

Large crop production losses induced by global ozone stress based on interval evaluation

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22 **Abstract**

23 Global crop yield loss due to ground-level ozone (O_3) concentrations is a major challenge to food
24 security, but a dose-response association is not easy to quantify. Here, we propose using a new metric,
25 O_3 sensitivity of crop yield (Y_o), to estimate yield loss under different O_3 time intervals using four
26 observational databases. The Y_o metric shows a non-linear parabola with elevated atmospheric O_3 for
27 wheat, maize, rice, soybean, and assorted vegetables. Spatial heterogeneity of yield loss varies as a
28 function of crop type and O_3 intervals. Estimates of yield loss from ozone suggest recent losses
29 (2017-2019) may reach as high as 537 million tonnes, with a significant proportion coming with lower
30 (30-40 ppb) exposure (325 million tonnes). Our results suggest that previous research, which only
31 included higher (>40 ppb ozone), may have had grossly underestimated the negative effect of
32 atmospheric O_3 on crop production. Suppose these results are endemic to global crop production. In
33 that case, additional research will be necessary to reassess ozone sensitivity and dose-responses, both
34 spatially and temporally, to determine future air pollution impacts.

35

36 **Introduction**

37 Air pollution, including atmospheric (or ground-level) concentrations of ozone (O₃), can significantly
38 damage plant growth and biomass accumulation in terrestrial ecosystems^{1, 2, 3}. Atmospheric O₃ enters
39 the plant body through leaf stomata and stimulates a series of biochemical reactions, which destroy the
40 cell structure and initiate physiological and metabolic disorders^{4, 5}. Such adverse reactions can decrease
41 stomatal conductance and net photosynthetic rate and further result in losses of biomass and yield.
42 Since the late 1800s, atmospheric O₃ has risen from approximately 10 ppb to 50 ppb today and will
43 increase by 40-60% until 2100⁶.

44 For agroecosystem, an accurate assessment of how elevated atmospheric O₃ affects crop productivity,
45 especially crop yield, is crucial for global food security^{3, 7}. Most researchers have focused on the total
46 loss of crop yield caused by atmospheric O₃ while turning out to have considerable variation and high
47 uncertainty^{8, 9, 10}. The value of 40 ppb O₃ is generally considered a threshold. Beyond 40 ppb,
48 atmospheric O₃ could cause significant crop yield loss^{11, 12}. Nonetheless, atmospheric O₃ may be
49 unlikely to fall directly below the crop threshold by taking measures in the short term¹. The knowledge
50 gap on crop yield change under different atmospheric O₃ intervals swamps the evaluation and
51 prediction of future atmospheric O₃ pollution on crop productivity. It thus is not conducive to the
52 establishment of effective mitigation strategies and policies.

53 As atmospheric O₃ rises, crop O₃ absorption does not increase proportionally because of stomatal
54 resistance⁴. Crops have specific adaptability and resistance to atmospheric O₃ damage through their
55 natural defenses (e.g., antioxidants, detoxification, and nocturnal remediation capabilities) and the
56 specific triggering responses¹³. These differences in ozone sensitivity may lead to significant variation
57 of yield responses relative to low and high O₃ concentrations and for a time of exposure to those
58 concentrations. However, this phenomenon to date is poorly noticed and understood when evaluating
59 O₃ impact^{4, 9, 14}. Here we propose a new approach with the O₃ sensitivity of crop yield (Y_o) by
60 estimating crop yield loss rate (%) per increase in O₃ concentration of 1 ppb above ambient levels for
61 one hour. Although individual experiments have reported average values of Y_o based on the
62 relationship between O₃ dose (hour mean of O₃ or accumulated O₃ over a threshold concentration) and
63 crop yield loss rate^{3, 14, 15}, explicit quantification of the values under smaller O₃ intervals is still scant
64 and remains a major challenge in formulating dose-responses for crop yield assessment. Due to the lack
65 of observational O₃ data, part of this challenge has been met using atmospheric models to predict

66 regional and global O₃-induced crop yield losses^{3, 8, 16}. However, with the establishment of numerous
67 O₃ monitoring stations in recent years, more accurate global real-time O₃ concentration data could be
68 obtained (Supplementary Fig. 5). An interval evaluation of Y_o based on observational real-time O₃ data
69 is crucial to consistent predictions of crop yield response to changes in atmospheric O₃.

70 Based on an evaluation of over 900 O₃ fumigation experiments, we present a detailed analysis to
71 provide a dose-response of O₃ exposure on yield of major crops (wheat, maize, rice, and soybean)
72 based on seven interval evaluations. 7246 hourly atmospheric O₃ monitoring stations, distribution of
73 crop cultivation, and crop production observational databases (see “Methods” section; Supplementary
74 Data). We first calculate the Y_o under different crop types and O₃ intervals (30-40, 40-50, 50-60, 60-70,
75 70-80, 80-90, and >90 ppb). Then we map the global distribution of yield loss rates for major crops
76 under 7 O₃ intervals. In addition, we provide a metric for the dose-response of O₃ for yield loss on a
77 regional crop basis. Finally, we discuss direct and indirect pathways by which atmospheric O₃ affects
78 crop yield through photosynthetic and agronomic indexes using a structural equation model¹⁷. In
79 general, the sizes of Y_o show a non-linear parabola with atmospheric O₃ elevation. Larger crop
80 production loss caused by atmospheric O₃ is estimated compared to the previously suggested value.

81 **Results and discussion**

82 **The O₃ sensitivity of crop yield.** Our synthesis quantified the Y_o of primary crop yield (wheat, maize,
83 rice, soybean, and vegetable) under 7 O₃ intervals at the global scale by involving 960 O₃ fumigation
84 experiments (Fig. 1). Previous studies have established different O₃ indicators to estimate the impact of
85 atmospheric O₃ on crop yield^{3, 12, 18, 19}. However, those O₃ indicators are mainly divided into two
86 categories: O₃ dose indicators (M₇ or M₁₂: 7-hr or 12-hr mean O₃; AOT₄₀, or SUM₆₀, hourly average O₃
87 concentration higher than 40 or 60 ppb) and O₃ stomatal absorption flux indicator (POD_Y, hourly O₃
88 stomatal flux higher than the cumulative flux of Y nmol m⁻² s⁻¹)^{3, 20}. The O₃ dose indicators unify the
89 atmospheric O₃ with the crop yield, ignoring the crop's resilience and adaptation to different
90 atmospheric O₃. The O₃ stomatal absorption flux indicator considers the influence of biological and
91 environmental factors on the stomatal O₃ absorption of plants; however, it is difficult to obtain the
92 actual value from observational data, especially at the global or regional level scale³. Based on a large
93 number of experimental data, the Y_o under different O₃ intervals can overcome the above defects better
94 to understand crop yield response to elevated atmospheric O₃.

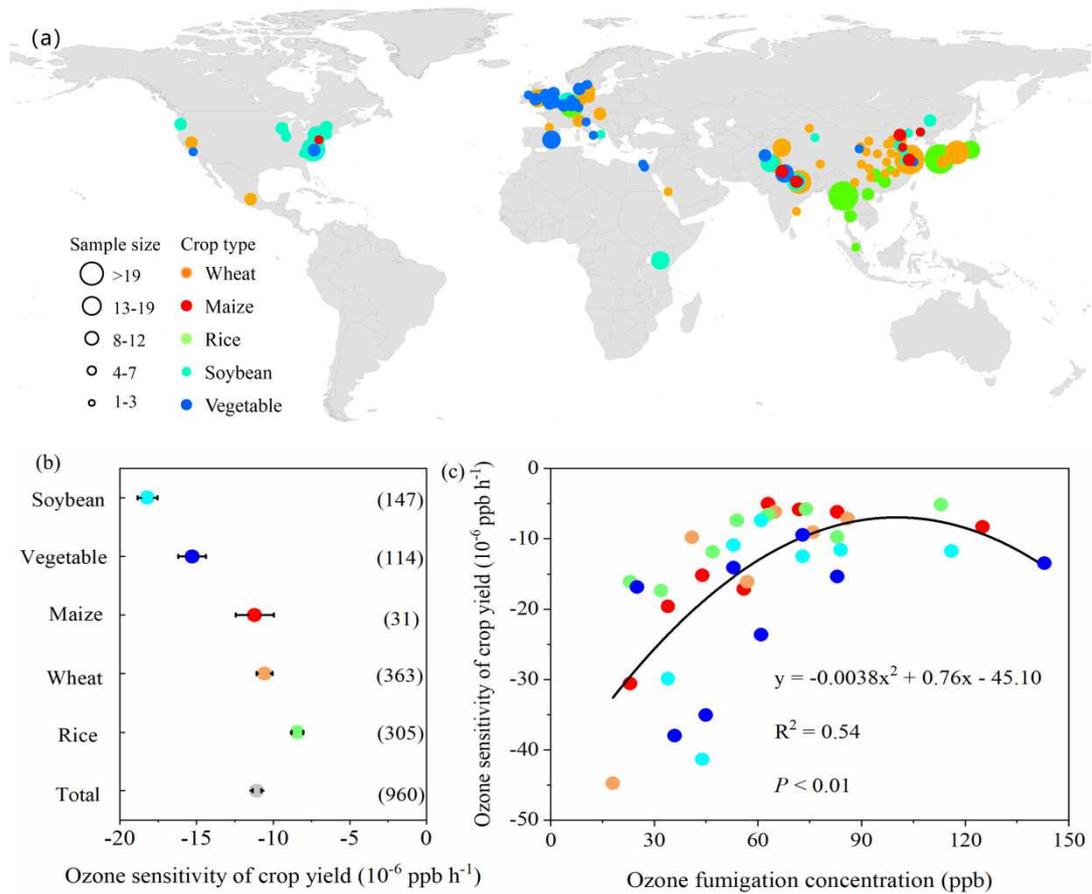
95 At the global scale, the average of Y_o was -11.1×10⁻⁶ ppb h⁻¹, with a low 95% confidence of -10.7 to

96 -11.4×10^{-6} ppb h⁻¹ ($N = 960$, Fig. 1b). The Y_o among crop types was significantly different with an
97 average of -18.2, -15.3, -11.2, -10.6, and -8.4×10^{-6} ppb h⁻¹ for soybean, vegetables, wheat, maize, and
98 rice, respectively (Fig. 1b). These differences may be due to different responses of crops in
99 photosynthesis with leaf stomatal and non-stomatal limitations under O₃ stress. For example, leaf
100 stomata can regulate O₃ absorbed doses, thus affecting crop sensitivity to O₃⁴. The decrease in stomatal
101 conductance under O₃ stress is considered the major reason for reducing the photosynthetic rate.
102 Generally, the stomatal conductance of soybean is more significant, and its response to O₃ was
103 significantly weaker than wheat and rice (Supplementary Fig. 10)^{9, 21, 22}. In terms of non-stomatal
104 factors, the mesophyll of dicotyledonous (e.g., soybean and most vegetables) is differentiated into
105 palisade tissue and spongy tissue compared with monocotyledonous (e.g., wheat, maize, and rice).
106 Palisade tissue is close to the upper epidermis and contains more chlorophyll. High O₃ can firstly
107 damage the palisade tissue and then cause cytoplasmic wall separation and cell content dispersion,
108 inhibiting crop growth and yield formation²³. Therefore, soybean and vegetable were more sensitive
109 to elevated O₃ than wheat, maize, and rice.

110 There were significant differences in the Y_o for the same crop type among O₃ fumigation
111 concentrations (Fig. 1c, Supplementary Fig. 6-9). Previous researches mainly focused on the overall
112 size and did not explicitly distinguish the different effects of O₃ levels^{11, 24}. Many studies indirectly
113 showed that different O₃ fumigation concentrations showed diverse impacts on crop growth and yield
114 formation based on data integration (Meta-analysis)^{21, 22}. In this study, we also demonstrate a
115 non-linear parabola between the Y_o and O₃ fumigation concentration (Fig. 1c). Our result provides
116 evidence of biological evolutionism to O₃ stress. This viewpoint can be supported by the crop's
117 photosynthetic physiologies, antioxidant system, and other multiple pathways resulting in adaptation
118 and recovery to O₃ stress^{4, 23}. As O₃ concentration increases, the detrimental effect of unit O₃
119 concentration on photosynthesis decreases gradually⁹. High O₃ stress can minimize stomatal
120 conductance through the plasma membrane, slow anion channel preferential response in guard cells,
121 and even cause stomatal closure, inhibiting leaf photosynthesis and lowering crop productivity⁴.
122 Simultaneously, the activities of superoxide dismutase, catalase, and peroxidase increase rapidly under
123 O₃ stress, which is the first step to defend against reactive oxygen species damage caused by O₃^{4, 25}.
124 Other pathways enhance crop resistance and lead to the parabola relationship between the Y_o and O₃
125 concentration, i.e., high O₃ stress can accelerate crop respiratory metabolisms and promote

126 nutrient absorption by stimulating related enzymes^{4, 26}.

127 Interestingly, when O₃ concentration exceeded 100 ppb, the crop yield loss rate increased with an
128 elevated O₃ concentration of 1 ppb h⁻¹ (Fig. 1c). Long-term high O₃ exposure can reduce stomatal
129 resistance and destroy the antioxidant system²⁶, which would lead to irreversible damage to the crop.
130 Overall, the parabola relationship between the Y₀ and O₃ concentrations is an advanced indicator of
131 how crop yield responds to different O₃ concentrations.



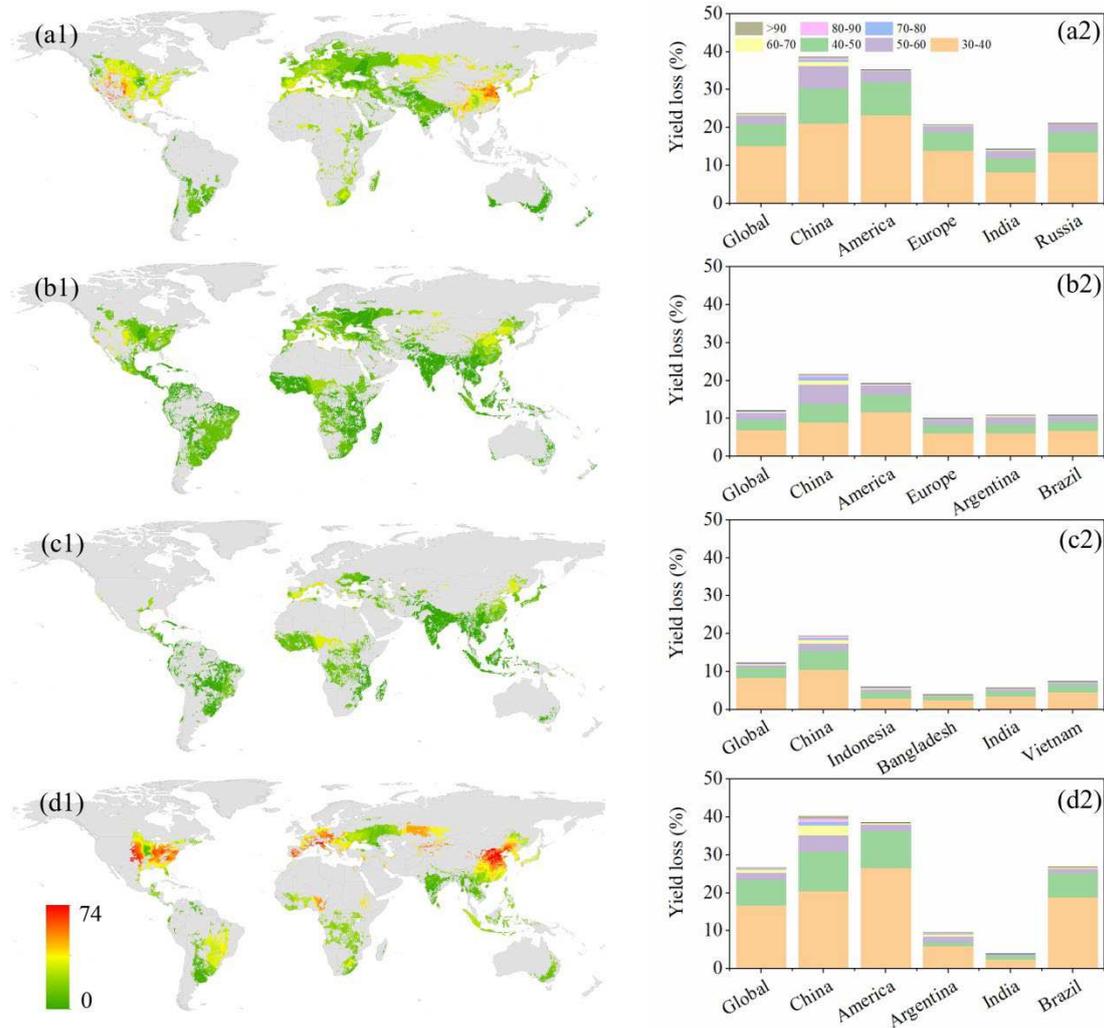
133 **Fig. 1 Ozone sensitivity of crop yield.** a, global distribution of the 960 experiments from 208
134 published papers in which the effect of elevated ozone concentration on crop yield was assessed. The
135 size of black circles represents the sample size ranging from 1 to more than 19. Crop types include
136 wheat (in brown), maize (in red), rice (in green), soybean (in turquoise), and vegetable (in blue). b,
137 response ratio (natural logarithm-transformed ratio of treatment to control) of the ozone sensitivity of
138 crop yield (ppb h⁻¹). Dot and bar represent the mean and range at 95% confidence intervals of ozone
139 sensitivity of crop yield. The value in parentheses represents the sample sizes. c, the relationship
140 between ozone sensitivity of crop yield and ozone fumigation concentration. Each dot represents the
141 average effect size under ozone fumigation intervals with <30, 30-40, 40-50, 50-60, 60-70, 70-80,

142 80-90, >90 ppb for each crop type according to the treatment groups in the database.

143

144 **The magnitude of crop production loss.** To our knowledge, this study is the first to present global
145 distribution maps of major crop yield (wheat, maize, rice, and soybean) loss under 7 O₃ intervals
146 (30-40, 40-50, 50-60, 60-70, 70-80, 80-90, and >90 ppb) based on integrating available
147 observation-based databases. Global annual crop yield loss rate (2017-2019) added up to 23.8, 12.2,
148 12.3, and 26.8% for wheat, maize, rice, and soybean, respectively (Fig. 2, Supplementary Fig. 6-9).
149 Although previous studies have estimated crop yield losses associated with O₃ exposures using a
150 meta-analysis method^{21, 22, 27}, their results are unlikely to be widely applied for two reasons. First, the
151 meta-analysis' findings were calculated using existing literature with a limited sample size (N<150)^{21, 22,}
152 ²⁷. The key factors causing variation in meta-analysis results, as stated in their articles ^{21, 22, 27}, are that
153 they used the different O₃ fumigation concentrations and duration of O₃ fumigation. For example, some
154 experimental fumigations did not last for the entire crop growth period, which would underestimate the
155 O₃ stress. Second, the crop yield loss rate is caused by the accumulation of O₃ concentration. The
156 atmospheric O₃ concentration and duration are different at the global and regional scales^{1, 28}. The
157 meta-analysis findings did not map the rate of crop yield loss caused by atmospheric O₃ in various
158 regions.

159 The U. S. Environmental Protection Agency and European researchers have suggested that
160 atmospheric O₃ concentrations above 60 and 40 ppb would affect local crop yields¹². Currently, most
161 studies estimate the crop yield loss rate caused by atmospheric O₃ based on AOT₄₀^{28, 29}. Our study
162 showed that O₃ concentration over 30 ppb had a significant effect on crop yield than O₃ concentration
163 below 30 ppb (Supplementary Fig. 3). Previous results based on AOT₄₀ are most likely underestimate
164 the impact of O₃ on crop production. Therefore, the crop yield loss rate evaluated with different O₃
165 intervals was robust by using the accurate Y_o and hourly O₃ data from more than 7,000 O₃ monitoring
166 stations in our study (Supplementary Fig. 4). The crop yield loss rate differed significantly among
167 divergent O₃ intervals (Fig. 2, Supplementary Fig. 6-9). This was mainly because the occurrence
168 frequency of low O₃ concentration was much higher than that of high concentration (Supplementary
169 Table 1). It is also affected by the relationship between the Y_o and O₃ concentration (Fig. 1c). Our
170 estimation based on the Y_o can provide a reference to diagnose the response of crop yield to
171 atmospheric O₃ change predicted by other empirical methods.



173 **Fig. 2 Global annual loss rate (%) of crop yield (2017-2019) due to atmospheric ozone**
 174 **concentration.** The maps showed the spatial distribution of cumulative crop yield loss (%) in the case
 175 of ozone concentration above 30 ppb for wheat (a1), maize (b1), rice (c1), and soybean (d1), averaged
 176 for the period 2017-2019. The spatial distribution of crop yield loss in the case of ozone above 30, 40,
 177 50, 60, 70, 80, and 90 ppb for wheat, maize, rice, and soybean were also shown in Supplementary Fig.
 178 6-9, respectively. All data were presented for the 0.0083° grid squares based on global 7246 real-time
 179 ozone monitoring stations (see Supplementary Fig. 5). a2, b2, c2, and d2 indicated crop yield loss of
 180 wheat, maize, rice, and soybean in the case of ozone concentration ranged with 30-40, 40-50, 50-60,
 181 60-70, 70-80, 80-90, and >90 ppb for global and 5 countries or regions (ranked top five in grain
 182 production) averaged value, respectively. The percentage of ozone monitoring stations involved in
 183 calculating the effects of ozone on crop yield at each range of ozone concentration was shown in
 184 Supplementary Table 1.
 185

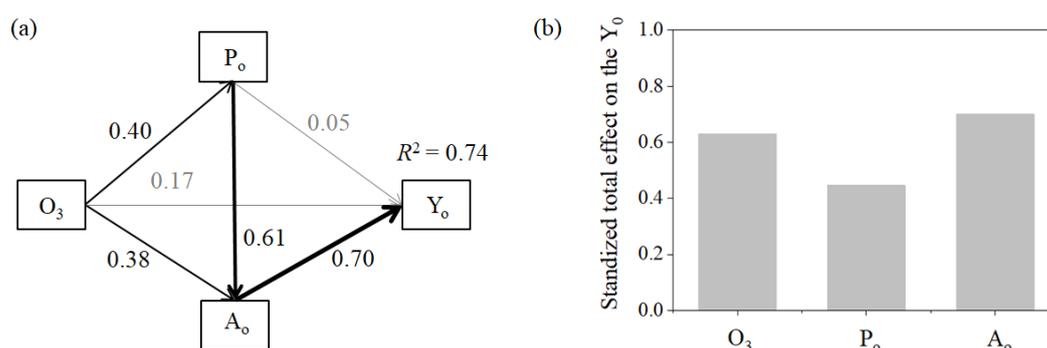
186 Global annual crop production loss (2017-2019) caused by atmospheric O₃ added up to 537 million
187 tonnes, of which 177, 182, 82, and 96 million tonnes for wheat, maize, rice, and soybean, respectively
188 (Table 1). The yield loss in our study (537 million tonnes) was about double that of a recent global
189 study (227 million tonne) that used the European Monitoring and Evaluation Programme model
190 according to global crop production data for 2010-2012³. This difference is mainly because the
191 chemical transport model does not accurately predict hourly O₃ concentration, especially for a
192 multi-year time series of atmospheric O₃ concentrations at a global scale of ^{30, 31}. Atmospheric O₃
193 concentrations have been increasing at an annual rate of 0.5-2.0% for the past few decades³². The data
194 of global hourly O₃ concentration and continued increase in O₃ over time are the main reasons leading
195 to the rise in crop yield loss estimated by our study (537 million tonnes) compared with Mills, Sharps ³
196 (227 million tonnes). Despite that good practices and advanced technologies were adopted in crop
197 cultivation, crop yield in many world regions stagnated in recent years. This might be partly explained
198 by the growing severe O₃ pollution, which damaged the yield formation of the crop. Atmospheric O₃
199 may be unlikely to fall directly below the injury threshold in the short term¹. Our results also indicate
200 that low levels of O₃ have a non-negligible effect on yield loss (Supplementary Figure 3). Hence, O₃
201 mitigation strategies and policies in agriculture are crucial for global food security.

202 **Table 1 Global and five areas (ranked top five in grain production) annual loss amount of wheat, maize, rice, and soybean production (million tonnes) in the case of**
 203 **ozone 30-40 ppb and above 40 ppb. Each country's annual loss rate of crop yield in the case of ozone above 30, 40, 50, 60, 70, 80, and 90 ppb and total yield for**
 204 **wheat, maize, rice, and soybean were shown in Supplementary Table 2-6.**

| Wheat | | | Maize | | | Rice | | | Soybean | | |
|---------|-----------|---------|-----------|-----------|---------|------------|-----------|---------|-----------|-----------|---------|
| Area | 30-40 ppb | >40 ppb | Area | 30-40 ppb | >40 ppb | Area | 30-40 ppb | >40 ppb | Area | 30-40 ppb | >40 ppb |
| Global | 112.44 | 64.94 | Global | 99.51 | 82.76 | Global | 48.08 | 33.49 | Global | 64.87 | 30.93 |
| China | 28.17 | 23.42 | America | 42.09 | 27.44 | China | 22.59 | 18.97 | America | 29.93 | 13.46 |
| Europe | 25.37 | 12.62 | China | 23.40 | 32.77 | India | 6.15 | 3.59 | Brazil | 21.74 | 9.35 |
| America | 11.74 | 5.97 | Europe | 6.86 | 4.39 | Vietnam | 2.00 | 1.23 | China | 3.20 | 3.11 |
| Russia | 10.48 | 5.93 | Brazil | 6.43 | 3.75 | Indonesia | 1.68 | 1.77 | Argentina | 2.99 | 1.75 |
| India | 8.27 | 6.15 | Argentina | 3.08 | 2.39 | Bangladesh | 1.37 | 0.75 | India | 0.31 | 0.18 |
| Other | 28.40 | 10.86 | Other | 17.64 | 12.02 | Other | 14.29 | 7.19 | Other | 6.70 | 3.08 |

205

206 **The mechanisms of crop production loss.** The magnitudes of Y_o varied greatly among experimental
 207 sites, ranging from -44.9×10^{-6} to -4.8×10^{-6} ppb h^{-1} (Fig. 1). Path analysis showed a network of
 208 inter-correlation of atmospheric O_3 concentration, photosynthetic indexes, and agronomic indexes in
 209 determining the Y_o (Fig. 3a), implying that the effect size of Y_o was regulated by multiple factors rather
 210 than a single factor. Agronomic indexes, especially aboveground biomass and grain number per ear, are
 211 the most critical factors directly determining crop yield under elevated O_3 . Photosynthesis indexes play
 212 their roles mainly by affecting agronomic indexes. When atmospheric O_3 particularly enters the crop
 213 body through the stomata, it can stimulate a series of biochemical reactions (photosynthetic rate,
 214 stomatal conductance, and enzyme activity), which further reduce the agronomic indexes leads to crop
 215 yield loss^{4,5}. Breeding new cultivars which have better resistance to O_3 damage is recognized widely^{23,}
 216 ³³. The inter-correlation between atmospheric O_3 concentration, photosynthetic indexes, and agronomic
 217 indexes in determining the Y_o suggested in our study can provide a scientific reference for crop
 218 breeding and O_3 -crop models optimization.



219
 220 **Fig. 3 Influence of ozone (O_3) and ozone sensitivity of photosynthetic (P_o) and agronomic (A_o)**
 221 **indexes on the ozone sensitivity of crop yield (Y_o).** **a**, path analysis results on the direct and indirect
 222 effects of O_3 , P_o , and A_o on the Y_o . Numbers show the path coefficients. Grey path and number indicate
 223 that the effect is insignificant. Arrow width is proportional to the standardized coefficient. The
 224 P_o includes ozone sensitivity of light-saturated rate, ozone sensitivity of stomatal conductance, and
 225 ozone sensitivity of chlorophyll (See supplementary Fig. 11). The A_o has the ozone sensitivity of above
 226 ground biomass, the ozone sensitivity of grain number per ear, and the ozone sensitivity of leaf area. **b**,
 227 the standardized total effect of O_3 , P_o , and A_o on the Y_o .

228
 229 **Limitation.** Although we have rigorously reviewed and synthesized multiple datasets from the
 230 available literature and public data to estimate global crop production losses and underlying potential

231 drivers, there are limitations to the current study. One area of importance is acknowledging
232 intra-specific variation to O₃ among crop cultivars^{3, 14, 34}. In recent years, many crop cultivars have
233 been developed to achieve higher yields and better resilience to O₃. Variation in crop cultivar response
234 will, in turn, provide uncertainties for Y_o. Secondly, we derived crop yield loss rates from observed
235 stations' global real-time atmospheric O₃ concentration data (see “Methods”). However, not all
236 countries or regions have established real-time atmospheric O₃ observatory stations or provided access
237 to the observational data. Consequently, atmospheric O₃ stations in Asia, Europe, and North America
238 are better represented than South America and Australia, with data from other Africa, the Middle East,
239 and Russia being problematic (Supplementary Fig. 5). Therefore, estimates of the crop yield loss rates
240 in Africa and Russia may be less accurate³⁵. Thirdly, although the crop yield loss rates with the 0.0083
241 ×0.0083° grid squares are reported based on MAPSPAM and the atmospheric O₃ database, the global
242 crop production in our study is based on country-specific FAO data and does not provide the exact
243 resolution. Such resolution can result in a mismatch between crop yield loss rates and crop production
244 and may affect the final estimates of food production loss. While we recognize these limitations, our
245 combined databases define and provide accurate values of the Y_o and robust estimate of crop
246 production loss under different atmospheric O₃ intervals than previous studies.

247 In summary, our global synthesis verifies that the Y_o, defined as yield loss rate (%) with an elevated
248 O₃ concentration of 1 ppb h⁻¹, shows a non-linear parabola with atmospheric O₃ increase and
249 significant differences among crop types. This indicator provides a new perspective and method for
250 improving the crop system models to predict yield loss by using real-time atmospheric O₃ accurately.
251 The crop yield responses to different atmospheric O₃ concentrations present significant variations and
252 indicate that low O₃ stress (30-40 ppb) has considerable damage to crop yield. Based on an interval
253 evaluation, we demonstrate the spatial quantification of crop yield loss rate under O₃ stress globally,
254 including much more significant crop production losses than previously reported. Finally, the
255 co-regulation of crop yield response to elevated O₃ by crop photosynthetic and agronomic indexes
256 signifies the necessity of comprehensive measures to improve crop resistance against O₃ stress. These
257 results are crucial to identifying crop yield-sensitive regions under global O₃ pollution and may help
258 facilitate an appropriate response at the scientific and policy level.

259 **Methods**

260 **Experimental data collection.** To establish a standardized and unified database of responses of crop

261 yield to atmospheric O₃ concentration, experimental data that met the following criteria were collected
262 through Web of Science (<http://apps.webofknowledge.com>), Google Scholar
263 (<https://scholar.google.com>), and China Knowledge Resource Integrated Database
264 (<http://www.cnki.net/>). A wide range of keywords ("ozone* yield", "ozone* wheat", "ozone* maize or
265 corn", "ozone* rice", "ozone* vegetable", "ozone* production", and "grain yield") were used. The
266 target literature was obtained directly from the corresponding authors because of paper download and
267 subscription permissions. The PRISMA flow chart showed the process of literature collection until
268 November 2020 (Supplementary Fig. 1).

269 To standardize the database, experimental data were only when the following criteria were met: (1)
270 the experiment included O₃ fumigation and no O₃ fumigation (control) treatments; (2) no
271 anthropogenic simulation (e.g., elevated carbon dioxide was included; (3) experimental period, O₃
272 fumigation concentration, O₃ exposure time (hour day⁻¹), and crop yield were reported via figures,
273 tables and text; (4) crop was planted directly in the soil, and the variety was given; (5) data were
274 excluded if they were previously reported. Get Data Graph Digitizer 2.24 (free software) was used to
275 derive data from figures. Data presented as equations were excluded.

276 To make the database consistent, units of partial data were converted. For crop yield, if the target
277 literature described only the percentage of O₃ fumigation effects on crop yield under O₃ fumigation
278 treatment, then the value of 1 and 1 - the percentage was recorded under control and O₃ fumigation
279 treatments. For O₃ concentration, the unit of part per billion (ppb) was considered as the only unit of O₃
280 concentration under O₃ fumigation and control treatments. The parts per million (ppm) and nmol mol⁻¹
281 were converted to ppb by using the following equation:

$$282 \quad 1 \text{ ppb} = 0.001 \text{ ppm} \quad (1)$$

$$283 \quad 1 \text{ ppb} = 22.4/48 \text{ ug/m}^3 \quad (2)$$

284 The data represented 960 O₃ fumigation experiments reported in 208 published articles/reports that
285 tested the effect of elevated O₃ concentration on crop yield. This included 363 experiments for wheat,
286 31 experiments for maize, 305 experiments for rice, 147 experiments for soybean, and 114 experiments
287 for various vegetables, respectively. Besides the experimental period, O₃ fumigation concentration,
288 exposure time, and crop yield measures, photosynthetic and agronomic indexes were also included in
289 the database, which explained the variation in Y_o. Photosynthetic indexes had a light-saturated rate
290 (191 experiments), stomatal conductance (169 experiments), leaf chlorophyll content (170

291 experiments), and leaf injury (41 experiments), correspondingly. Agronomic indexes included
292 aboveground biomass (266 experiments), total biomass (218 experiments), grain number per ear (316
293 experiments), ear number per plant (343 experiments), grain-setting percentage (180 experiments),
294 1000-grain weight (379 experiments), and harvest index (225 experiments), and leaf area index (190
295 experiments), respectively. The mean, standard deviations (SD), and sample size (N) of photosynthetic
296 and agronomic indexes under O₃ fumigation and control treatments were recorded in the database
297 together with the crop yield. If only the standard error (SE) was reported in the target literature, SD was
298 transformed by: $SD = SE\sqrt{N}$. If SD or SE was not reported, the missing SD was replaced by
299 multiplying the corresponding mean times with the coefficient of 0.05. If N was not reported, the
300 disappeared N was replaced as the mean sample sizes of each crop type. Additionally, latitude and
301 longitude were extracted only to show the global distribution of the 960 experiments (Fig. 1a). In cases
302 where latitude and longitude were not reported across the target literature of the O₃ fumigation
303 experiments (34% of target literature did not report latitude and longitude), the approximate latitude
304 and longitude were obtained by inputting the name of the experimental site into Google Earth 7.0 (the
305 free version). It did not affect the major results. Furthermore, the author and publication year were
306 recorded and used to test the publication bias (Supplementary Fig. 2). Overall, the sites of our global
307 study spanned from -1.26° to 57.92° and -123.23° to 140.21° in latitude and longitude, respectively.

308 **The O₃ sensitivity of crop yield.** The Y_o is crop yield loss rate (%) relative to elevated O₃
309 concentration per 1 ppb h⁻¹. This is applied to normalize the effects of atmospheric O₃ on crop
310 production. One primary objective of our study was to precisely define the Y_o under different O₃
311 intervals using a meta-analysis approach. Meta-analysis is a comprehensive statistical strategy to
312 systematically combine and quantitatively evaluate multiple independent research results with a
313 common research purpose, which is particularly suitable for the large-scale study³⁶.

314 Before performing the meta-analysis, the quality of experimental data was using the "metainf"
315 package (Supplementary Fig. 2)³⁷. This is a method of combining publication bias and treatment to
316 explore any source of publication bias. Such discrimination could reduce the small-sample effects by
317 publication bias and ensure the credibility of the results³⁸. If a control corresponds to more than one
318 experimental treatment at a study site, such treatments are considered non-independent of sampling.
319 Previous studies have shown that the non-independence of the sample can significantly affect the
320 research results, but such studies have also offered solutions^{36, 39, 40}. According to the principles of

321 statistics and study purpose, the data of different O₃ concentrations under O₃ fumigation treatment was
 322 regarded as non-independence of sampling compared with the same control treatment. Therefore, the
 323 mean and SD of non-independence were weighted based on the concentration of different O₃
 324 fumigation. The following equation calculated the weight of O₃ fumigation concentrations (W_f):

$$325 \quad W_f = \frac{C}{\sum_{i=1}^n C_i} \quad (3)$$

326 where C is the concentration of different O₃ fumigation (ppb); n is the number of O₃ fumigation
 327 concentrations under the same control treatment.

328 To quantify the magnitude of O₃ sensitivity of crop yield, we first calculated the response ratio (RR)
 329 of O₃ fumigation on crop yield. The RR of treatment was calculated as the following equation:

$$330 \quad RR = \frac{X_f}{X_c} \quad (4)$$

331 where X_f and X_c are the crop yield under O₃ fumigation and control treatments, respectively.

332 The value of RR less than 1 indicates a negative effect of O₃ fumigation on crop yield. The
 333 meta-analysis is the comparison of treatments including even different variables⁴¹. The RR is natural
 334 log-transformed to approach the normal distribution:

$$335 \quad \ln(RR) = \ln\left(\frac{X_f}{X_c}\right) = \ln(X_f) - \ln(X_c) \quad (5)$$

336 The overall response ratio (Ln(RR₊)) of a group was calculated as follow:

$$337 \quad \ln(RR_+) = \frac{\sum_{i=1}^n \ln(RR_i) * W_i}{\sum_{i=1}^n W_i} \quad (6)$$

338 where n is the number of a group. W_i is the weighting factor of the *i*th data in the group. The W_i is
 339 calculated as the following equation:

$$340 \quad W_i = \frac{1}{V_i} \quad (7)$$

341 where V_i is the variance of *i*th data. The V_i was calculated as the following equation:

$$342 \quad V_i = \frac{SD_f^2}{n_f X_f^2} + \frac{SD_c^2}{n_c X_c^2} \quad (8)$$

343 where n_f and n_c are the numbers of samples for O₃ fumigation and control treatments, respectively. SD_f
 344 and SD_c are the standard deviations for O₃ fumigation and control treatments, respectively.

345 The standard error (SD(Ln(RR₊))) and 95% confidence interval (CI) of the Ln(RR₊) were calculated
 346 by the following equations:

$$347 \quad SD(\ln(RR_+)) = \frac{1}{\sqrt{\sum_{i=1}^n W_i}} \quad (9)$$

348
$$95\%CI = \text{Ln}(RR_+) \pm 1.96SD(\text{Ln}(RR_+)) \quad (10)$$

349 O_3 fumigation significantly affects the crop yield if the 95% confidence interval does not overlap
 350 with 1. If the 95% confidence interval of two variables does not overlap, they are considered
 351 significantly different. The following equation transformed the effect size (ES, %):

352
$$ES = (e^{\text{Ln}(RR_+)} - 1) * 100\% \quad (11)$$

353 A value of ES less than 0 indicates a negative effect of O_3 fumigation on crop yield. A meta-analysis
 354 should be performed to correctly and effectively detect heterogeneity in data before merging it, i.e.,
 355 heterogeneity test. Previously, a chi-square test was used as a tool for testing heterogeneity. However, it
 356 has been found that the chi-square test lacks efficacy and has no statistical significance for the
 357 existence of heterogeneity for small samples. At present, the most commonly used heterogeneity testing
 358 methods can be divided into two kinds: the graphical method and the systematic measurement method.
 359 For latter, if $P > 0.1$ and $I^2 < 50\%$ (no heterogeneity), the fixed-effect model is selected for
 360 meta-analysis. Otherwise, the random effect model is selected. The meta-analysis was performed using
 361 the MetaWin 2.0⁴².

362 The Y_o (ppb h⁻¹) was calculated as follows:

363
$$Y_o = \frac{ES}{(O_f - O_c) * d * h} \quad (12)$$

364 where O_f and O_c are O_3 concentrations (ppb) for O_3 fumigation and control treatments, respectively. d
 365 and h are the days of O_3 fumigation and hours of O_3 fumigation per day, respectively.

366 Previous studies indicated that atmospheric O_3 concentrations above 40 ppb could significantly
 367 affect crop yield^{28,43}. Based on our dataset, atmospheric O_3 can dramatically affect crop yield even at
 368 30-40ppb (Supplementary Fig. 3). Therefore, we calculated the Y_o of different crop (wheat, maize, rice,
 369 soybean, and vegetable) yield under 7 intervals (30-40, 40-50, 50-60, 60-70, 70-80, 80-90, and >90 ppb)
 370 according to O_3 concentration of fumigation treatment (Fig. 1c). The Y_o of maize under the 30-40 ppb
 371 interval was obtained using a fitting equation due to the missing data (Supplementary Fig. 4). Based on
 372 the above principles and methods, we also calculated the O_3 sensitivity of photosynthetic indexes
 373 (light-saturated rate, stomatal conductance, leaf chlorophyll, and leaf injury) and agronomic indexes
 374 (aboveground biomass, total biomass, grain number per ear, ear number per plant, setting percentage,
 375 thousand seed weight, harvest index, and leaf area index), respectively. This was done to try and
 376 determine the mechanistic basis for any O_3 damage relative to proportional changes in crop yield.

377 **Global real-time O_3 concentration data.** The global crop yield loss rate is dependent on global

378 real-time O₃ concentrations during the crop growing season. In this study, we used hourly O₃
 379 concentrations as the standard unit. The hourly O₃ concentrations, in turn, were derived by searching
 380 and/or directly accessing air quality monitoring stations in each country. An air quality monitoring
 381 station had to meet the following three criteria in our database: (1) air quality monitoring station must
 382 report the exact latitude and longitude; (2) O₃ concentration must be recorded on an hourly basis. If
 383 only the average of one day was given, data were excluded; (3) a complete record of at least two years
 384 of data from 2017 to 2019. Any air quality monitoring station reported in the parts per million, or nmol
 385 mol⁻¹ units were converted to ppb using equations 1-2. Overall, the hourly O₃ database includes about
 386 160 million data pairs (latitude, longitude, time, and O₃ concentration) from 7246 stations in more than
 387 60 countries or regions (Supplementary Fig. 5). Except for missing data for some countries (e.g., Japan
 388 for 2019), the integrity rate of the database is more than 94%.

389 The hourly O₃ concentration was divided into 7 intervals (30-40, 40-50, 50-60, 60-70, 70-80, 80-90,
 390 and >90 ppb). If the O₃ concentration is below 30 ppb for an hour, the 7 intervals are assigned 0 ppb. If
 391 the O₃ concentration for an hour is more than 90 ppb, the first 6 intervals are assigned 10 ppb, and the
 392 7th interval (>90 ppb) is the difference of O₃ concentration of that hour and 90 ppb. For every air
 393 quality monitoring station, the crop yield loss rate (%) of total and each O₃ interval was calculated as
 394 follows:

$$395 \quad \text{Crop yield loss rate} = \sum_{g=1}^{ng} \left(\sum_{m=1}^{nm} \left(\sum_{i=1}^{ni} \left((-OY_{ith}) * O_{ith} \right) \right) \right) * 100\% \quad (13)$$

396 where i, m, and g are the *i*th O₃ intervals, the *m*th hour and *g*th day, respectively. *n_i* is total O₃ intervals
 397 for an hour. *n_m* is 12, meaning 12 hours per day (08:00-20:00). *n_g* is the number of days of crop
 398 growing season.

399 Although we have tried to obtain crop yield loss rate for more than 7,000 stations worldwide,
 400 however, the distribution of those stations is limited. To get the spatial predictions of the crop yield loss
 401 rate in each country or region, kriging classifications combined with regression were used in ArcGIS
 402 10.4.1 (version). The result was presented for 0.0083×0.0083° grid squares across the entire terrestrial
 403 system of the globe⁴⁴. Meantime, different crop distribution data (about 2 million) were obtained at the
 404 MAPSPAM (<https://www.mapspam.info/>) and used to get the spatial distribution of the crop yield loss
 405 rate in the cropland (Supplementary Fig. 6-9).

406 **Global crop production data.** To estimate the crop production loss caused by atmospheric O₃, the
 407 global crop production was obtained from the Food and Agriculture Organization of the United Nations

408 (FAO, <http://www.fao.org/faostat/en/#data/QC>). The FAO reports wheat, maize, rice, and soybean crop
409 productions for 200 countries or regions for the years 1961-2019. However, crop production data for
410 2017-2019 have been selected under real-time O₃ data as the national crop production data is not
411 consistent with that of the crop yield loss rate data on a grid square. Furthermore, this is the most recent
412 and complete observational data which we can get. Therefore, crop production loss of total and each O₃
413 interval was estimated on a national basis using the following equation:

$$414 \quad \text{Crop production loss} = \sum_{i=1}^{ni} (\text{Crop production} * \text{Crop yield loss rate}_{ith}) \quad (14)$$

415 **Structural equation model analysis.** Structural equation model (SEM) analysis was performed for
416 quantitative partitioning the direct and indirect pathways and determining whether atmospheric O₃
417 concentration influences the Y_o through crop indexes¹⁷. The 7 O₃ intervals (30-40, 40-50, 50-60, 60-70,
418 70-80, 80-90, and >90ppb) and four crop types (wheat, maize, rice, and soybean) were regarded as
419 treatment gradient to conducted the SEM analysis focusing on the overall effect. To simplify the model
420 framework, crop indexes were divided into two categories: agronomic indexes and photosynthetic
421 indexes. For agronomic indexes, the O₃ sensitivity of the aboveground biomass, grain number per ear,
422 and leaf area index were retained as latent variables according to loading scores. For the latent variable
423 photosynthetic indexes, three potential indicators were: O₃ sensitivity of light-saturated rate, stomatal
424 conductance, and chlorophyll (Supplementary Fig. 11). The following possible pathways were
425 hypothesized: first, O₃ concentration, agronomic indexes, and photosynthetic indexes have a direct
426 effect on the Y_o; second, O₃ concentration may indirectly affect the Y_o via its impact on agronomic and
427 photosynthetic indexes. Finally, photosynthetic indexes may indirectly affect the Y_o via their effects on
428 agronomic indexes. SEM analysis assumes that the variance-covariance matrix of the observed variable
429 is a function of a set of parameters. Its estimation requires minimizing the difference between the
430 variance-covariance value of the sample and the value estimated by the model and taking the difference
431 as the residual. The maximum likelihood (estimate means and intercepts) was used to assess the path
432 parameter. A nonparametric bootstrap method was adopted to calculate the accuracy of parameter
433 estimate¹⁷. The standardized total effect was shown to express the relative impact of O₃,
434 photosynthetic indexes, and agronomic indexes on the Y_o. Amos 17.0 for Windows (version) was used
435 to perform the SEM analysis.

436 **Data availability**

437 All data related to this manuscript are available from the Dryad Digital Repository:

438 <https://figshare.com/s/92384712469fe5942df0>.

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536

537 **Figure Legends**

538 **Fig. 1 Ozone sensitivity of crop yield.** **a**, global distribution of the 960 experiments from 208
539 published papers in which the effect of elevated ozone concentration on crop yield was assessed. The
540 size of black circles represents the sample size ranging from 1 to more than 19. Crop types include
541 wheat (in brown), maize (in red), rice (in green), soybean (in turquoise), and vegetable (in blue). **b**,
542 response ratio (natural logarithm-transformed ratio of treatment to control) of the ozone sensitivity of
543 crop yield (ppb h⁻¹). Dot and bar represent the mean and range at 95% confidence intervals of ozone
544 sensitivity of crop yield. The value in parentheses represents the sample sizes. **c**, the relationship
545 between ozone sensitivity of crop yield and ozone fumigation concentration. Each dot represents the
546 average effect size under ozone fumigation intervals with <30, 30-40, 40-50, 50-60, 60-70, 70-80,
547 80-90, >90 ppb for each crop type according to the treatment groups in the database.

548 **Fig. 2 Global annual loss rate (%) of crop yield (2017-2019) due to atmospheric ozone**
549 **concentration.** The maps showed the spatial distribution of cumulative crop yield loss (%) in the case
550 of ozone concentration above 30 ppb for wheat (**a1**), maize (**b1**), rice (**c1**), and soybean (**d1**), averaged
551 for the period 2017-2019. The spatial distribution of crop yield loss in the case of ozone above 30, 40,
552 50, 60, 70, 80, and 90 ppb for wheat, maize, rice, and soybean were also shown in Supplementary Fig.
553 6-9, respectively. All data were presented for the 0.0083° grid squares based on global 7246 real-time
554 ozone monitoring stations (see Supplementary Fig. 5). **a2, b2, c2, and d2** indicated crop yield loss of
555 wheat, maize, rice, and soybean in the case of ozone concentration ranged with 30-40, 40-50, 50-60,
556 60-70, 70-80, 80-90, and >90 ppb for global and 5 countries or regions (ranked top five in grain
557 production) averaged value, respectively. The percentage of ozone monitoring stations involved in
558 calculating the effects of ozone on crop yield at each range of ozone concentration was shown in

559 Supplementary Table 1.

560 **Fig. 3 Influence of ozone (O_3) and ozone sensitivity of photosynthetic (P_o) and agronomic (A_o)**
561 **indexes on the ozone sensitivity of crop yield (Y_o).** a, path analysis results on the direct and indirect
562 effects of O_3 , P_o , and A_o on the Y_o . Numbers show the path coefficients. Grey path and number indicate
563 that the effect is insignificant. Arrow width is proportional to the standardized coefficient. The
564 P_o includes ozone sensitivity of light-saturated rate, ozone sensitivity of stomatal conductance, and
565 ozone sensitivity of chlorophyll (See supplementary Fig. 11). The A_o has the ozone sensitivity of above
566 ground biomass, the ozone sensitivity of grain number per ear, and the ozone sensitivity of leaf area. b,
567 the standardized total effect of O_3 , P_o , and A_o on the Y_o .

568 **Tables**

569 **Table 1 Global and five areas (ranked top five in grain production) annual loss amount of wheat,**
570 **maize, rice, and soybean production (million tonnes) in the case of ozone 30-40 ppb and above 40**
571 **ppb. Each country's annual loss rate of crop yield in the case of ozone above 30, 40, 50, 60, 70, 80,**
572 **and 90 ppb and total yield for wheat, maize, rice, and soybean were shown in Supplementary**
573 **Table 2-6.**

574

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579 **Author contributions**

580 A.D., B., W.J., and Y.E. designed the study. A.D. and T.J. collected the data. A.D. analyzed the data. All
581 authors contributed significantly to the writing of the manuscript.

582 **Competing interests**

583 The authors declare no competing interests.

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