

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

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10	Stratospheric ozone depletion and tropospheric ozone increases drive Southern Ocean
11	interior warming
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Atmospheric ozone has undergone distinct changes in the stratosphere and troposphere 32 during the second half of the twentieth century, with depletion in the stratosphere and an 33 increase in the troposphere. Until now, the effect of these changes on ocean heat uptake has 34 been unclear. Here we show that both stratospheric and tropospheric ozone changes have 35 contributed to Southern Ocean interior warming, with the latter being more important. The 36 ozone changes between 1955 and 2000 induced about 30% of the net simulated ocean heat 37 content increase in the upper 2000 m of the Southern Ocean, with around 60% attributed to 38 tropospheric increases and 40% to stratospheric depletion. Moreover, these two warming 39 contributions show distinct physical mechanisms: Tropospheric ozone increases cause a 40 subsurface warming in the Southern Ocean primarily via the deepening of isopycnals, while 41 stratospheric ozone depletion via spiciness changes along isopycnals. Our results highlight 42 that tropospheric ozone is more than an air pollutant and, as a greenhouse gas, has been 43 pivotal to the Southern Ocean warming. 44

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Atmospheric ozone has experienced distinct changes in the stratosphere and troposphere during 46 the second half of the twentieth century. Notable ozone depletion has occurred in the stratosphere, 47 most strikingly as the ozone hole over Antarctica, which has been attributed primarily to 48 anthropogenic emissions of ozone-depleting substances^{1,2,3,4}. In contrast, ozone increases in the 49 troposphere have been observed (Extended Data Fig. 1) as a result of anthropogenic emissions of 50 ozone precursors such as methane, non-methane volatile organic compounds, carbon monoxide 51 and nitrogen oxides^{5,6,7,8}. These atmospheric ozone changes have profound impacts on Earth's 52 climate system. For example, stratospheric ozone depletion has significantly altered the 53 tropospheric circulation by displacing the Southern Hemisphere westerly winds poleward during 54

austral summer^{9,10,11,12}, though these Southern Hemisphere circulation trends paused around 2000
 and are expected to reverse the sign owing to reduced emissions of ozone depleting substances
 following the signing of the Montreal Protocol and its Amendments^{13,14,15}. By contrast, ozone
 impacts on oceans, especially those due to tropospheric ozone changes, are relatively less well
 explored.

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The Fifth Assessment Report of the United Nations Intergovernmental Panel on Climate Change 61 indicates that ozone constitutes the third-most important contribution to greenhouse gas forcing 62 since pre-industrial times after carbon dioxide and methane¹⁶. Stratospheric and tropospheric 63 ozone changes substantially modulate Earth's radiation balance¹⁷, and thus could also affect 64 global ocean heat uptake. The role of the Southern Ocean is critical in the context of climate 65 change as it is one of the most important regions for taking up excess heat in a warming 66 climate^{18,19}, and is markedly affected by Southern Hemisphere westerly winds^{20,21,22,23}. During the 67 past several decades, the Southern Ocean has shown a rapid subsurface warming^{24,25}, only a small 68 part of which, however, has been attributed to stratospheric ozone depletion^{26,27,28}. Given the 69 concurrent (but opposite) ozone changes in both the stratosphere and troposphere, one gap remains 70 71 in our current knowledge of ozone-driven Southern Ocean warming: The impact of the increase in tropospheric ozone. Here, we employ historical simulations and accompanying ozone 72 single-forcing experiments with a broad set of climate models from the Coupled Model 73 Intercomparison Projects Phase 5/6 (CMIP5/6) to probe the mechanisms and impacts of 74 stratospheric and tropospheric ozone changes on Southern Ocean interior warming during the 75 second half of the twentieth century. 76

78 **Results**

We first examine the ozone single-forcing experiments from the CMIP5 models in which the 79 models were only forced with historical integrations of atmospheric ozone concentrations instead 80 of all historical forcings (see Methods). These ozone single-forcing experiments demonstrate the 81 effects of both stratospheric and tropospheric ozone changes together. Between 1955 and 2000, 82 ozone depletion generates a strong stratospheric cooling trend in the Southern Hemisphere high 83 latitudes (Extended Data Fig. 2), which leads to a poleward intensification of Southern 84 Hemisphere westerly winds in the troposphere reminiscent of "annular mode-like" responses²⁹ 85 (Fig. 1a). Along with the response in the atmosphere, ozone changes also produce a pronounced 86 subsurface warming in the Southern Ocean. Within 40-50°S, the warming rate is larger than 0.01 87 K/decade in the upper 1000 m (Fig. 1c). When we integrate ocean heat content (OHC) over the 88 upper 2000 m between 30°S and 60°S where ocean warming mainly occurs, we find a significant 89 increase of OHC, with a trend of 5.63 ± 2.36 ZJ/decade (1 ZJ = 10^{21} joule; multi-model mean ± 1 90 standard deviation among models, see Methods) between 1955 and 2000 (Fig. 2a). Our results 91 from these CMIP5 model simulations thus suggest a substantial Southern Ocean subsurface 92 warming in response to stratospheric and tropospheric ozone changes. 93

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

westerly winds, which indicates a major role of stratospheric ozone depletion in the atmospheric 101 response during 1955-2000 (Fig. 1b). However, in the Southern Ocean, we find a much weaker 102 subsurface warming in the CMIP6 stratospheric ozone only experiments with a pattern consistent 103 with previous studies^{27,30}. Between 40°S and 50°S, the warming rate is much smaller than 0.01 104 K/decade in the upper 2000 m (Fig. 1d). The upper 2000-m OHC between 30°S and 60°S 105 exhibits a marginal increase between 1955 and 2000, with a trend of 0.45±1.22 ZJ/decade 106 (multi-model mean ± 1 standard deviation among models; Fig. 2a). We further find no 107 statistically significant difference in transient climate sensitivity between the CMIP5 and CMIP6 108 models (see Methods) but the Southern Ocean OHC trend in the CMIP5 simulations is one order 109 of magnitude larger than that in the CMIP6 simulations, indicating that the difference in model 110 climate sensitivity cannot serve as the major cause of such distinct warming trends in the 111 Southern Ocean. On the other hand, the comparison between CMIP5 and CMIP6 model 112 simulations implies that the tropospheric ozone increase is a key driver of Southern Ocean 113 interior warming. Nevertheless, it is worth noting that this comparison cannot allow for a 114 conclusive quantification of the impact nor shed light on the mechanism of tropospheric ozone 115 increases on Southern Ocean warming, since the differences in prescribed historical ozone 116 117 datasets between CMIP5 and CMIP6 models (Extended Data Fig. 1) and model responses to ozone forcing would need to be considered. 118

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120 Quantifying the ozone impacts on Southern Ocean warming

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

warming generally in alignment with the ensemble mean result of CMIP6 stratospheric ozone 124 only experiments (Fig. 2a). The first CanESM5 ensemble is forced with historical changes in 125 both stratospheric and tropospheric ozone, equivalent to the CMIP5 simulations described above 126 but adopting the CMIP6 simulation protocol^{31,32}. The second ensemble simulation is equivalent 127 to the CMIP6 ozone experiments described above in which the model is forced with historical 128 integrations of solely stratospheric ozone changes (see Methods). The difference between the 129 two ensemble simulations therefore isolates the effect of tropospheric ozone change. Relative to 130 preindustrial times, we find that both ensemble experiments from CanESM5 simulate a 131 stratospheric cooling in the southern high latitudes and a significant poleward intensification of 132 Southern Hemisphere westerlies in the troposphere between 1955 and 2000 (Fig. 3a,b). On the 133 other hand, tropospheric ozone increases lead to a warming in the troposphere and a cooling in 134 the stratosphere³³ (Extended Data Fig. 3), together with a significant upward intensification of 135 Southern Hemisphere westerly winds in the upper levels and a poleward intensification of 136 westerly winds towards surface (Fig. 3c). These tropospheric-ozone-produced atmosphere 137 temperature and circulation changes are comparable to those induced by stratospheric ozone 138 depletion towards the surface layers, suggesting that tropospheric ozone changes could 139 140 potentially have considerable impacts on the oceans underneath.

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

ozone depletion, which is, however, mostly limited to the upper 600 m (Fig. 3e). On the other 147 hand, the vertical extension of the tongue of warming waters depends essentially on tropospheric 148 ozone forcing. The increase of tropospheric ozone creates such a deep warming in the Southern 149 Ocean that warming larger than 0.01 K/decade is found to penetrate as deep as 1000 m within 150 40-50°S (Fig. 3f). To the north of the tongue of warming waters, there is a tongue of cooling 151 waters in the upper levels of the Southern Ocean, which results principally from stratospheric 152 ozone depletion and secondarily from tropospheric ozone increases (Fig. 3e,f). A similar cooling 153 feature is found at high latitudes south of 55°S (Fig. 3d,f). It is worth noting that the warming 154 pattern due to tropospheric ozone increases is different from that due to the rising well-mixed 155 greenhouse gases such as carbon dioxide. The rise of well-mixed greenhouse gases induces 156 ubiquitous while vertically decaying warming in the upper 2000-m ocean in the Southern 157 Ocean^{20,21,27,34}. 158

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Here we estimate the OHC variations in the upper 2000 m between 30° S and 60° S in the two sets 160 of CanESM5 simulations. We find that atmospheric ozone changes induce a robust upward OHC 161 trend of 4.58 ± 3.35 ZJ/decade (ensemble mean ± 1 standard deviation among ensembles) between 162 163 1955 and 2000, of which about two-fifths (1.84 \pm 2.50 ZJ/decade, ensemble mean \pm 1 standard deviation among ensembles) can be attributed to stratospheric ozone depletion while the other 164 three-fifths (2.74 ZJ/decade, the difference of the ensemble means between the two suites of 165 ozone simulations) is driven by tropospheric ozone increases. Our results confirm the importance 166 of tropospheric ozone to Southern Ocean heat uptake and storage. Importantly, the increases in 167 tropospheric ozone have been more effective in driving the interior warming over the Southern 168 Ocean during the second half of the twentieth century compared to stratospheric ozone depletion. 169

Moreover, to set in context the effect of ozone forcing on the historical OHC increase in the 171 Southern Ocean over the 1955-2000 period, we compare OHC changes in the upper 2000 m 172 within 30-60°S between the CanESM5 historical simulations that also include the other 173 greenhouse gas forcings from carbon dioxide, methane and nitrous oxide and the combined 174 stratosphere-troposphere ozone single-forcing experiment (Fig. 2b). We find that CanESM5 175 simulates a general long-term increase of OHC in the Southern Ocean, at a rate of 13.77±4.29 176 ZJ/decade (ensemble mean ± 1 standard deviation among ensembles) between 1955 and 2000, 177 which is consistent with the OHC trends inferred from observations (16.25 ZJ/decade) and 178 CMIP5 models (14.60 \pm 5.27 ZJ/decade, multi-model mean \pm 1 standard deviation among models) 179 (Fig. 2b). Using the CanESM5 ozone experiment, we further find that about 33.2% of the net

historical OHC increase between 1955 and 2000 is caused by atmospheric (both stratospheric 181 and tropospheric) ozone changes. This ratio is in line with that suggested by the historical 182 CMIP5 model ozone experiment ($38.4 \pm 10.4\%$, multi-model mean ± 1 standard deviation among 183 models). 184

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Physical mechanisms of ozone-driven Southern Ocean warming 186

To further understand the mechanisms by which stratospheric and tropospheric ozone changes 187 drive Southern Ocean interior warming, we decompose the temperature and salinity changes 188 between 1955 and 2000 at depth levels into the spiciness changes along isopycnals and 189 heave-related changes owing to the vertical heave of isopycnals³⁵ (see Methods). The spiciness 190 reveals alterations in water mass properties as a result of the subduction of surface temperature 191 and salinity anomalies and changes by interior mixing processes. The heave of isopycnals could 192

be linked to changes in wind-driven ocean circulation and the redistribution of heat and salt in
 the interior ocean³⁶.

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We first depict the temperature and salinity responses to total atmospheric ozone variations. 196 During 1955-2000, the zonally averaged spiciness changes on density surfaces exhibit strong 197 warming and salification trends in the upper ocean toward isopycnal outcrops, especially in the 198 latitudes between 40°S and 60°S (Fig. 4a,b). These warming and salification trends primarily 199 result from stratospheric ozone depletion and not from tropospheric ozone increases (Fig. 4c,d). 200 Between 40°S and 60°S, the Southern Ocean takes heat from the atmosphere but loses freshwater 201 in response to stratospheric ozone depletion (Fig. 5a,b), both contributing to warming and 202 salification spiciness trends³⁶. The peak of Southern Ocean surface heat uptake is around 55°S 203 (Extended Data Fig. 4), essentially due to the increase of downward turbulent latent heat flux³⁷ 204 over the Indian Ocean sector (Extended Data Fig. 5). Increases in surface shortwave radiation 205 fluxes also contribute to Southern Ocean heat uptake in these latitudes (Extended Data Fig. 3). 206 On the other hand, the reduction in surface freshwater flux can be mostly attributed to changes in 207 precipitation minus evaporation (P-E) to the north of 54°S but is likely related to sea-ice 208 variations to the south (Extended Data Fig. 6). Between 40°S and 54°S, the P-E reduction results 209 from both precipitation decreases and evaporation increases and is especially robust over the 210 Pacific sector (Extended Data Fig. 7). 211

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Besides the warming and salification trends in the isopycnal outcropping region between 40° S and 60° S, we also find cooling and freshening spiciness changes to the north of 40° S on density surfaces between 26.3 and 27.0 kg/m³ (Fig. 4a,b), within the density ranges of the Subantarctic

Mode Water (SAMW) and Antarctic Intermediate Water (AAIW) simulated by climate models³⁸. Particularly, the spiciness changes in the AAIW density range (26.7-27.0 kg/m³) can be attributed mostly to stratospheric ozone depletion (Fig. 4c,d) while those in the SAMW density range (26.3-26.6 kg/m³) mainly to tropospheric ozone increases (Fig. 4e,f). These ozone-induced cooling signals contribute to the cooling trend found at corresponding locations from observations and historical simulations during 1955-2000 (Extended Data Fig. 8).

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

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We further analyze the heave component of Southern Ocean temperature change. We find a 230 subsurface warming region (>0.01 K/decade) extending equatorward between 36°S and 51°S and 231 232 downward in 300-1100 m (Fig. 6b) in response to atmospheric ozone changes, accompanied by a cooling tongue to the north and in upper levels. This pair of warming and cooling anomalies has 233 been linked to poleward intensified surface westerly winds and indicates heat redistribution 234 within the Southern Ocean^{21,39}. Specifically, stratospheric ozone depletion drives an 235 intensification of surface westerly winds at and to the south of the Antarctic Circumpolar Current 236 but a relaxation to the north (Fig. 5c, Extended Data Fig. 9), in a pattern consistent with other 237 CMIP6 models (Extended Data Fig. 10). The zonally averaged zonal wind change exhibits a 238

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

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Tropospheric ozone increases, on the other hand, engender different changes in surface winds 247 from those due to stratospheric ozone depletion (Fig. 5c, Extended Data Fig. 9). The zonally 248 averaged surface zonal wind change also reflects a dipole-like pattern but located more 249 northward, with positive and negative anomalies occurring to the south and north of around 42°S. 250 This pattern indicates less poleward displaced surface westerlies than their counterparts driven 251 by stratospheric ozone depletion. Tropospheric ozone increases also drive poleward-intensified 252 Southern Hemisphere precipitation and significantly increase evaporation at lower latitudes where 253 the tropospheric ozone increases are stronger (Extended Data Fig. 6f). In the ocean, the 254 255 wind-driven Ekman pumping (Extended Data Fig. 9d) produces isopycnals deepening in much lower latitudes, around 42°S, and warming heave changes there (Fig. 6f). These heave-related 256 warming changes are much stronger than those induced by stratospheric ozone depletion, which 257 is likely due to the fact that the oceanic thermocline is more strongly stratified at lower 258 latitudes³⁶, allowing the wind-driven downwelling more effectively to create warming heave 259 changes there. 260

262 Discussion

In summary, we have examined the climate impacts of atmospheric ozone changes during the 263 second half of the twentieth century, with a focus on disentangling effects of stratospheric ozone 264 depletion and tropospheric ozone increases. We show that while stratospheric ozone depletion 265 plays a dominant role in atmospheric temperature and wind changes in southern high latitudes in 266 the stratosphere and upper levels of the troposphere, tropospheric ozone increases have made a 267 larger contribution to Southern Ocean interior warming. Between 1955 and 2000, about one-third 268 of the historical OHC increase in the upper 2000 m of the Southern Ocean between 30°S and 269 60°S was induced by atmospheric ozone changes, of which around three-fifths can be attributed 270 to tropospheric ozone increases and the other two-fifths to stratospheric ozone depletion. 271 Tropospheric ozone increases cause Southern Ocean subsurface warming primarily via the 272 deepening of isopycnals. They give rise to an intensification of surface westerly winds over the 273 Southern Ocean such that the wind-driven Ekman pumping brings about isopycnal deepening 274 275 around 42°S and prompts heave-induced warming there. On the other hand, stratospheric ozone depletion promotes warming in the Southern Ocean mainly through spiciness changes along 276 isopycnals in the upper 500 m. In response to stratospheric ozone depletion, the net surface 277 278 downward heat flux increases but the freshwater flux decreases over the Southern Ocean between 40°S and 60°S, contributing to the warming and salification spiciness changes in the 279 isopycnal outcropping regions of the Southern Ocean. 280

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

depleting substances (ODSs)⁴⁰. However, the response to ODSs, inferred by differencing 285 historical simulations with all anthropogenic forcings and simulations with fixed ODSs, omits 286 changes in tropospheric ozone induced by precursor omissions, but includes radiative effects of 287 ODSs themselves, and hence these results are not directly comparable with our study of the 288 direct effects of tropospheric and stratospheric ozone changes. Furthermore, our results suggest 289 that, when the effect of tropospheric ozone increases is considered, the ozone impacts on 290 Southern Ocean interior warming are much larger than previous estimates that only considered 291 stratospheric ozone depletion²⁷. Between 1955 and 2000, tropospheric ozone increases 292 significantly affect the P-E over the Southern Ocean. As such, our results highlight that 293 tropospheric ozone, besides being an air pollutant, is an important contributor to ocean heat 294 uptake and hydrological cycle change in the Southern Hemisphere. 295

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427	W.L. conceived the study, performed the analysis and wrote the original draft of the manuscript.					
428	S.L. contributed to the analysis. W.L., M.I.H., R.CG., S.L., N.P.G., K.L., X.Z. and N.C.S.					
429	contributed to interpreting the results and made substantial improvements to the manuscript.					
430						
431	Competing Interests Statement					
432	The authors declare no competing interests.					
433						

434 Figure legends

Figure 1. Changes in Southern Hemisphere westerlies and Southern Ocean temperature in 435 response to ozone changes in CMIP5 and CMIP6 simulations. (top row) Trends of annual and 436 zonal mean zonal winds during 1955-2000 (shading in m/s/decade) of the multi-model means 437 (MMMs) from (a) CMIP5 and (b) CMIP6 models ozone single-forcing experiments. The annual 438 climatologies of zonal mean zonal winds (contour in m/s, with an interval of 5 m/s and the zero 439 contours thickened) of the MMMs from CMIP5 and CMIP6 models preindustrial control runs 440 are superimposed on both panels, respectively. (bottom row) Trends of annual and zonal ocean 441 temperature during 1955-2000 (shading in K/decade) of the MMMs from (c) CMIP5 and (d) 442 CMIP6 models ozone single-forcing experiments. Stippling indicates that the change is 443 statistically insignificant at the 95% confidence level of the Mann-Kendall trend significance test 444 (see Methods). 445

446

Figure 2. Observed and simulated Southern Ocean heat content. (a) Ocean heat content 447 (OHC) anomalies (relative to the value in 1955) integrated over the upper 2000 m between 30°S 448 and 60°S from ozone single-forcing experiments with four CMIP5 models (MMM, medium blue; 449 inter-model spread, light blue; see Methods) and four CMIP6 models (MMM, magenta; 450 inter-model spread, light magenta), and the ensemble means from CanESM5 stratospheric and 451 tropospheric ozone experiment (blue), stratospheric ozone only experiment (red) as well as the 452 difference between the two (black) indicating the effect of tropospheric ozone change. The 453 inter-model spread is calculated as one standard deviation of the ensemble means of individual 454 models. (b) Same as panel (a) but for OHC anomalies from the IAP observation data (orange) 455 and historical simulations with the four CMIP5 models (MMM, medium purple; inter-model 456 spread, light purple) and CanESM5 (ensemble mean, purple). OHC anomalies from CMIP5 457

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

461

Figure 3. Changes in Southern Hemisphere westerlies and Southern Ocean temperature in 462 response to ozone changes in CanESM5 simulations. (top row) Changes in annual and zonal 463 mean zonal winds (shading in m/s) during 1955-2000 (relative to preindustrial control run) for 464 the ensemble means in CanESM5 (a) stratospheric and tropospheric ozone experiment and (b) 465 stratospheric ozone only experiment as well as (c) the difference between the two indicating the 466 effect of tropospheric ozone change. Stippling indicates that the change is statistically 467 insignificant at the 95% confidence level of the Student's t-test (see Methods). (bottom row) 468 Same as the top row but for trends of annual and zonal mean ocean temperature during 469 1955-2000 (shading in K/decade). Stippling indicates that the change is statistically insignificant 470 at the 95% confidence level of the Mann-Kendall trend significance test (see Methods). 471

472

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

Figure 5. Surface heat flux, freshwater flux and zonal winds changes in CanESM5 ozone 481 experiments. (a) Changes in annual and zonal mean net surface heat fluxes over the Southern 482 Ocean during 1955-2000 (relative to preindustrial control run) for the ensemble means in 483 CanESM5 stratospheric and tropospheric ozone experiment (light blue; significant, blue) and 484 stratospheric ozone only experiment (orange; significant, red) as well as the difference between 485 the two (gray; significant, black) indicating the effect of tropospheric ozone change. Panels (b) 486 and (c) are the same as (a) but for changes in annual and zonal mean net surface freshwater 487 fluxes over the ocean and surface zonal winds. The variable of surface zonal wind is obtained 488 from atmosphere model outputs and land is then masked out for the variable so that winds are on 489 the liquid ocean water surface in most parts of the Southern Ocean but on sea ice surface around 490 or south of 60°S where sea ice exists. Heat and freshwater fluxes are positive downward. In all 491 the panels, changes are tested based on the Student's t-test and denoted statistically significant 492 when exceeding the 95% confidence level (see Methods). 493

494

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

502

503 Methods

504 **Observations**

To evaluate the performance of CanESM5 in simulating the historical warming in the Southern 505 Ocean during the second half of the twentieth century, we use one objectively analyzed ocean 506 dataset, the Institute of Atmospheric Physics (IAP) ocean temperature analysis⁴¹. The IAP ocean 507 temperature analysis has global ocean coverage of 1-degree horizontal resolution on 41 vertical 508 levels from the surface down to 2000 m. It has a monthly resolution from 1940 to the present. This 509 ocean temperature analysis minimizes the errors from ocean sampling by in situ observations and 510 allows for accurate estimates of regional and global OHC changes during the past several decades, 511 especially those in the Southern Ocean. 512

513

CMIP5 and CMIP6 preindustrial control, historical and ozone single-forcing simulations 514 We use the preindustrial control runs of four CMIP5 (CCSM4, CESM1-CAM5, FGOALS-g2 515 and GISS-E2-H) and four CMIP6 (CanESM5, GISS-E2-1-G, IPSL-CM6A-LR and MIROC6) 516 models. For either CMIP5 or CMIP6, the four-model ensemble has an average transient climate 517 response (TCR)^{42,43,44} that is very close to the mean TCR reported by previous studies^{43,44}, 518 suggesting that the models we used can well represent the transient climate sensitivity of the 519 520 models of either generation. For all the models except CESM1-CAM5 and GISS-E2-1-G, we estimate each model's climate drift in ocean temperatures as a 500-year temperature trend 521 (during the last 500 years) in each model's preindustrial control run. As CESM1-CAM5 and 522 GISS-E2-1-G only have 320 and 345 years of simulation available in the CMIP5 and CMIP6 523 archives, for either model, we estimate its climate drift in ocean temperature as the temperature 524 trend during the last 200 years of the preindustrial control run. For CanESM5 and GISS-E2-1-G, 525 the preindustrial simulations of "p1" and "f2" are adopted to be consistent with their ozone 526

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

531

The CMIP5 historical simulations are performed including all the natural and anthropogenic 532 forcings during the historical period. The CMIP5 ozone single-forcing experiments on the other 533 hand are forced by stratospheric and tropospheric ozone only during the historical period while 534 the other forcings are fixed at their preindustrial levels⁴⁵. In the four CMIP5 models, the ozone 535 chemistries are either semi-offline calculated or prescribed¹². In this study, we adopt 11 536 ensemble members of ozone single-forcing (stratospheric and tropospheric ozone) experiments 537 (2 from CCSM4, 3 from CESM1-CAM5, 1 from FGOALS-g2 and 5 from GISS-E2-H) and 21 538 ensemble members of historical simulations with the four CMIP5 models (6 from CCSM4, 4 539 from CESM1-CAM5, 5 from FGOALS-g2 and 6 from GISS-E2-H). Note here, for CCSM4 540 ozone experiment, there are three ensemble members while temperature outputs in 2000-2005 541 are not available for one member in the CMIP5 archives; so only the other two ensemble 542 543 members are used. For CCSM4 and GISS-E2-H preindustrial and historical simulations, ensembles of "p1" perturbation are adopted to be consistent with the perturbation in the ozone 544 experiments. We calculate the ensemble mean for each model and then calculate the multi-model 545 mean (MMM) based on the ensemble means of the four models to minimize the effects of 546 internal climate variability and model differences. The inter-model difference is estimated as one 547 standard deviation of the ensemble means of the models. 548

The ozone single-forcing experiments with CMIP6 models are akin to their historical simulations 550 but forced by stratospheric ozone variations only. For models without coupled chemistry, they 551 prescribe the same stratospheric ozone concentrations as used in their historical simulations⁴⁶. 552 For models with coupled chemistry, their chemistry schemes are turned off. Note here, that while 553 these model configurations neglect to represent potential feedbacks of changing dynamics on the 554 ozone fields in a self-consistent way, we consider such effects to be of second-order relevance. 555 Such an assumption is justified given that the climate response, for example, the response of the 556 polar vortex breakdown to equivalent effective stratospheric chlorine, does not show a 557 systematic difference between models with prescribed and interactive ozone⁴⁷. 558

559

The CMIP6 models prescribe the ensemble mean monthly mean three-dimensional stratospheric 560 ozone concentrations as simulated in their historical runs but have fixed three-dimensional 561 long-term monthly mean tropospheric ozone concentrations from their preindustrial control runs. 562 In particular, grid cells are categorized tropospheric when they have an ozone concentration 563 below 100 ppbv (parts per billion by volume) in the climatology of the preindustrial control run. 564 This definition of the troposphere is consistent throughout the historical period and facilitates 565 inter-model comparisons⁴⁸. Albeit the tropopause height may alter with climate change, several 566 studies^{6,17} suggest that the tropopause choice only has a marginal effect on radiative forcing. To 567 examine the ozone impacts on Southern Ocean interior warming during the second half of the 568 twentieth century, we adopt 28 ensemble members of ozone single-forcing (stratospheric ozone 569 only) experiments with the four CMIP6 models (10 from CanESM5, 5 from GISS-E2-1-G, 10 570 from IPSL-CM6A-LR and 3 from MIROC6) and calculate the MMM and inter-model difference 571 of the CMIP6 models. 572

Besides, we compare the transient climate responses (TCRs) between CMIP5 and CMIP6 574 models. For CMIP5 models^{42,43}, the TCRs of CCSM4, CESM1-CAM5, FGOALS-g2 and 575 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 576 models⁴⁴, the TCRs of CanESM5, GISS-E2-1-G, IPSL-CM6A-LR and MIROC6 are 2.66 K, 577 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 578 mean TCRs reported by previous studies^{43,44} based on 29 CMIP5 models and 34 CMIP6 models, 579 respectively. This result suggests that, for either CMIP5 or CMIP6, the four-model ensemble 580 well represents the transient climate sensitivity of the models of either generation. The Student's 581 t-test result further shows that the difference of TCR between CMIP5 and CMIP6 model means 582 is insignificant at the 95% confidence level, which suggests that there is no statistically 583 significant difference in transient climate sensitivity between the CMIP5 and CMIP6 models used 584 in the current study. 585

586

587 **CanESM5 and associated simulations**

CanESM5 is a fully coupled climate model participating in CMIP6⁴⁹. The atmosphere 588 589 component is the Canadian Atmosphere Model (CanAM5), which employs a spectral dynamical core with a T63 truncation (an approximate 2.8-degree horizontal resolution) and a hybrid 590 sigma-pressure coordinate with 49 vertical layers up to about 1 hPa. The land component 591 incorporates the Canadian Land Surface Scheme (CLASS) and the Canadian Terrestrial 592 Ecosystem Model (CTEM). The ocean component is a modified version of the Nucleus for 593 European Modelling of the Ocean model (NEMO), which includes ocean biogeochemistry 594 represented by the Canadian Model of Ocean Carbon (CMOC) and employs a ~1-degree 595

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

598

A 25-member historical simulation labeled as perturbed physics member 1 ("p1") has been 599 performed with CanESM5 during 1850-2014. Individual ensemble members are initialized at 600 different years from preindustrial control run and perturbed by the conservative remapping 601 wind-stress fields. We use these 25 ensembles of CanESM5 historical simulation as they share 602 the same perturbation scheme ("p1") with CanESM5 ozone simulations. We compare the trend 603 of zonal mean temperature in the ensemble mean of the CanESM5 historical simulation with that 604 in the IAP data during 1955-2000 and find that the CanESM5 historical simulation is able to well 605 capture the observed warming tongue (>0.03 K/decade) in the upper 1000 m between 40°S and 606 50° S (Extended Data Fig. 8). This result demonstrates the model fidelity in simulating the 607 Southern Ocean temperature response to external climate forcings. 608

609

Besides the 10-ensemble stratospheric ozone only experiment as in line with several other 610 CMIP6 models, CanESM5 provides a 10-ensemble member historical total ozone-only 611 612 experiment in which the model prescribes the monthly mean three-dimensional ozone concentrations from the historical simulation through the depth of the atmosphere. This total 613 ozone-only experiment is consistent with the CMIP5 ozone single-forcing (stratospheric and 614 tropospheric ozone) experiments. We adopt the simulations of this pair of ozone experiments to 615 isolate and quantify the effects of stratospheric and tropospheric ozone on Southern Ocean 616 interior warming. 617

619 The spiciness and heave decomposition

The spiciness and heave decomposition follows previous studies^{35,36}. For changes in potential temperature (θ) and salinity (S) at depth z, i.e., $\theta'|_z$ and $S'|_z$, they can be decomposed as:

$$\theta'|_{z} \cong \theta'|_{n} + N'\theta_{z}$$
(1)

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

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

628

620

621

629 **The OHC calculation**

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

$$0HC = \rho_0 C_p \int_{z_1}^{z_2} \theta dz \quad (3)$$

where ρ_0 denotes sea water density and C_p denotes the specific heat capacity of sea water.

633

634 The statistical significance test

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

- statistical significances of trends of CMIP5 and CMIP6 MMMs and CanESM5 ensemble means
- based on the Mann-Kendall trend significance test.
- 644

645 **Data availability**

- All the raw CMIP5 model simulation data are publically available at
- 647 <u>https://esgf-node.llnl.gov/search/cmip5/</u>
- All the raw CMIP6 model simulation data are publically available at
- 649 <u>https://esgf-node.llnl.gov/projects/cmip6/</u>
- ⁶⁵⁰ The IAP observation data are publically available at
- 651 <u>http://www.ocean.iap.ac.cn/</u>
- 652

653 Code availability

- ⁶⁵⁴ Figures are generated via the NCAR Command Language (NCL, Version 6.5.0) [Software].
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- The codes and processed variables to generate Figures 1-6 are available at $Zenodo^{50}$.

657

658 Methods References

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