## 1 **Text S1**

2 We use the following multivariate predictive model (e.g., Stern and Kaufmann 2013, Mosedale

3 *et al* 2006) to estimate the causal links between the ENSO and ozone concentration:

4 
$$X_t = \sum_{i=1}^p \alpha_i X_{t-i} + \sum_{i=1}^p \beta_i Y_{t-i} + \sum_{j=1}^m \sum_{i=1}^p \delta_{j,i} Z_{j,t-i} + \varepsilon_t$$
(1)

5 where  $X_t$  is the annual mean (or seasonal mean) ozone concentration for year t,  $Y_t$  is the ENSO index, and  $Z_{j,t}$  is the confounding factor j for year t. In the predictive model shown in equation 1, 6 while estimating the influence of Y on X (i.e., the contribution of the term  $\sum_{i=1}^{p} \beta_i Y_{t-i}$  in 7 8 predicting X), the contribution of past X events are already taken into account by adding the term  $\sum_{i=1}^{p} \alpha_i X_{t-i}$ . Thus, the causal influence of Y on X, if detected, is robust and the contribution of 9 past X events are already considered in our analyses. Here, m is number of confounding factors 10 11 and  $p \ge 1$  is the order of the multivariate predictive model. The optimal order p is computed by 12 minimizing the Schwarz criterion or the Bayesian information criterion (Schwarz 1978). The 13 optimal orders might be different for each model.

14 The ENSO index was computed as the average sea surface temperature (SST) anomalies in the Niño 3.4 area (120-170°W; 5°N-5°S) in boreal winter (December-January-February, DJF). 15 16 Confounding factors (i.e., the dipole mode index (DMI; Saji et al., 1999), the Southern Annular 17 Mode (SAM) and the North Atlantic Oscillation (NAO; e.g., Hurrell et al., 2003)) may have 18 effects on the connections between ENSO and ozone concentration. The DMI was given as the 19 difference in boreal fall (September-October-November, SON) SST anomalies between two 20 Indian Ocean regions of the western pole (50–70°E; 10°N–10°S) and southeastern pole (90– 21 110°E; 0°N–10°S). The SAM (Cai et al., 2011) was calculated as the first empirical orthogonal 22 function (EOF) of the boreal summer (June-July-August, JJA) sea level pressure (SLP) 23 anomalies for the region of 40-70°S. The NAO index is computed as the EOF of boreal winter (DJF) SLP anomalies in the North Atlantic area (90°W-40°E, 20°-70°N). 24

We estimate the probability of no Granger causality by applying a test of Granger causality (Le and Bae, 2020; Mosedale et al., 2006; Stern and Kaufmann, 2013) for the multivariate predictive model shown in equation 1. For computing the degree of uncertainty, we followed recent guidance (Stocker et al., 2013) and utilized the terms 'very unlikely', 'unlikely', 'likely' for the 0–10%, 0–33%, and 66–100% probability of the likelihood of the outcome, respectively. For

- 30 example, if the *p*-value is less than 0.33, the result indicates that ENSO is unlikely to display no
- 31 Granger causality on ozone concentration. In this instance, we conclude that ENSO has 'causal
- 32 effect' on ozone concentration.

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- 56

| No. | Model name      | Modelling center, country | Atmospheric Chemistry model |
|-----|-----------------|---------------------------|-----------------------------|
| 1   | BCC_CSM2_MR     | BCC, China                | None                        |
| 2   | BCC_ESM1        | BCC, China                | BCC-AGCM3-Chem              |
| 3   | CESM2           | NCAR, United States       | MAM4                        |
| 4   | CESM2_FV2       | NCAR, United States       | MAM4                        |
| 5   | CESM2_WACCM     | NCAR, United States       | MAM4                        |
| 6   | CESM2_WACCM_FV2 | NCAR, United States       | MAM4                        |
| 7   | CNRM_CM6_1      | CNRM-CERFACS, France      | OZL_v2                      |
| 8   | CNRM_CM6_1_HR   | CNRM-CERFACS, France      | OZL_v2                      |
| 9   | CNRM_ESM2_1     | CNRM-CERFACS, France      | REPROBUS-C_v2               |
| 10  | IPSL_CM6A_LR    | IPSL, France              | None                        |
| 11  | MPI_ESM_1_2_HAM | MPI-M, Germany            | Sulfur chemistry (unnamed)  |
| 12  | MPI_ESM1_2_LR   | MPI-M, Germany            | None                        |

**Table S1.** List of CMIP6 models used in this study.

MODELS STD OF ANNUAL OZ300 (ppbv) PERIOD 1850-2014 EXPERIMENT HISTORICAL

MODELS STD OF ANNUAL OZ500 (ppbv) PERIOD 1850-2014 EXPERIMENT HISTORICAL



MODELS STD OF ANNUAL OZ850 (ppbv) PERIOD 1850-2014 EXPERIMENT HISTORICAL



MODELS STD OF ANNUAL OZ1000 (ppbv) PERIOD 1850-2014 EXPERIMENT HISTORICAL



- 59
- 60 Figure S1. Multi-model mean map of standard deviation of annual ozone concentrations (ppbv) for the
- 61 historical experiment over the 1850-2014 period at 300 hPa (a), 500 hPa (b), 850 hPa (c) and 1000 hPa
- 62 (d) pressure levels, respectively.



- 64 **Figure S2.** Probability for the absence of Granger causality from ENSO to annual ozone concentrations at
- 65 300 hPa pressure level for the historical experiment over the 1850-2014 period of 12 individual models
- 66 (see Table S1). The cyan and yellow contour lines signify p-value = 0.33 and 0.1, respectively. Brown
- 67 shades denote a low probability for the absence of Granger causality. ENSO: El Niño–Southern
- 68 Oscillation.



- **Figure S3.** As in Figure S2, but for the absence of Granger causality from ENSO to annual ozone concentrations at 500 hPa pressure level. ENSO: El Niño–Southern Oscillation.
- 71



74 **Figure S4.** As in Figure S2, but for the absence of Granger causality from ENSO to annual ozone concentrations at 850 hPa pressure level. ENSO: El Niño–Southern Oscillation.

MODELS MEAN: ENSO - SPRING OZONE (300 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL MODELS MEAN: ENSO - SUMMER OZONE (300 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL



MODELS MEAN: ENSO - FALL OZONE (300 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL MODELS MEAN: ENSO - WINTER OZONE (300 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL





76 **Figure S5.** Multi-model mean probability map for the absence of Granger causality from ENSO during

boreal winter [defined as D(t)JF(t+1); t denotes year t] to seasonal mean ozone concentrations at 300 hPa

78 pressure level over the period 1850-2014. (a) Spring [March, April, May; MAM(t+1)]. (b) Summer [June,

July, August; JJA(t+1)]. (c) Fall [September, October, November; SON(t+1)]. (d) Winter [December,

Solution January, February; D(t+1)JF(t+2)]. Stippling demonstrates that at least 70% of total models show

81 agreement on the mean probability of all models at a given grid point. The cyan contour line signifies *p*-

82 value = 0.33. Brown shades denote a low probability for the absence of Granger causality. ENSO: El

83 Niño–Southern Oscillation.

MODELS MEAN: ENSO - SPRING OZONE (500 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL MODELS MEAN: ENSO - SUMMER OZONE (500 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL



MODELS MEAN: ENSO - FALL OZONE (500 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL MODELS MEAN: ENSO - WINTER OZONE (500 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL



- 84
- 85 **Figure S6.** As in Figure S5, but for multi-model mean probability map for the absence of Granger
- 86 causality from ENSO to seasonal mean ozone concentrations at 500 hPa pressure level. ENSO: El Niño-
- 87 Southern Oscillation.

MODELS MEAN: ENSO - SPRING OZONE (850 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL MODELS MEAN: ENSO - SUMMER OZONE (850 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL



MODELS MEAN: ENSO - FALL OZONE (850 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL MODELS MEAN: ENSO - WINTER OZONE (850 hPa) PERIOD 1850-2014 EXPERIMENT HISTORICAL



- 89 **Figure S7.** As in Figure S5, but for multi-model mean probability map for the absence of Granger
- 90 causality from ENSO to seasonal mean ozone concentrations at 850 hPa pressure level. ENSO: El Niño-
- 91 Southern Oscillation.