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14        **Challenges for the recovery of the ozone layer**

15    Xuekun Fang<sup>1\*</sup>, John A. Pyle<sup>2,3</sup>, Martyn P. Chipperfield<sup>4,5</sup>, John S. Daniel<sup>6</sup>, Sunyoung Park<sup>7</sup>,  
16    Ronald G. Prinn<sup>1</sup>

17    <sup>1</sup>Center for Global Change Science, Massachusetts Institute of Technology, Cambridge, Massachusetts,  
18    USA

19    <sup>2</sup>Department of Chemistry, University of Cambridge, Cambridge CB2 1EW, UK.

20    <sup>3</sup>National Centre for Atmospheric Science, UK

21    <sup>4</sup>School of Earth and Environment, University of Leeds, Leeds LS2 9JT, UK.

22    <sup>5</sup>National Centre for Earth Observation, University of Leeds, Leeds LS2 9JT, UK

23    <sup>6</sup>Chemical Sciences Division, Earth System Research Laboratory, National Oceanic and Atmospheric  
24    Administration, Boulder, CO, USA

25    <sup>7</sup>Department of Oceanography, Kyungpook National University, Daegu, Republic of Korea

## 26 **Abstract**

27 [From the Editor---FIRST PARAGRAPH: We have already edited your first paragraph for  
28 clarity and to match our style. The revised first paragraph is included at the end of this email.  
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30 and include the revised first paragraph in your revised manuscript]  
31 [< 200 words]

32 The recovery of stratospheric ozone from past depletion is underway, due to the 1987 Montreal  
33 Protocol and its subsequent amendments, which have been effective in phasing out the  
34 production and consumption of the major ozone-depleting substances (ODSs). However, the rate  
35 at which this recovery will continue faces a number of uncertainties. Here, we present our  
36 insights on anthropogenic challenges for future ozone recovery, based on recent findings and  
37 updated observational data on atmospheric ODSs. These challenges partly relate to controlled  
38 ODSs, e.g., unexpected emissions of CFC-11 ( $\text{CCl}_3\text{F}$ ) and slower-than-expected declines in  
39 atmospheric  $\text{CCl}_4$ . Further challenges include emissions of uncontrolled short-lived ODSs (e.g.,  
40  $\text{CH}_2\text{Cl}_2$  and  $\text{CHCl}_3$ ), which observations show have been increasing in the atmosphere through  
41 2017, potential emission increases in natural ODSs (e.g.,  $\text{CH}_3\text{Cl}$  and  $\text{CH}_3\text{Br}$ ) induced by climate  
42 change, changes in atmospheric concentrations of greenhouse gases  $\text{N}_2\text{O}$  and  $\text{CH}_4$ , and  
43 stratospheric geoengineering. We discuss the implications of these challenges for future policy  
44 and science, and suggest how they can be addressed to ensure effective monitoring of the success  
45 of the Protocol, e.g., expanding geographic coverage of atmospheric observations on ODSs,  
46 enhancing the ability of ODS source attribution modeling, and improving understanding of the  
47 interactions between climate change and ozone recovery.

## 48 Main

49 [~500 words]

50 The stratospheric ozone layer (approximately 15 to 35 kilometers above Earth's surface) protects  
51 life on Earth by preventing harmful incoming solar ultraviolet radiation that can cause skin  
52 cancers, damage to ecosystems, and other harmful effects, from reaching the surface. The  
53 Montreal Protocol was signed in 1987, and subsequently strengthened through amendments and  
54 adjustments (see Figure 1a), to minimize the depletion of the ozone layer by phasing out the  
55 anthropogenic production and consumption of ozone-depleting substances (ODSs), such as  
56 chlorofluorocarbons (CFCs), bromochlorofluorocarbons (halons), and hydrochlorofluorocarbons  
57 (HCFCs)<sup>1, 2</sup>. The total global emissions of ODSs have been declining since the late 1980s (Figure  
58 1b), and the observed atmospheric abundances of most ODSs are declining after peaking in the  
59 1990s and 2000s<sup>3, 4</sup>. The emergence of ozone recovery following the phase-out of ODSs under  
60 the Montreal Protocol has been demonstrated, particularly in the Antarctic and in the upper  
61 stratosphere, after accounting for atmospheric circulation, temperature, and volcanic factors<sup>5, 6, 7</sup>.  
62 Outside of the tropics, it is projected that total column ozone will return to 1980 historical levels  
63 in the coming decades, e.g., about 2060 for Antarctic spring<sup>8</sup>.

64 While the long-term ozone layer recovery from the ODS phase-out is well underway,  
65 challenges are posed for the exact timing and extent of recovery in the coming decades. Figure 2  
66 is a schematic illustration of ozone recovery, and the return date of ozone to 1980 levels.

67 Unexpected emission increases in several ODSs (e.g., CFC-11 (CCl<sub>3</sub>F)<sup>9</sup>, CFC-13 (CClF<sub>3</sub>)<sup>10</sup>,  
68 CFC-113a (CF<sub>3</sub>CCl<sub>3</sub>)<sup>11</sup>, CFC-114<sup>a, 10</sup>, CFC-115 (CF<sub>3</sub>CClF<sub>2</sub>)<sup>10</sup>, HCFC-133a (CF<sub>3</sub>CH<sub>2</sub>Cl)<sup>11</sup>,  
69 CH<sub>2</sub>Cl<sub>2</sub><sup>12</sup>, and CHCl<sub>3</sub><sup>13</sup>) have been uncovered by recent studies and could represent an important

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<sup>a</sup>CFC-114 represents the combination of CFC-114 (CClF<sub>2</sub>CClF<sub>2</sub>) and its isomer CFC-114a (CCl<sub>2</sub>FCF<sub>3</sub>).

70 challenge for future ozone layer recovery. For example, a delay of nearly 30 years has been  
71 projected if the atmospheric abundance of a  $\text{CH}_2\text{Cl}_2$  follows a “continued growth” scenario<sup>12</sup>,  
72 demonstrating the possible damage to the ozone layer of continued emission of chlorinated  
73 ODSs (for more discussion see below). Other important challenges include the increases in ODS  
74 emissions from natural sources induced by climate change or future potential applications of  
75 geoengineering<sup>8, 14</sup>. Future ozone increases might also be challenged in the short term by  
76 sporadic volcanic eruptions, which can cause transient ozone depletion over periods of 3-5 years.  
77 Such eruptions are part of natural variability and would not alter long-term ozone recovery<sup>14</sup>.

78 Here, we present our insights on anthropogenic challenges for future ozone recovery (Figure  
79 3), based on recent findings and updated observational data on atmospheric ODSs. An often used  
80 metric of ozone recovery is the date when the atmospheric ozone abundance ‘returns’ to some  
81 previous value, usually the abundance in 1980 (see Figure 2). Such a crude model metric does  
82 not make allowance for atmospheric variability, which is usually removed by smoothing and  
83 averaging results from multiple simulations<sup>15</sup>. Furthermore, if recovery is approached slowly, the  
84 difference between the recovery value and the actual ozone value may be very small, with the  
85 values not statistically different from each other, but recovery as defined will be deemed not to  
86 have occurred. We use recovery dates below, but these caveats should be borne in mind.

## 87 **Unexpected emissions in controlled ODSs**

88 [Subheadings – no more than 58 characters]

89 [~600 words]

90 CFC-11, a potent ODS, was widely used primarily as a foam-blowing agent, as a refrigerant and  
91 in a range of other smaller uses. Global production and consumption of CFC-11 was supposed to

92 be fully phased out by 2010 as required by the Montreal Protocol<sup>2</sup>. However, atmospheric mole  
93 fractions of CFC-11 show that the rate of decline in 2014-2016 was only two-thirds as fast as in  
94 2002-2012<sup>9</sup>. Subsequent modeling shows that there has been an unexpected global CFC-11  
95 emission increase ( $13 \pm 5$  kilotonnes per year ( $25 \pm 13\%$ )) over 2014-2016 relative to 2002-2012,  
96 indicating that new production had occurred, which had not been reported to the United Nations  
97 Environment Programme (UNEP)<sup>9</sup>. Although the country or countries involved were not  
98 pinpointed, it was suggested that new production of CFC-11 in East Asia contributed<sup>9</sup>. A recent  
99 study using atmospheric measurements and inverse modeling has traced the significant CFC-11  
100 emission increase to eastern China, explaining a substantial fraction (approximately 40–60% or  
101 more) of the global emission rise<sup>16</sup>. Emissions of some other controlled ODSs, with much lower  
102 atmospheric abundances, have also increased. For example, compared to 2008–2012, for the  
103 period 2012–2016, atmospheric mole fractions of CFC-13 continued to increase, CFC-114 and  
104 CFC-114a declined more slowly, and CFC-115 reversed from near-zero change to positive  
105 growth, which is not expected given their phase-out for emissive uses under the Montreal  
106 Protocol<sup>4, 10</sup>. CFC-113a mole fractions and emissions also increased much more rapidly from  
107 2010 to mid-2012 than 1980-2010<sup>11</sup>. However, the primary causes of these CFC emission  
108 increases are not clear.

109 These unexpected emissions in controlled ODSs pose a potential challenge for timely future  
110 ozone layer recovery, depending on their magnitude and, in turn, on what mitigating action is  
111 taken. Assuming global CFC-11 emissions continue at their average level of 67 kilotonnes yr<sup>-1</sup> in  
112 2002–2016, the return of mid-latitude and polar equivalent effective stratospheric chlorine  
113 (EESC, a measure of stratospheric chlorine and bromine levels which determine ozone layer  
114 recovery) to its 1980 values could be delayed by about 7 and 20 years, respectively, compared to

115 the return date expected when no unreported production is considered<sup>17</sup>. Moreover, unregulated  
116 emissions of other ODSs might also occur in the coming decades. For example, HCFCs were  
117 widely used as replacements for CFCs in the 1990s and 2000s, but are now being phased out  
118 gradually (e.g., production and consumption frozen in 2013 and complete phase-out by 2040 in  
119 developing countries)<sup>2</sup>. Beginning in the middle of the 2020s in developing countries (e.g., 2024  
120 for China and 2028 for India) and in 2019 and 2020 in developed countries, production and  
121 consumption of hydrofluorocarbons (HFCs), which are being widely used to replace HCFCs,  
122 will be controlled and gradually phased out by the Montreal Protocol. This phase-out period is a  
123 time when increased scientific vigilance is warranted to ensure production of HCFCs and CFCs  
124 remain in compliance with the Montreal Protocol and does not increase in response to the HFC  
125 controls.

126 Carbon tetrachloride (CCl<sub>4</sub>) is another important ODS<sup>4</sup>, whose production for emissive uses  
127 was banned from 2010 onwards<sup>2</sup>. Atmospheric observations show that CCl<sub>4</sub> mole fractions have  
128 been declining at a lower rate than expected and that inferred emissions (~ 35 kilotonnes yr<sup>-1</sup>) are  
129 much greater than those derived from feedstock uses (< 4 kilotonnes yr<sup>-1</sup> in 2012) as reported to  
130 UNEP<sup>18</sup>. Some of the discrepancies (total ~ 25 kilotonnes yr<sup>-1</sup>) have been identified as by-  
131 product emissions from chloromethane and perchloroethylene plants and fugitive emissions from  
132 the chlor-alkali process<sup>19</sup>. A recent inverse modeling study found that CCl<sub>4</sub> emissions were  
133 concentrated in eastern China and did not decline between 2009 and 2016, in contrast to the  
134 bottom-up estimates showing significant decreases<sup>20</sup>. If future global CCl<sub>4</sub> emissions do not  
135 decline but remain at the current level, the return of mid-latitude EESC to 1980 levels would be  
136 delayed by almost two years<sup>17</sup>.

### 137 **Emission increases in uncontrolled ODSs**

138 [~450 words]

139 Important uncertainties in the timing of ozone layer recovery also arise from the uncertain impact  
140 of very short-lived substances (VSLs) with atmospheric lifetimes of typically <6 months.

141 Historically, VSLs were thought to have minimal impact on ozone layer depletion because they  
142 largely degrade in the troposphere, but observations show VSLs do reach the stratosphere<sup>21</sup>. For  
143 many of these gases, anthropogenic emissions (e.g., industrial processes) dominate over natural  
144 sources (e.g., ocean and soil)<sup>4</sup>. However, VSLs, such as dichloromethane (CH<sub>2</sub>Cl<sub>2</sub>) and  
145 chloroform (CHCl<sub>3</sub>), are currently not regulated under the Montreal Protocol<sup>2</sup>.

146 Atmospheric measurements show increases in both atmospheric CH<sub>2</sub>Cl<sub>2</sub> mole fractions (e.g.,  
147 from ~30 ppt in 2004 to ~60 ppt in 2015 in the Northern Hemisphere; ppt refers to parts per  
148 trillion as a dry air mole fraction) and in derived global emissions over 2004-2015<sup>12, 21</sup>. Global 3-  
149 D model simulations show that return of Antarctic ozone to 1980 values could be substantially  
150 delayed by 17-31 years under the assumption of “continued growth” through 2050, or by even 5  
151 years if it is assumed they remain at 2015 levels<sup>12</sup>. It should be noted that the “continued growth”  
152 scenario is strictly hypothetical and is not based on a market analysis or future demand. New  
153 observational data from the Advanced Global Atmospheric Gases Experiment (AGAGE)  
154 network<sup>3</sup> show that CH<sub>2</sub>Cl<sub>2</sub> continued to rise up to 2018, at a rate of 1.8 ppt yr<sup>-1</sup> over 2015-2017  
155 (3.0 ppt yr<sup>-1</sup> over 2016-2017) compared to the growth rate of 1.4 ppt yr<sup>-1</sup> over 2004-2014 (see  
156 Figure 4).

157 Another recent study found that atmospheric CHCl<sub>3</sub> mole fractions were relatively stable or  
158 decreasing during 1990-2010, but were followed by a rapid increase through 2015<sup>13</sup>. Using high  
159 frequency measurement data from two AGAGE stations in East Asia, combined with regional  
160 inverse modelling, hot spots of CHCl<sub>3</sub> emissions have been found in eastern China and the total

161 annual emissions derived for eastern China grew by 49 (41–59) kilotonnes between 2010 and  
162 2015, which could explain the entire global emission increases<sup>13</sup>. By comparison with the results  
163 for CH<sub>2</sub>Cl<sub>2</sub> described above, if atmospheric CHCl<sub>3</sub> growth continued at the average rate observed  
164 during 2010-2015, the ozone layer recovery delay could be 4–8 years; alternatively, the delay  
165 could be 0.4 years if no further growth in atmospheric CHCl<sub>3</sub> mole fractions exists beyond  
166 2015<sup>13</sup>. More recent observational data from the AGAGE network<sup>3</sup> show that CHCl<sub>3</sub> mole  
167 fractions continued to grow at a rate of 0.35 ppt yr<sup>-1</sup> over 2015-2017 (0.89 ppt yr<sup>-1</sup> over 2016-  
168 2017), compared to the growth rate of 0.33 ppt yr<sup>-1</sup> over 2010-2014 (see Figure 4).

169 Thus, CH<sub>2</sub>Cl<sub>2</sub> and CHCl<sub>3</sub> are potential threats for future ozone layer recovery if their  
170 atmospheric abundances continue to grow. It is important to ensure continuous measurements of  
171 CH<sub>2</sub>Cl<sub>2</sub> and CHCl<sub>3</sub> to understand their emission sources and to develop models to simulate better  
172 the impacts of these gases on stratospheric ozone loss. Elimination of CH<sub>2</sub>Cl<sub>2</sub> and CHCl<sub>3</sub>  
173 emissions in the future would rapidly limit their impacts on stratospheric ozone, since these  
174 short-lived gases would be cleansed from the atmosphere within a few years.

## 175 **Climate change and ODS emissions from natural sources**

176 [350 words]

177 Ozone layer recovery during this century will be controlled not only by the decline in ODSs but  
178 also, to a large and increasing extent, by the increase in greenhouse gases (GHGs) and the  
179 associated future climate change. There is a strong coupling between climate change and  
180 stratospheric ozone change in both directions. For example, the influence of the Antarctic ozone  
181 hole has led to a range of significant summertime surface climate changes in the Southern  
182 Hemisphere<sup>22</sup>. Studies have also shown that GHG-induced climate change can cause ozone  
183 changes in different regions<sup>8, 14, 23, 24</sup>, e.g., an increase in ozone in the upper stratosphere at all

184 latitudes and a decrease the lower stratospheric ozone in the tropics<sup>8,23</sup>. This is because GHG-  
185 induced climate change impacts both chemical reactions (e.g., upper stratospheric cooling  
186 reduces gas-phase catalytic ozone loss rates and can lead to “super-recovery” of ozone) and  
187 atmospheric circulation (e.g., likely increase in mid-latitude lower stratospheric ozone because  
188 the Brewer-Dobson circulation transporting ozone from the tropics to the extratropics may be  
189 speeding up<sup>8,25</sup>). The wide range of possible future levels of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O limits accurate  
190 projections of the ozone layer recovery<sup>8</sup>.

191 One future challenge for ozone recovery is that human-induced climate change may increase  
192 natural emissions of halogenated ODSs (CH<sub>3</sub>Cl, CH<sub>3</sub>Br, CHBr<sub>3</sub>, etc.) from ocean and land. Past  
193 studies suggest that atmospheric levels of CH<sub>3</sub>Cl from the ocean may rise due to future GHG-  
194 driven warming, with a response of about 90 ppt CH<sub>3</sub>Cl/°C<sup>26</sup>, from current levels of ~550 ppt in  
195 2015<sup>4</sup>. Production and emissions of CH<sub>3</sub>Br from the ocean will be enhanced in a warming  
196 climate<sup>27</sup>. Emissions of CHBr<sub>3</sub> from the ocean are projected to increase by 31% during 2010-  
197 2100 under a scenario of high greenhouse gas emissions<sup>28</sup>. Models project that Antarctic ozone  
198 recovery will be delayed by ~20 years if atmospheric concentrations of natural ODSs increase at  
199 ~0.2%/yr in a future climate warming<sup>29</sup>. Mitigating climate warming would mitigate the risk of  
200 increases in ODS emissions from natural sources.

## 201 **N<sub>2</sub>O and CH<sub>4</sub> included in climate agreements**

202 [**~400 words**]

203 N<sub>2</sub>O is a powerful greenhouse gas included in climate agreements, e.g., the 2015 Paris  
204 Agreement, which aims to keep global temperature rise this century to less than 2°C above pre-  
205 industrial levels. Anthropogenic sources, which include agriculture, stationary and mobile  
206 combustion, nitric and adipic acid production, biomass burning, and wastewater treatment,

207 contribute approximately one-third of total N<sub>2</sub>O emissions, while natural sources (mainly  
208 microbial activity in the oceans and natural soils) comprise the rest<sup>30</sup>. Atmospheric N<sub>2</sub>O mole  
209 fractions have been growing continuously from 319 ppb (parts per billion) in 2005 to 330 ppb in  
210 2017 as observed in the AGAGE network<sup>3</sup> (Figure 4). N<sub>2</sub>O is also an important ODS, and  
211 increases in its atmospheric abundance lead to ozone depletion. Currently, N<sub>2</sub>O provides the  
212 largest contribution among all individual ODS emissions to stratospheric ozone loss<sup>31</sup>. There are  
213 multiple options for mitigating N<sub>2</sub>O in anthropogenic sectors<sup>32</sup>. N<sub>2</sub>O is not included in the  
214 Montreal Protocol although it has been proposed to regulate N<sub>2</sub>O under the ozone regime as  
215 well<sup>32</sup>. The return of globally averaged total column ozone to 1980 levels will be more than 5  
216 years later compared with the scenario in which no anthropogenic N<sub>2</sub>O emissions occur after  
217 2020<sup>17</sup>. Therefore, atmospheric N<sub>2</sub>O levels will exert an important influence on future ozone  
218 layer recovery.

219 CH<sub>4</sub>, with atmospheric mole fractions of ~1840 ppb in 2016<sup>4</sup>, is also a powerful greenhouse  
220 gas and is included in climate agreements. Its future growth rate is uncertain and, in consequence,  
221 so is its potential future impact on climate, which could be very large. Anthropogenic methane is  
222 emitted from industry, agriculture, and waste management activities. CH<sub>4</sub> also impacts ozone by:  
223 (1) increasing atmospheric odd hydrogen species and H<sub>2</sub>O, which decrease ozone; (2) cooling of  
224 the stratosphere which decreases ozone loss; (3) deactivating chlorine which decreases ozone  
225 loss; and (4) increasing the NO<sub>x</sub>-ozone production in the troposphere. Studies show that the  
226 return of global average total ozone relative to 1980 level would be accelerated by ~15 years if  
227 CH<sub>4</sub> follows the high-emission scenario of Representative Concentration Pathways (RCPs; RCP-  
228 8.5<sup>33</sup>; 3750 ppb in 2100)<sup>17</sup>. However, the return of global average total ozone relative to 1980  
229 level would be delayed by ~35 years if CH<sub>4</sub> follows the low-emission RCP scenario (RCP-2.6<sup>33</sup>;

230 1254 ppb in 2100)<sup>17</sup>. Thus, the impact of CH<sub>4</sub> on future ozone layer recovery depends  
231 significantly on the rather uncertain future CH<sub>4</sub> emission trajectory.

## 232 **Stratospheric geoengineering**

233 [**~150 words**]

234 To tackle climate change, geoengineering projects like injecting sulfate aerosol into the  
235 stratosphere for solar-radiation management have been proposed<sup>e.g., 34</sup>. Such injections might  
236 have some appeal because they are the only known way at present to reduce temperatures  
237 quickly and effectively, but they would also entail new risks, including likely depletion of  
238 stratospheric ozone. Thus, stratospheric geoengineering may become another challenge for future  
239 ozone recovery. Specifically, modeling studies show that an injection of sulfate aerosol large  
240 enough to compensate for surface warming caused by the doubling of atmospheric CO<sub>2</sub> would  
241 cause the Antarctic ozone hole to persist into the next century<sup>35</sup>. It has been proposed that the  
242 injection of alkaline calcite (CaCO<sub>3</sub>) aerosol particles rather than acidic sulfate aerosol would  
243 enable stratospheric geoengineering while reducing or even reversing ozone depletion<sup>36</sup>.  
244 However, more studies are needed to investigate further the effects of using calcite aerosols and  
245 other alternatives.

## 246 **Implications for future policy and science**

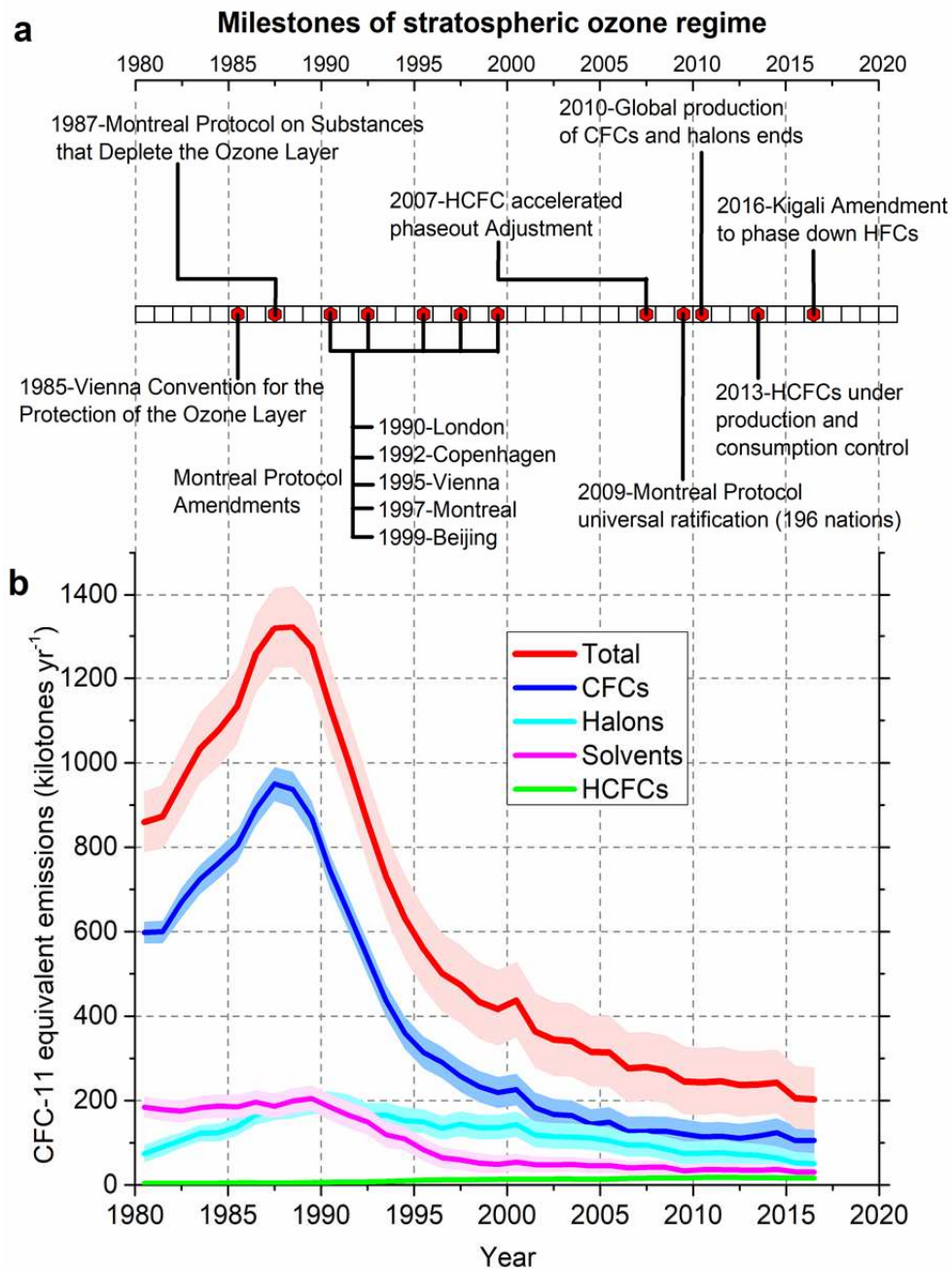
247 [**~300 words**]

248 To mitigate the threats to future ozone recovery, it is first crucial to ensure that the 32-year-old  
249 Montreal Protocol and its amendments are implemented effectively in firmly controlling ODSs.  
250 Effective implementation requires that the current issue of unexpected emissions of CFC-11, and  
251 other minor CFCs be promptly addressed. Going forward, since other ODSs in addition to those

252 addressed here may show unexpected emission increases, careful monitoring of the phase-out  
253 activities of all ODSs is essential. Given the recent increases in atmospheric  $\text{CH}_2\text{Cl}_2$  and  $\text{CHCl}_3$   
254 concentrations, emissions and their potential threat to future ozone recovery, it is important to  
255 understand their sources prior to any discussion about mitigation. Threats from  $\text{N}_2\text{O}$  to ozone  
256 recovery, and to climate, could be minimized if anthropogenic  $\text{N}_2\text{O}$  becomes controlled  
257 effectively. Efforts to mitigate future climate warming may also help avoid any significant  
258 increases in natural emissions of some ODSs. The research community must be vigilant in  
259 highlighting any geoengineering options which could represent a potential threat to the ozone  
260 layer.

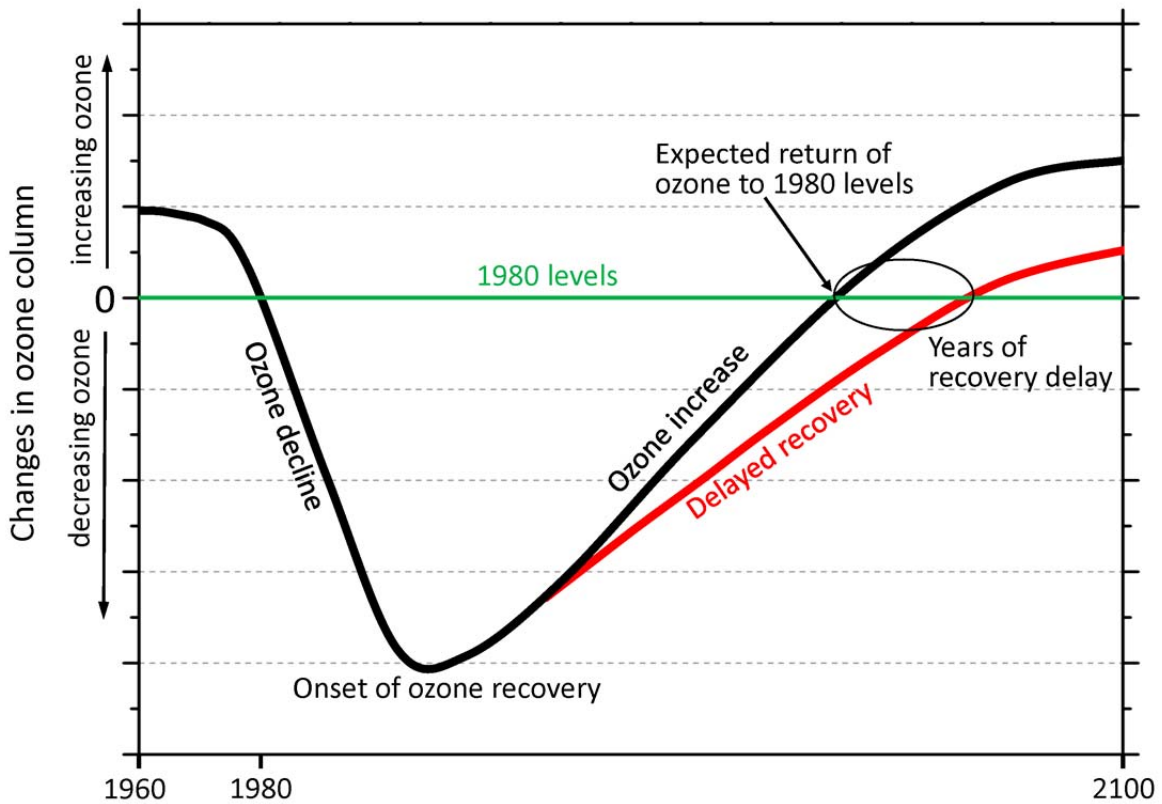
261 With all these potential future challenges for ozone recovery, it is imperative that accurate  
262 monitoring of atmospheric ozone, ODSs, GHGs, and climate change continue throughout the  
263 coming decades. A substantial proportion of the world, e.g., western China, South and Southeast  
264 Asia, and South America, is currently not covered by existing atmospheric ODS measurement  
265 networks<sup>3,4</sup>, so that ODS-relevant information (e.g., emission strengths and spatial distributions)  
266 for those regions is severely lacking. Thus, enhancing the spatial coverage of the monitoring  
267 networks is crucial at global and regional scales. Methods need to be improved for pinpointing  
268 emission locations by inverse modeling, attributing emissions among anthropogenic and natural  
269 sources, and understanding the interplay between climate change and ozone layer recovery. In  
270 the future, alarms informed by regional inverse modeling studies could precede a global alarm,  
271 since regional atmospheric measurements respond much faster to unexpected emission increases  
272 than remote global measurements. Finally, our ability to model the future interactions between  
273 climate change and ozone recovery in coupled Earth system models needs to be improved  
274 through coordinated model development and evaluation.



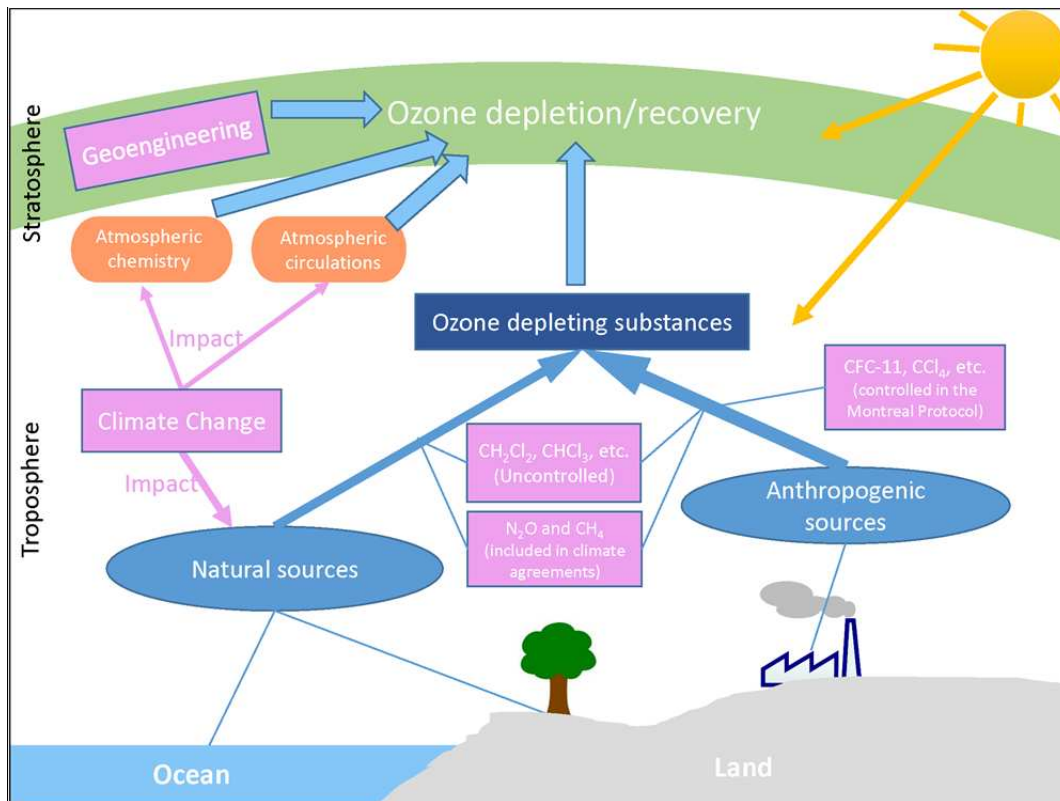


276

277 **Figure 1. (a) Milestones of the stratospheric ozone control regime and (b) CFC-11**  
 278 **equivalent emissions during 1980-2016.** Emissions are derived from the WMO 2018 report<sup>4</sup>  
 279 using atmospheric measurement data, converted to CFC-11-equivalents using their Ozone  
 280 Depletion Potential (ODP) values<sup>8</sup>. Species are grouped into CFCs, halons, solvents (CCl<sub>4</sub> and  
 281 CH<sub>3</sub>CCl<sub>3</sub>), and HCFCs. Shading indicates the 1-sigma uncertainty.

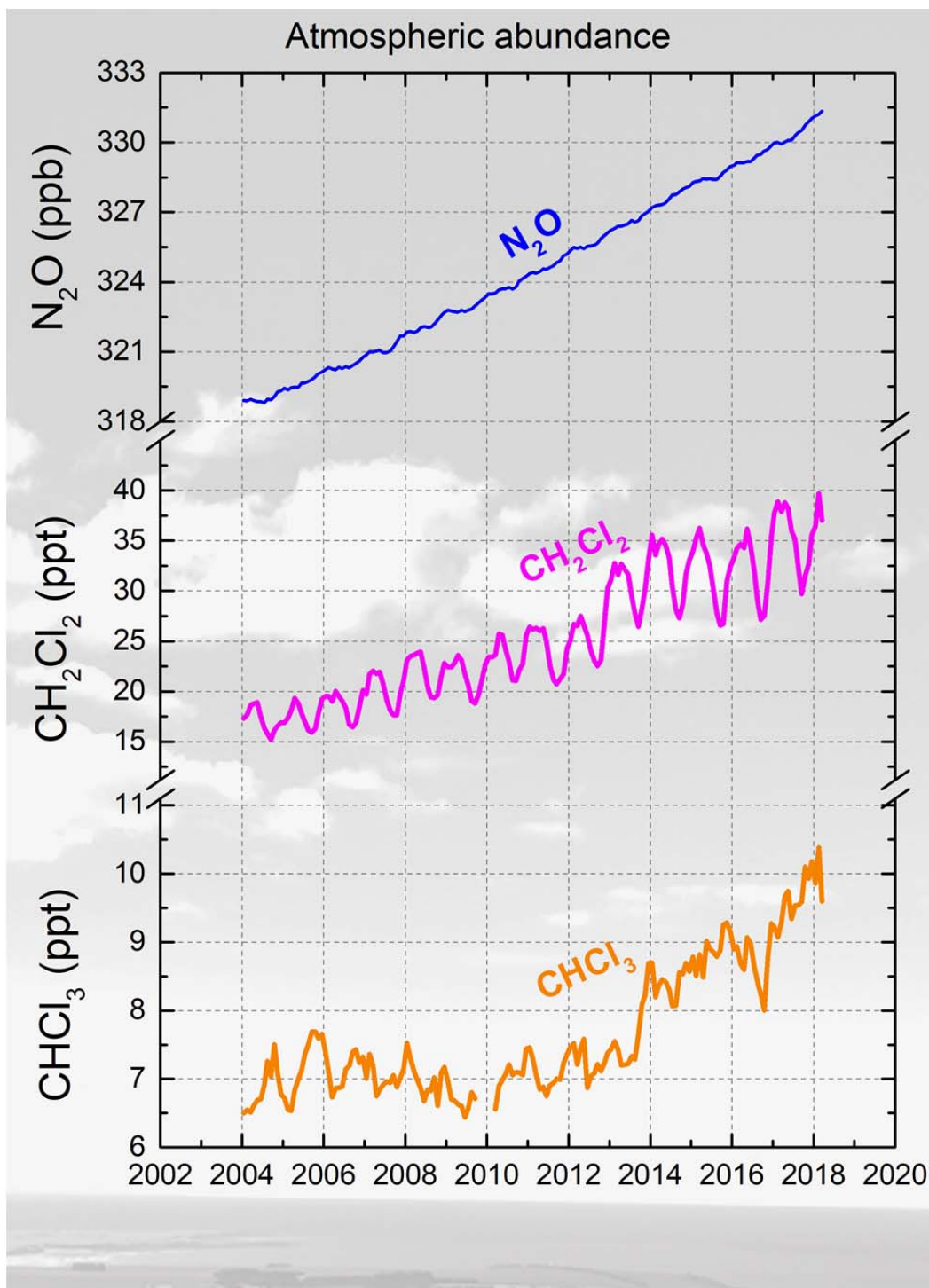


282  
 283 **Figure 2. Schematic illustration of ozone recovery.** The solid black line represents ozone  
 284 depletion from 1960, turnround and then ozone recovery towards 2100. The zero line is drawn at  
 285 the 1980 ozone value. The red line illustrates a future scenario with larger emissions of ODSs  
 286 which causes a delay in ozone recovery and later return to the 1980 value. Please refer to Figures  
 287 6-4 and 6-5 in WMO (2018)<sup>17</sup> as examples for quantitative historical ozone trajectory and  
 288 projected trajectories from various scenarios.



289

290 **Figure 3. Schematic illustration of potential challenges for ozone recovery.** Ozone-depleting  
 291 substances are released from both anthropogenic and natural sources. Some controlled  
 292 substances (e.g., CFC-11 and CCl<sub>4</sub>) are only from anthropogenic sources. Some substances (e.g.,  
 293 CH<sub>2</sub>Cl<sub>2</sub> and N<sub>2</sub>O) are from both anthropogenic and natural sources. Climate change impacts  
 294 atmospheric chemistry, circulation, and emissions of ODSs from natural sources. The purple  
 295 boxes show the five potential challenges for the recovery of the ozone layer.



296  
 297 **Figure 4. Continuous growth of global atmospheric abundances of  $N_2O$  (ppb),  $CH_2Cl_2$  (ppt),**  
 298 **and  $CHCl_3$  (ppt) from 2004 to 2017 derived from the AGAGE global observation network<sup>3</sup>.**  
 299 The data were accessed on March 11, 2019 via <http://agage.mit.edu/data/>. The term of ‘ppt’  
 300 refers to parts per trillion; ‘ppb’ refers to parts per billion.

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## 427 **Author contributions**

428 X.F. and R.G.P. were responsible for the overall project design. All authors wrote up the paper  
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## 430 **Competing financial interests**

431 The authors declare no competing financial interests.

## 432 **Corresponding author**

433 \*e-mail: [fangxk@mit.edu](mailto:fangxk@mit.edu) (X.F.);

434 **ORCID**

435 Xuekun Fang: 0000-0002-7055-0644

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