6c) while tropospheric ozone  
219 increases primarily account for the spiciness warming below (Fig. 6e).

220  
221 We further analyze the heave component of Southern Ocean temperature change. We find a  
222 subsurface warming region (>0.01 K/decade) extending equatorward between 36°S and 51°S and  
223 downward in 300-1100 m (Fig. 6b) in response to atmospheric ozone changes, accompanied by a  
224 cooling tongue to the north and in upper levels. This pair of warming and cooling anomalies has  
225 been linked to poleward intensified surface westerly winds and indicates heat redistribution  
226 within the Southern Ocean<sup>21,39</sup>. Specifically, stratospheric ozone depletion drives an  
227 intensification of surface westerly winds at and to the south of the Antarctic Circumpolar Current  
228 but a relaxation to the north (Fig. 5c, Extended Data Fig. 9), in a pattern consistent with other  
229 CMIP6 models (Extended Data Fig. 10). The zonally averaged zonal wind change exhibits a

230 dipole-like pattern, with positive and negative anomalies to the south and north of around 50°S.  
231 The resultant anomalous Ekman transport convergence and wind-driven downwelling produces a  
232 deepening of isopycnals in the latitudes around 50°S and hence heave-induced changes of  
233 warming. While to the north of about 43°S, the weakening of surface westerlies progressively  
234 decays, which prompts an anomalous Ekman transport divergence and wind-driven upwelling  
235 (Extended Data Fig. 9d) and thus leads to shallower isopycnals and cooling heave changes in  
236 these latitudes (Fig. 6d).

237

238 Tropospheric ozone increases, on the other hand, engender different changes in surface winds  
239 from those due to stratospheric ozone depletion (Fig. 5c, Extended Data Fig. 9). The zonally  
240 averaged surface zonal wind change also reflects a dipole-like pattern but located more  
241 northward, with positive and negative anomalies occurring to the south and north of around 42°S.  
242 This pattern indicates less poleward displaced surface westerlies than their counterparts driven  
243 by stratospheric ozone depletion. Tropospheric ozone increases also drive poleward-intensified  
244 Southern Hemisphere precipitation and significantly increase evaporation at lower latitudes  
245 where the tropospheric ozone increases are stronger (Extended Data Fig. 6f). In the ocean, the  
246 wind-driven Ekman pumping (Extended Data Fig. 9d) produces isopycnals deepening in much  
247 lower latitudes, around 42°S, and warming heave changes there (Fig. 6f). These heave-related  
248 warming changes are much stronger than those induced by stratospheric ozone depletion, which  
249 is likely due to the fact that the oceanic thermocline is more strongly stratified at lower  
250 latitudes<sup>36</sup>, allowing the wind-driven downwelling more effectively to create warming heave  
251 changes there.

252

253 **Discussion**

254 In summary, we have examined the climate impacts of atmospheric ozone changes during the  
255 second half of the twentieth century, with a focus on disentangling effects of stratospheric ozone  
256 depletion and tropospheric ozone increases. We show that while stratospheric ozone depletion  
257 plays a dominant role in atmospheric temperature and wind changes in southern high latitudes in  
258 the stratosphere and upper levels of the troposphere, tropospheric ozone increases have made a  
259 larger contribution to Southern Ocean interior warming. Between 1955 and 2000, about one-third  
260 of the historical OHC increase in the upper 2000 m of the Southern Ocean between 30°S and  
261 60°S was induced by atmospheric ozone changes, of which around three-fifths can be attributed  
262 to tropospheric ozone increases and the other two-fifths to stratospheric ozone depletion.  
263 Tropospheric ozone increases cause Southern Ocean subsurface warming primarily via the  
264 deepening of isopycnals. They give rise to an intensification of surface westerly winds over the  
265 Southern Ocean such that the wind-driven Ekman pumping brings about isopycnal deepening  
266 around 42°S and prompts heave-induced warming there. On the other hand, stratospheric ozone  
267 depletion promotes warming in the Southern Ocean mainly through spiciness changes along  
268 isopycnals in the upper 500 m. In response to stratospheric ozone depletion, the net surface  
269 downward heat flux increases but the freshwater flux decreases over the Southern Ocean  
270 between 40°S and 60°S, contributing to the warming and salification spiciness changes in the  
271 isopycnal outcropping regions of the Southern Ocean.

272

273 In our study, the finding that stratospheric and tropospheric ozone changes contributed to around  
274 one-third of the historical OHC increase during the second half of the twentieth century is  
275 consistent with the result from previous studies examining simulations with fixed ozone

276 depleting substances (ODSs)<sup>40</sup>. However, the response to ODSs, inferred by differencing  
277 historical simulations with all anthropogenic forcings and simulations with fixed ODSs, omits  
278 changes in tropospheric ozone induced by precursor omissions, but includes radiative effects of  
279 ODSs themselves, and hence these results are not directly comparable with our study of the  
280 direct effects of tropospheric and stratospheric ozone changes. Furthermore, our results suggest  
281 that, when the effect of tropospheric ozone increases is considered, the ozone impacts on  
282 Southern Ocean interior warming are much larger than previous estimates that only considered  
283 stratospheric ozone depletion<sup>27</sup>. Between 1955 and 2000, tropospheric ozone increases  
284 significantly affect the P-E over the Southern Ocean. As such, our results highlight that  
285 tropospheric ozone, besides being an air pollutant, is an important contributor to ocean heat  
286 uptake and hydrological cycle change in the Southern Hemisphere.

287

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411

412 **Acknowledgments**

413 W.L. is supported by the Alfred P. Sloan Foundation as a Research Fellow and by U.S. National  
414 Science Foundation (AGS-2053121, OCE 2123422). K.L. and X.Z. are funded by the Centre for  
415 Southern Hemisphere Oceans Research (CSHOR), jointly funded by the Qingdao National  
416 Laboratory for Marine Science and Technology (QNLM, China) and the Commonwealth  
417 Scientific and Industrial Research Organisation (CSIRO, Australia).

418

419 **Author Contributions Statement**

420 W.L. conceived the study, performed the analysis and wrote the original draft of the manuscript.  
421 S.L. contributed to the analysis. W.L., M.I.H., R.C.-G., S.L., N.P.G., K.L., X.Z. and N.C.S.  
422 contributed to interpreting the results and made substantial improvements to the manuscript.

423

424 **Competing Interests Statement**

425 The authors declare no competing interests.

426

427 **Figure legends**

428 **Figure 1. Changes in Southern Hemisphere westerlies and Southern Ocean temperature in**  
429 **response to ozone changes in CMIP5 and CMIP6 simulations.** (top row) Trends of annual  
430 and zonal mean zonal winds during 1955-2000 (shading in m/s/decade) of the multi-model  
431 means (MMMs) from (a) CMIP5 and (b) CMIP6 models ozone single-forcing experiments. The  
432 annual climatologies of zonal mean zonal winds (contour in m/s, with an interval of 5 m/s and  
433 the zero contours thickened) of the MMMs from CMIP5 and CMIP6 models preindustrial  
434 control runs are superimposed on both panels, respectively. (bottom row) Trends of annual and  
435 zonal ocean temperature during 1955-2000 (shading in K/decade) of the MMMs from (c) CMIP5  
436 and (d) CMIP6 models ozone single-forcing experiments. Stippling indicates that the change is  
437 statistically insignificant at the 95% confidence level of the Mann-Kendall trend significance test  
438 (see Methods).

439

440 **Figure 2. Observed and simulated Southern Ocean heat content.** (a) Ocean heat content  
441 (OHC) anomalies (relative to the value in 1955) integrated over the upper 2000 m between 30°S  
442 and 60°S from ozone single-forcing experiments with four CMIP5 models (MMM, medium blue;  
443 inter-model spread, light blue; see Methods) and four CMIP6 models (MMM, magenta;  
444 inter-model spread, light magenta), and the ensemble means from CanESM5 stratospheric and  
445 tropospheric ozone experiment (blue), stratospheric ozone only experiment (red) as well as the  
446 difference between the two (black) indicating the effect of tropospheric ozone change. The  
447 inter-model spread is calculated as one standard deviation of the ensemble means of individual  
448 models. (b) Same as panel (a) but for OHC anomalies from the IAP observation data (orange)  
449 and historical simulations with the four CMIP5 models (MMM, medium purple; inter-model  
450 spread, light purple) and CanESM5 (ensemble mean, purple). OHC anomalies from CMIP5

451 ozone single-forcing experiments are also included in the panel. Note that the OHC from the IAP  
452 observations is a single realization, which has larger interannual variations than the other OHCs  
453 from MMM.

454

455 **Figure 3. Changes in Southern Hemisphere westerlies and Southern Ocean temperature in**  
456 **response to ozone changes in CanESM5 simulations.** (top row) Changes in annual and zonal  
457 mean zonal winds (shading in m/s) during 1955-2000 (relative to preindustrial control run) for  
458 the ensemble means in CanESM5 (a) stratospheric and tropospheric ozone experiment and (b)  
459 stratospheric ozone only experiment as well as (c) the difference between the two indicating the  
460 effect of tropospheric ozone change. Stippling indicates that the change is statistically  
461 insignificant at the 95% confidence level of the Student's t-test (see Methods). (bottom row)  
462 Same as the top row but for trends of annual and zonal mean ocean temperature during  
463 1955-2000 (shading in K/decade). Stippling indicates that the change is statistically insignificant  
464 at the 95% confidence level of the Mann-Kendall trend significance test (see Methods).

465

466 **Figure 4. Temperature and salinity spiciness changes on density surfaces in CanESM5**  
467 **ozone experiments.** (left column) Spiciness changes in annual and zonal mean ocean  
468 temperature trends during 1955-2000 (shading in K/decade) on density surfaces for the ensemble  
469 means in CanESM5 (a) stratospheric and tropospheric ozone experiment and (c) stratospheric  
470 ozone only experiment as well as (e) the difference between the two indicating the effect of  
471 tropospheric ozone change. (right column) Same as the left column but for spiciness changes in  
472 annual and zonal mean ocean salinity trends (shading in  $10^{-2}$  psu/decade).

473

474 **Figure 5. Surface heat flux, freshwater flux and zonal winds changes in CanESM5 ozone**  
475 **experiments.** (a) Changes in annual and zonal mean net surface heat fluxes over the Southern  
476 Ocean during 1955-2000 (relative to preindustrial control run) for the ensemble means in  
477 CanESM5 stratospheric and tropospheric ozone experiment (light blue; significant, blue) and  
478 stratospheric ozone only experiment (orange; significant, red) as well as the difference between  
479 the two (gray; significant, black) indicating the effect of tropospheric ozone change. Panels (b)  
480 and (c) are the same as (a) but for changes in annual and zonal mean net surface freshwater  
481 fluxes over the ocean and surface zonal winds. The variable of surface zonal wind is obtained  
482 from atmosphere model outputs and land is then masked out for the variable so that winds are on  
483 the liquid ocean water surface in most parts of the Southern Ocean but on sea ice surface around  
484 or south of 60°S where sea ice exists. Heat and freshwater fluxes are positive downward. In all  
485 the panels, changes are tested based on the Student's t-test and denoted statistically significant  
486 when exceeding the 95% confidence level (see Methods).

487

488 **Figure 6. Spiciness and heave changes of ocean temperature in CanESM5 ozone**  
489 **experiments.** (left column) Spiciness changes in annual and zonal mean ocean temperature  
490 trends during 1955-2000 (shading in K/decade) above 2000 m but below the mixed layer (~150  
491 m) in the Southern Ocean for the ensemble means in CanESM5 (a) stratospheric and  
492 tropospheric ozone experiment and (c) stratospheric ozone only experiment as well as (e) the  
493 difference between the two indicating the effect of tropospheric ozone change. (right column)  
494 Same as the left column but for heave changes.

495

496 **Methods**

497 **Observations**

498 To evaluate the performance of CanESM5 in simulating the historical warming in the Southern  
499 Ocean during the second half of the twentieth century, we use one objectively analyzed ocean  
500 dataset, the Institute of Atmospheric Physics (IAP) ocean temperature analysis<sup>41</sup>. The IAP ocean  
501 temperature analysis has global ocean coverage of 1-degree horizontal resolution on 41 vertical  
502 levels from the surface down to 2000 m. It has a monthly resolution from 1940 to the present.  
503 This ocean temperature analysis minimizes the errors from ocean sampling by *in situ*  
504 observations and allows for accurate estimates of regional and global OHC changes during the  
505 past several decades, especially those in the Southern Ocean.

506

507 **CMIP5 and CMIP6 preindustrial control, historical and ozone single-forcing simulations**

508 We use the preindustrial control runs of four CMIP5 (CCSM4, CESM1-CAM5, FGOALS-g2  
509 and GISS-E2-H) and four CMIP6 (CanESM5, GISS-E2-1-G, IPSL-CM6A-LR and MIROC6)  
510 models. For either CMIP5 or CMIP6, the four-model ensemble has an average transient climate  
511 response (TCR)<sup>42,43,44</sup> that is very close to the mean TCR reported by previous studies<sup>43,44</sup>,  
512 suggesting that the models we used can well represent the transient climate sensitivity of the  
513 models of either generation. For all the models except CESM1-CAM5 and GISS-E2-1-G, we  
514 estimate each model's climate drift in ocean temperatures as a 500-year temperature trend  
515 (during the last 500 years) in each model's preindustrial control run. As CESM1-CAM5 and  
516 GISS-E2-1-G only have 320 and 345 years of simulation available in the CMIP5 and CMIP6  
517 archives, for either model, we estimate its climate drift in ocean temperature as the temperature  
518 trend during the last 200 years of the preindustrial control run. For CanESM5 and GISS-E2-1-G,  
519 the preindustrial simulations of “p1” and “f2” are adopted to be consistent with their ozone

520 experiments, respectively. We remove climate drifts from the trends of ocean temperatures in the  
521 historical and ozone single-forcing simulations with these CMIP5 and CMIP6 models. We also  
522 remove the climate drift in ocean salinity for CanESM5 (the salinity trend of its preindustrial  
523 control run) when conducting the spiciness and heave decomposition.

524

525 The CMIP5 historical simulations are performed including all the natural and anthropogenic  
526 forcings during the historical period. The CMIP5 ozone single-forcing experiments on the other  
527 hand are forced by stratospheric and tropospheric ozone only during the historical period while  
528 the other forcings are fixed at their preindustrial levels<sup>45</sup>. In the four CMIP5 models, the ozone  
529 chemistries are either semi-offline calculated or prescribed<sup>12</sup>. In this study, we adopt 11  
530 ensemble members of ozone single-forcing (stratospheric and tropospheric ozone) experiments  
531 (2 from CCSM4, 3 from CESM1-CAM5, 1 from FGOALS-g2 and 5 from GISS-E2-H) and 21  
532 ensemble members of historical simulations with the four CMIP5 models (6 from CCSM4, 4  
533 from CESM1-CAM5, 5 from FGOALS-g2 and 6 from GISS-E2-H). Note here, for CCSM4  
534 ozone experiment, there are three ensemble members while temperature outputs in 2000-2005  
535 are not available for one member in the CMIP5 archives; so only the other two ensemble  
536 members are used. For CCSM4 and GISS-E2-H preindustrial and historical simulations,  
537 ensembles of “p1” perturbation are adopted to be consistent with the perturbation in the ozone  
538 experiments. We calculate the ensemble mean for each model and then calculate the multi-model  
539 mean (MMM) based on the ensemble means of the four models to minimize the effects of  
540 internal climate variability and model differences. The inter-model difference is estimated as one  
541 standard deviation of the ensemble means of the models.

542

543 The ozone single-forcing experiments with CMIP6 models are akin to their historical simulations  
544 but forced by stratospheric ozone variations only. For models without coupled chemistry, they  
545 prescribe the same stratospheric ozone concentrations as used in their historical simulations<sup>46</sup>.  
546 For models with coupled chemistry, their chemistry schemes are turned off. Note here, that while  
547 these model configurations neglect to represent potential feedbacks of changing dynamics on the  
548 ozone fields in a self-consistent way, we consider such effects to be of second-order relevance.  
549 Such an assumption is justified given that the climate response, for example, the response of the  
550 polar vortex breakdown to equivalent effective stratospheric chlorine, does not show a  
551 systematic difference between models with prescribed and interactive ozone<sup>47</sup>.  
552

553 The CMIP6 models prescribe the ensemble mean monthly mean three-dimensional stratospheric  
554 ozone concentrations as simulated in their historical runs but have fixed three-dimensional  
555 long-term monthly mean tropospheric ozone concentrations from their preindustrial control runs.  
556 In particular, grid cells are categorized tropospheric when they have an ozone concentration  
557 below 100 ppbv (parts per billion by volume) in the climatology of the preindustrial control run.  
558 This definition of the troposphere is consistent throughout the historical period and facilitates  
559 inter-model comparisons<sup>48</sup>. Albeit the tropopause height may alter with climate change, several  
560 studies<sup>6,17</sup> suggest that the tropopause choice only has a marginal effect on radiative forcing. To  
561 examine the ozone impacts on Southern Ocean interior warming during the second half of the  
562 twentieth century, we adopt 28 ensemble members of ozone single-forcing (stratospheric ozone  
563 only) experiments with the four CMIP6 models (10 from CanESM5, 5 from GISS-E2-1-G, 10  
564 from IPSL-CM6A-LR and 3 from MIROC6) and calculate the MMM and inter-model difference  
565 of the CMIP6 models.

566

567 Besides, we compare the transient climate responses (TCRs) between CMIP5 and CMIP6  
568 models. For CMIP5 models<sup>42,43</sup>, the TCRs of CCSM4, CESM1-CAM5, FGOALS-g2 and  
569 GISS-E2-H are 1.7 K, 2.33 K, 1.4 K and 1.7 K, so their average TCR is 1.78 K. For CMIP6  
570 models<sup>44</sup>, the TCRs of CanESM5, GISS-E2-1-G, IPSL-CM6A-LR and MIROC6 are 2.66 K,  
571 1.68 K, 2.32 K and 1.52 K, so their average TCR is 2.05 K. Both averages are very close to the  
572 mean TCRs reported by previous studies<sup>43,44</sup> based on 29 CMIP5 models and 34 CMIP6 models,  
573 respectively. This result suggests that, for either CMIP5 or CMIP6, the four-model ensemble  
574 well represents the transient climate sensitivity of the models of either generation. The Student's  
575 t-test result further shows that the difference of TCR between CMIP5 and CMIP6 model means  
576 is insignificant at the 95% confidence level, which suggests that there is no statistically  
577 significant difference in transient climate sensitivity between the CMIP5 and CMIP6 models  
578 used in the current study.

579

## 580 **CanESM5 and associated simulations**

581 CanESM5 is a fully coupled climate model participating in CMIP6<sup>49</sup>. The atmosphere  
582 component is the Canadian Atmosphere Model (CanAM5), which employs a spectral dynamical  
583 core with a T63 truncation (an approximate 2.8-degree horizontal resolution) and a hybrid  
584 sigma-pressure coordinate with 49 vertical layers up to about 1 hPa. The land component  
585 incorporates the Canadian Land Surface Scheme (CLASS) and the Canadian Terrestrial  
586 Ecosystem Model (CTEM). The ocean component is a modified version of the Nucleus for  
587 European Modelling of the Ocean model (NEMO), which includes ocean biogeochemistry  
588 represented by the Canadian Model of Ocean Carbon (CMOC) and employs a ~1-degree

589 horizontal resolution and 45 vertical levels. The Louvain-la-Neuve sea-Ice Model version 2  
590 (LIM2) also operates within the NEMO framework.

591

592 A 25-member historical simulation labeled as perturbed physics member 1 (“p1”) has been  
593 performed with CanESM5 during 1850-2014. Individual ensemble members are initialized at  
594 different years from preindustrial control run and perturbed by the conservative remapping  
595 wind-stress fields. We use these 25 ensembles of CanESM5 historical simulation as they share  
596 the same perturbation scheme (“p1”) with CanESM5 ozone simulations. We compare the trend  
597 of zonal mean temperature in the ensemble mean of the CanESM5 historical simulation with that  
598 in the IAP data during 1955-2000 and find that the CanESM5 historical simulation is able to well  
599 capture the observed warming tongue ( $>0.03$  K/decade) in the upper 1000 m between 40°S and  
600 50°S (Extended Data Fig. 8). This result demonstrates the model fidelity in simulating the  
601 Southern Ocean temperature response to external climate forcings.

602

603 Besides the 10-ensemble stratospheric ozone only experiment as in line with several other  
604 CMIP6 models, CanESM5 provides a 10-ensemble member historical total ozone-only  
605 experiment in which the model prescribes the monthly mean three-dimensional ozone  
606 concentrations from the historical simulation through the depth of the atmosphere. This total  
607 ozone-only experiment is consistent with the CMIP5 ozone single-forcing (stratospheric and  
608 tropospheric ozone) experiments. We adopt the simulations of this pair of ozone experiments to  
609 isolate and quantify the effects of stratospheric and tropospheric ozone on Southern Ocean  
610 interior warming.

611

612 **The spiciness and heave decomposition**

613 The spiciness and heave decomposition follows previous studies<sup>35,36</sup>. For changes in potential  
614 temperature ( $\theta$ ) and salinity ( $S$ ) at depth  $z$ , i.e.,  $\theta'|_z$  and  $S'|_z$ , they can be decomposed as:

615 
$$\theta'|_z \cong \theta'|_n + N'\theta_z \quad (1)$$

616 
$$S'|_z \cong S'|_n + N'S_z \quad (2)$$

617 where  $\theta'|_n$  and  $S'|_n$  denote the spiciness changes of temperature and salinity that are  
618 density-compensating along neutral density surfaces;  $N'\theta_z$  and  $N'S_z$  denote the heave changes  
619 of temperature and salinity that are related to the neutral density surface height change  $N'$   
620 (positive downward).

622 **The OHC calculation**

623 At each location, the OHC within a layer between the depths  $z_1$  and  $z_2$  is calculated as

624 
$$OHC = \rho_0 C_p \int_{z_1}^{z_2} \theta dz \quad (3)$$

625 where  $\rho_0$  denotes sea water density and  $C_p$  denotes the specific heat capacity of sea water.

627 **The statistical significance test**

628 We examine the statistical significance of climate response to ozone forcing in CanESM5 based  
629 on the Student's t-test. We divide 500 years of CanESM5 preindustrial simulation into 10  
630 truncations and treat each truncation as one ensemble member. Hence we construct 10  
631 preindustrial ensembles with non-overlapping 50-year periods. We apply the Student's t-test to  
632 the three pairs of ensemble simulations—total-ozone versus preindustrial, stratospheric-ozone  
633 versus preindustrial and total-ozone versus stratospheric-ozone—to estimate the statistical  
634 significance of total, stratospheric and tropospheric ozone effects. Besides, we examine the

635 statistical significances of trends of CMIP5 and CMIP6 MMMs and CanESM5 ensemble means  
636 based on the Mann-Kendall trend significance test.

637

638 **Data availability**

639 All the raw CMIP5 model simulation data are publically available at

640 <https://esgf-node.llnl.gov/search/cmip5/>

641 All the raw CMIP6 model simulation data are publically available at

642 <https://esgf-node.llnl.gov/projects/cmip6/>

643 The IAP observation data are publically available at

644 <http://www.ocean.iap.ac.cn/>

645

646 **Code availability**

647 Figures are generated via the NCAR Command Language (NCL, Version 6.5.0) [Software].

648 (2018). Boulder, Colorado: UCAR/NCAR/CISL/TDD. <http://dx.doi.org/10.5065/D6WD3XH5>

649 The codes and processed variables to generate Figures 1-6 are available at Zenodo<sup>50</sup>.

650

651 **Methods References**

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