

Supporting Information for

**Climate-mediated surface ozone responses to
simultaneous multi-regional marine cloud brightening**

Yiqian Ni^{1,2}, Yang Yang^{1,2*}, Hailong Wang³, Yang Yu⁴, Pinya Wang^{1,2}, Hong
Liao^{1,2}

¹State Key Laboratory of Climate System Prediction and Risk
Management/Jiangsu Key Laboratory of Intelligent Atmospheric Environment
Monitoring and Carbon–Pollution Co-control/Jiangsu Collaborative Innovation
Center of Atmospheric Environment and Equipment Technology/Joint International
Research Laboratory of Climate and Environment Change, Nanjing University of
Information Science and Technology, Nanjing, Jiangsu, China

²School of Environmental Science and Engineering, Nanjing University of
Information Science and Technology, Nanjing, Jiangsu, China

³Atmospheric, Climate, and Earth Sciences Division, Pacific Northwest National
Laboratory, Richland, WA, USA.

⁴Department of Earth and Environmental Sciences, Faculty of Science, The
Chinese University of Hong Kong, Sha Tin, Hong Kong SAR, China

Contents of this file

Figures S1-S7, Tables S1-S2, and Text S1-S2.

Introduction

This auxiliary material contains supporting information for Climate-mediated surface ozone responses to simultaneous multi-regional marine cloud brightening.

*Correspondence to yang.yang@nuist.edu.cn



Figure S1. Effective radiative forcing (ERF) as a function of sea salt aerosol (SSA) injection (unit: Tg per region). Points represent model results and the dashed curve shows the quadratic fit.

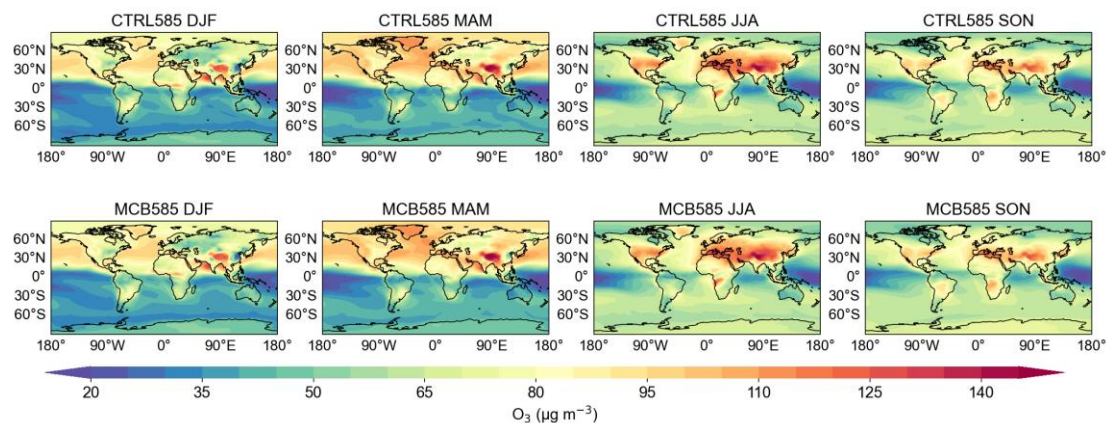


Figure S2. Seasonal mean surface O_3 concentrations ($\mu g m^{-3}$) in CTRL₅₈₅ (top) and MCB₅₈₅ during (bottom) DJF (December–January–February), MAM (March–April–May), JJA (June–July–August), and SON (September–October–November).

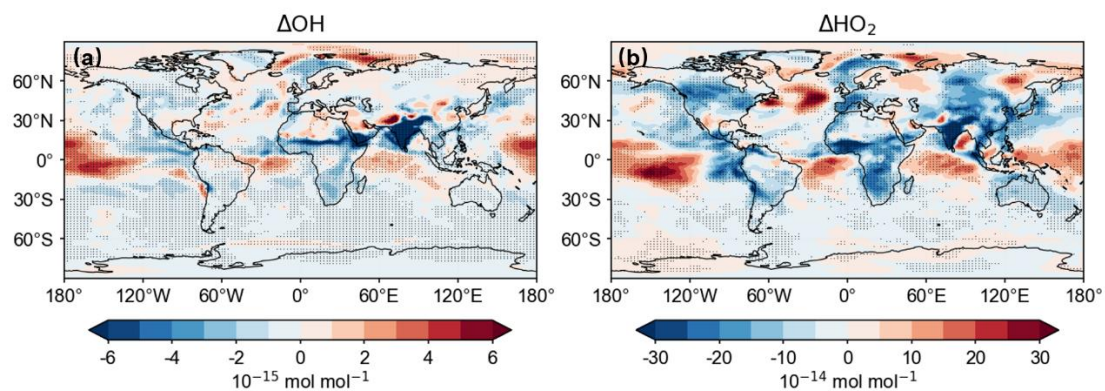


Figure S3. Changes in surface hydroxyl radical concentration (ΔOH , mol mol^{-1}) and hydroperoxyl radical concentration (ΔHO_2 , mol mol^{-1}) due to MCB-induced climate change during boreal summer (June to August). Stippling marks grid cells with $p < 0.05$ based on independent two-sample t-tests.

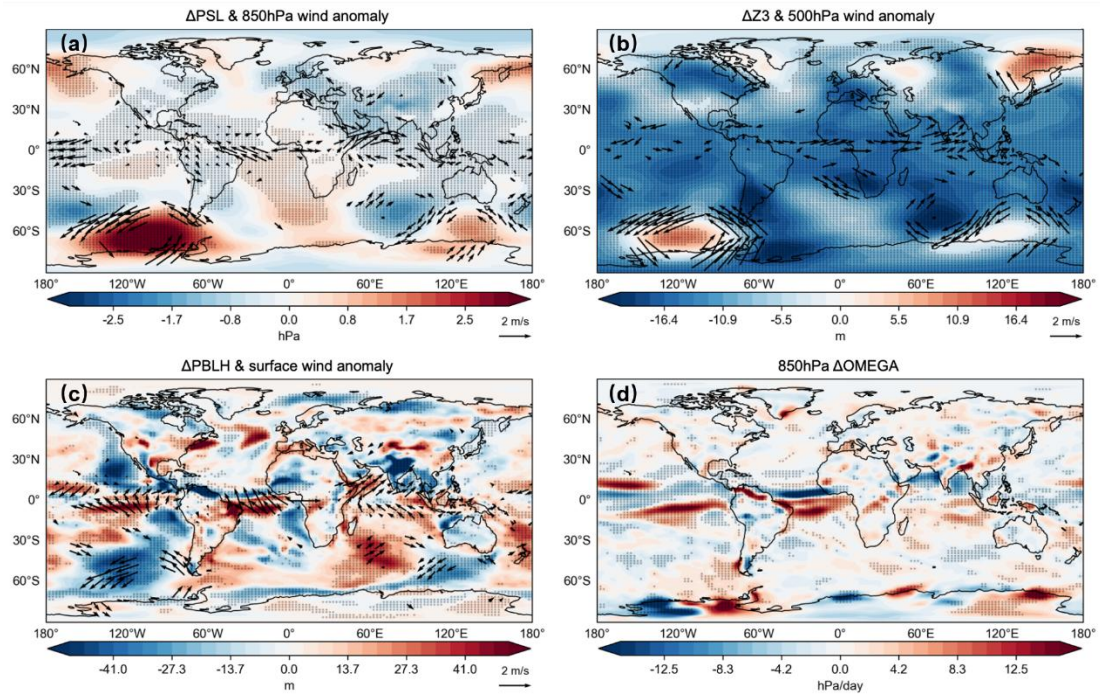


Figure S4. Changes in (a) sea level pressure (PSL, Pa) and 850 hPa winds, (b) 500 hPa geopotential height (Z3, m) and winds, (c) planetary boundary-layer height (PBLH, m) and surface winds, and (d) 850 hPa vertical pressure velocity (OMEGA, hPa day⁻¹) due to MCB-induced climate change during boreal summer (June to August). Stippling marks grid cells with $p < 0.05$ based on independent two-sample t-tests.

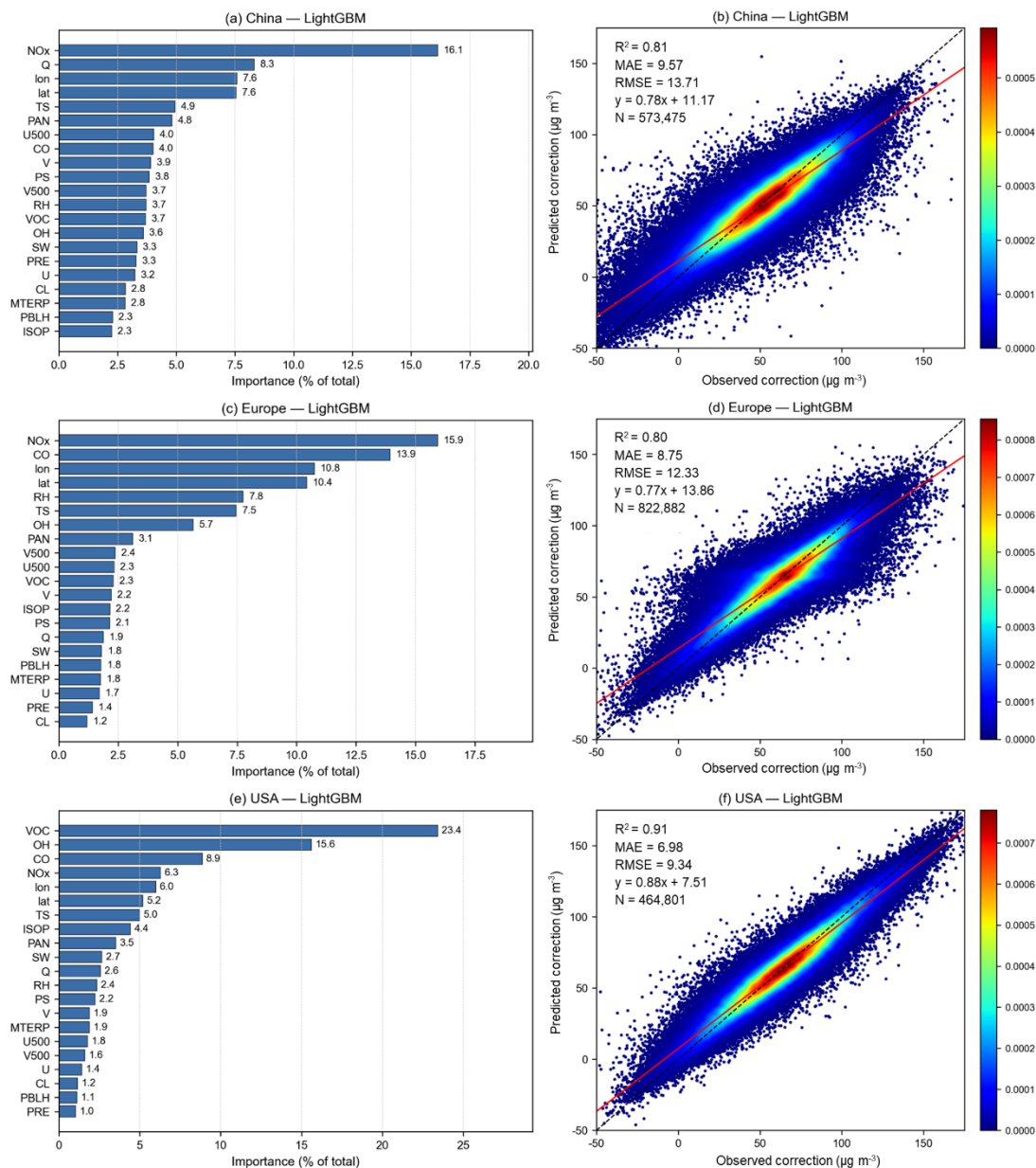


Figure S5. Gain-based feature importance, normalized to the total importance, and density scatter plots of predicted versus observed surface O₃ correction for (a, b) China, (c, d) Europe and (e, f) the United States. Right panels show held-out test-set performance, with color indicating bivariate kernel density, the dashed black line showing the 1:1 relationship, and the solid red line showing the linear fit. R^2 , MAE, RMSE, and sample size are reported in each panel.

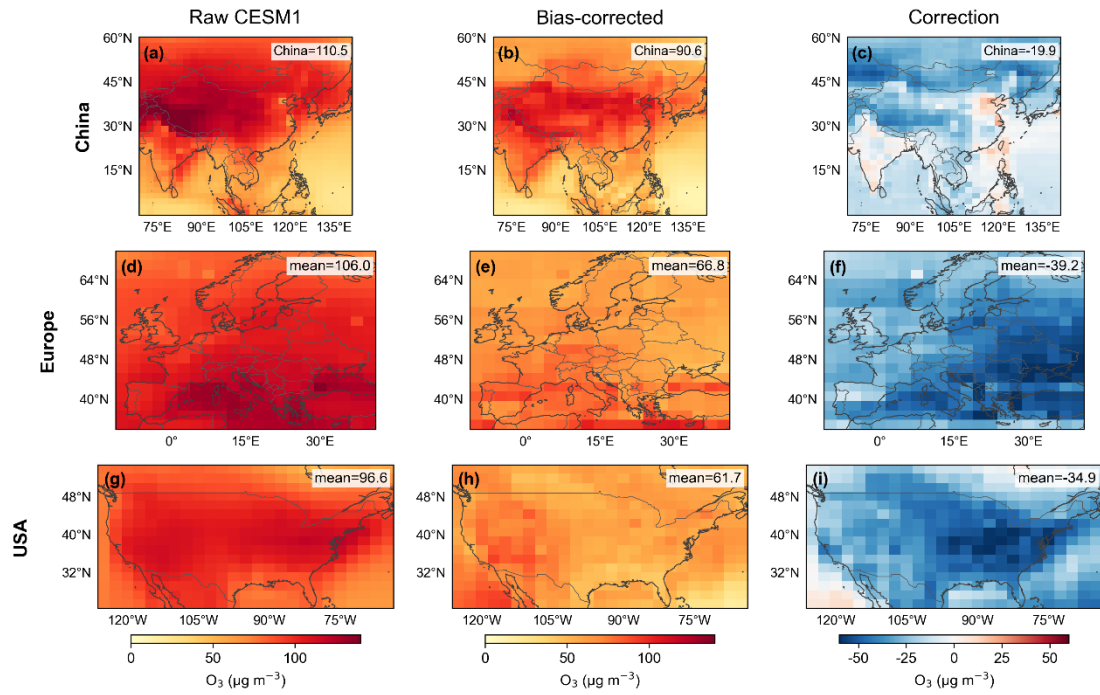


Figure S6. Spatial distribution of the surface O_3 concentrations during boreal summer (June to August) in CTRL₅₈₅ over the China, Europe, and the United States. Columns show raw CESM1 O_3 , bias-corrected O_3 , and the correction predicted by the correction model relative to the raw CESM1 values.

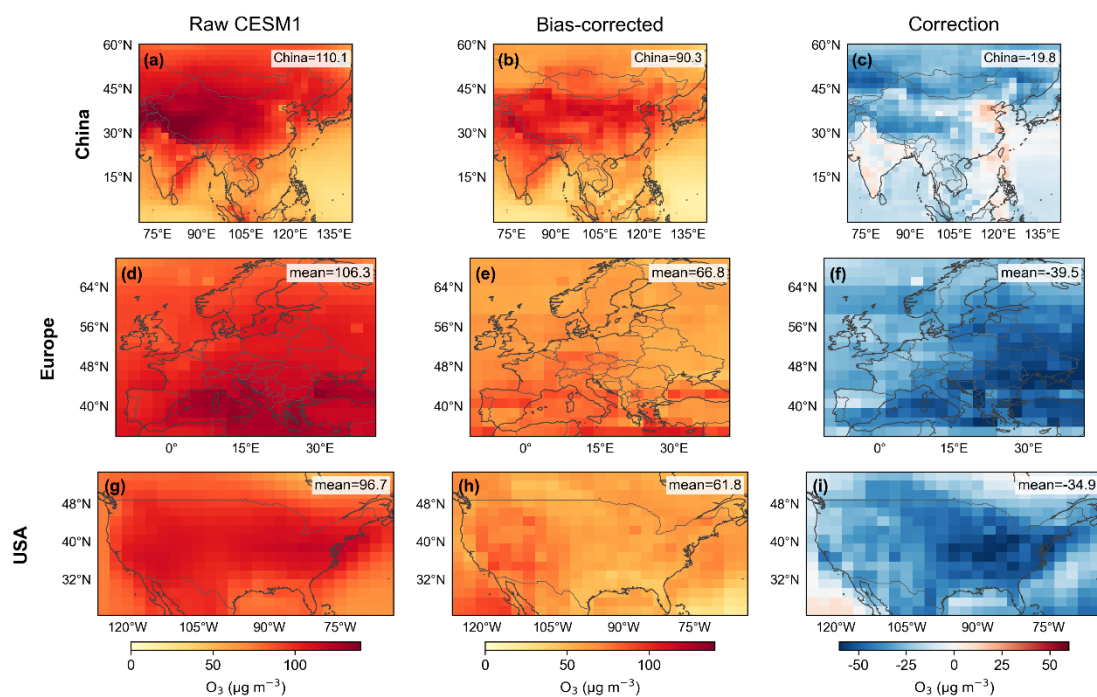


Figure S7. Same as Figure S6 but for MCB₅₈₅.

Table S1. Predictors used in the regional LightGBM bias-correction models.

Type	Abbreviation	Description	Spatial resolution	Time resolution (period)
Meteorology	LON	Longitude	/	/
	LAT	Latitude		
	TS	Surface temperature	1.9°×2.5°	Daily (2014-2019)
	PS	Surface pressure		
	U	10-meter zonal wind		
	U500	500 hPa zonal wind		
	V	10-meter meridional wind		
	V500	500 hPa meridional wind		
	RH	Relative humidity		
	Q	Specific humidity		
	PRE	Total precipitation rate		
	CL	Cloud cover fraction		
	SW	Net solar flux at surface		
	PBLH	Planetary Boundary Layer Height		
Chemistry	NO _x	Sum of NO and NO ₂ concentration		
	CO	CO concentration		
	OH	OH concentration		
	VOC	VOC concentration		
	MTERP	MTERP concentration		
	ISOP	ISOP concentration		
	PAN	PAN concentration		

Text S1. Training and evaluation of the LightGBM bias-correction models

Separate LightGBM models (Ke et al., 2017) were trained for China, Europe, and the United States to predict the CESM1 surface O₃ correction, defined as observed minus simulated O₃. Each model used 21 site-level daily predictors for 2014–2019, including longitude and latitude, near-surface meteorology (TS, U, V, U500, V500, RH, Q, PS, PBLH, PRE, SW, and CL), and surface-layer chemistry (CO, NO_x, OH, VOC, ISOP, MTERP, and PAN) (Table S1). The data were divided into an 80% training set and a 20% held-out test set using `random_state = 42`. Hyperparameters were optimized with 50 Optuna trials (Akiba et al., 2019) using only the training data.

Model performance was evaluated using R², MAE, and RMSE (Table S2; Figure S5). Test-set R² values are 0.81 for China, 0.80 for Europe, and 0.91 for the United States. The corresponding MAE values are 9.57, 8.75, and 6.98 µg m⁻³, and RMSE values are 13.71, 12.33, and 9.34 µg m⁻³. These results indicate that the models reproduce much of the observed spatial and temporal variability in the CESM1 correction within the sampled data. Because the test set was generated by random splitting, the reported metrics mainly characterize interpolation within the available station-period coverage and may not fully represent transfer to unsampled locations or future conditions.

Gain-based feature importance differs among the regional models (Figure S5). NO_x is the leading predictor over China (16.1%) and Europe (15.9%), followed by Q over China (8.3%) and CO over Europe (13.9%). Over the United States, VOC (23.4%) and OH (15.6%) have the highest importance.

Text S2. Training and evaluation of LightGBM models for meteorological attribution of corrected O₃

To quantify the relative contributions of meteorological perturbations to the MCB-induced ozone response, we constructed regional LightGBM regression models using the bias-corrected surface O₃ response as the dependent variable. For each grid cell, one sample was generated from the 50-year JJA mean difference between the MCB and CTRL simulations. The predictors were calculated consistently as the 50-year JJA mean MCB–CTRL differences of TS, QREFHT, RH, PBLH, PS, FSNS, CL, U, V, OMEGA, and PRE. The numbers of valid grid cells were 246 for China, 228 for Europe, and 295 for the USA.

The JJA means were calculated from the 600-month CESM simulations by selecting the 150 JJA months and averaging the three summer months for each year, followed by averaging across the 50 simulation years. Cloud fraction was represented by the lowest-level CLOUD variable. Total precipitation was calculated as the sum of large-scale and convective precipitation ($PRE=PRECL+PRECC$).

For each region, the data were randomly divided into training and test subsets using a 80:20 split with a fixed random seed of 42. Hyperparameters were selected using four-fold shuffled K-fold cross-validation within the training subset. The selected hyperparameters were region-specific.

Model performance was evaluated using the coefficient of determination (R^2) and root-mean-square error (RMSE) on the held-out test subsets. The test R^2 values were 0.69, 0.68, and 0.52 for China, Europe, and the USA, respectively, with corresponding RMSE values of 0.681, 0.492, and 0.692 in the units of corrected O₃.

Model interpretation was performed using TreeSHAP. Mean absolute SHAP values were calculated for the test samples and normalized to obtain the relative contribution of each meteorological variable. The individual variables were further aggregated into five process-based groups:

Thermal: TS,

Moisture: QREFHT, RH, PRE,

Radiative forcing: FSNS, CL,

Boundary layer mixing: PBLH, PS,

Transport: U, V, OMEGA.

The variables were grouped according to their expected physical roles in ozone photochemistry, moisture and wet removal, radiation, boundary-layer mixing, stagnation, and atmospheric transport. The contribution of each process was calculated as the sum of the normalized mean absolute SHAP values of its constituent variables. These physically motivated groups are not strictly independent because meteorological factors can co-vary. Thus, grouped SHAP values indicate the relative importance of process-related predictor sets within the fitted model rather than independent causal effects.

Table S2. Test-set performance of the regional LightGBM bias-correction models. R^2 is the coefficient of determination. MAE and RMSE are the mean absolute error and root-mean-square error ($\mu\text{g m}^{-3}$). N is the number of test samples.

Region	N	R^2	MAE	RMSE
China	573,475	0.809	9.57	13.71
Europe	822,882	0.800	8.75	12.33
United States	464,801	0.909	6.98	9.34

References From the Supporting Information

- Akiba, T., Sano, S., Yanase, T., Ohta, T., & Koyama, M. (2019). Optuna: A next-generation hyperparameter optimization framework. In Proceedings of the 25th ACM SIGKDD International Conference on Knowledge Discovery & Data Mining (pp. 2623–2631). Association for Computing Machinery. <https://doi.org/10.1145/3292500.3330701>
- Ke, G., Meng, Q., Finley, T., Wang, T., Chen, W., Ma, W., Ye, Q., & Liu, T.-Y. (2017). LightGBM: A highly efficient gradient boosting decision tree. In Advances in Neural Information Processing Systems 30 (NIPS 2017) (pp. 3146–3154). Curran Associates. <https://proceedings.neurips.cc/paper/2017/hash/6449f44a102fde848669bdd9eb6b76fa-Abstract.html>