Supplemental Information for

# Source specific bias correction of US background ozone modeled in CMAQ

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## Description of CMAQ simulations and O<sub>3</sub> components

Table S1. Simulation names and descriptions for hemispheric-scale and regional-scale simulations. Table adapted from 2020 O<sub>3</sub> Policy Assessment Table 2-1 (USEPA, 2020). Table S1 is reproduced from Table 1 in the main text to aid in interpreting Tables S2 and S3.

Simulation	Description
BASE	All emission sectors are included
ZUSA	All U.S. anthropogenic emissions are removed including prescribed fires.*
ZROW	All international anthropogenic emissions are removed including prescribed fires where
	possible.**
ZCANMEX	All anthropogenic emissions from Canada and Mexico are removed including prescribed fires
	where possible.**
ZANTH	All anthropogenic emissions are removed including prescribed fires.**
STRAT	Tracer species for O <sub>3</sub> injected into the upper troposphere/lower stratosphere based on CMAQ
	potential vorticity parameterization for stratospheric O <sub>3</sub> .***

\* Emissions estimated to be associated with intentionally set fires ("prescribed fires") are grouped with anthropogenic fires. \*\* Only for PA simulations

\*\*\* Only for EQUATES simulations.

Table S2. Expressions used to calculate contributions from specific sources for Policy Assessment simulations described in Table S1. Table adapted from 2020 O<sub>3</sub> Policy Assessment Table 2-2 (USEPA, 2020).

Label	Name	Description	Expression
BASE	Total	Total Concentration	BASE
USB	USB	US Background	ZUSA
USA	USA	US Anthropogenic	BASE – ZUSA
INTL	International	Rest of the World	BASE – ZROW
		Contribution	
CANMEX	Canada & Mexico	Canada & Mexico	BASE – ZCANMEX
		Contribution	
LINTL	Long-range international	Contribution from	INTL – CANMEX
		countries other than the	
		US, Canada, and Mexico	
NAT	Natural	Natural Contribution	ZANTH
RES-ANTH	Residual anthropogenic	Anthropogenic	BASE – ZANTH – INTL
		contribution that is not	- USA
		attributed directly to	= BASE $-$ ZANTH $-$
		either the US or	(BASE – ZROW) –
		International due to non-	(BASE – ZUSA)
		linear chemistry	= ZROW + ZUSA -
			BASE – ZANTH

Table S3. Expressions used to calculate contributions from specific sources for EQUATES simulations described in Table S1.

Label	Name	Description	Expression
BASE	Total	Total Concentration	BASE
USB	USB	US Background	ZUSA
STRAT	Stratospheric	Stratospheric O <sub>3</sub> estimate	STRAT
		from potential vorticity	
		tracer species	

USB_NOSTRAT	USB non-stratospheric	Estimate of USB O <sub>3</sub> from	ZUSA – STRAT
		sources other than	
		stratospheric O <sub>3</sub>	

Table S4. Summary of emissions used for CTM simulations.

	PA continental	PA H-CMAQ	EQUATES continental	EQUATES H-CMAO
US anthropogenic	2016 emissions modeling platform (2016fe) (USEPA, 2019a)	2016fe	Foley et al. (2023)	Foley et al. (2023)
non-US (except Canada and Mexico)	from lateral boundary conditions	EDGAR-HTAP* projected to 2014 (USEPA, 2019b) China: Tsinghua University (Zhao et al., 2018) rest of Asia: MIXv1 (Li et al., 2017)	from lateral boundary conditions	EDGAR-HTAP projected to 2014 China: Tsinghua University
Canada and Mexico	2016fe	2016fe	Canada: Air Pollutant Emission Inventory by Environment and Climate Change Canada Mexico: Inventory from Mexico's Secretariat of Environment and Natural Resources (SEMARNAT)	Canada: Air Pollutant Emission Inventory by Environment and Climate Change Canada Mexico: Inventory from Mexico's Secretariat of Environment and Natural Resources (SEMARNAT)
Lightning	None	GEIA**	CMAQ inline module (Kang et al., 2019)	GEIA
Biogenics	Biogenic Emission Inventory System (BEIS)	Model of Emissions of Gases and Aerosols from Nature (MEGAN), except BEIS over North America	BEIS	Hourly CAMS biogenic VOCs v2.2 data (Sindelarova et al., 2014); extension of MEGAN2.1
Soil NOx	BEIS	MEGAN, except BEIS over North America	BEIS	Hourly CAMS soil NO v2.1 data
Wildfires	2016fe	FINNv1.5 (Wiedinmyer et al., 2011), except 2016fe over North America	SMARTFIRE2 + Bluesky	FINNv1.5; SMARTFIRE2 + Bluesky within North America
Methane	set to constant value in CMAQ (1850 ppb)	set to constant value in CMAQ (1850 ppb)	set to constant value in CMAQ (1850 ppb)	set to constant value in CMAQ (1850 ppb)
Stratospheric O <sub>3</sub>	from LBCs, otherwise none	potential vorticity parameterization in CMAQ (Xing et al., 2016; Mathur et al., 2017)	from LBCs, otherwise none	potential vorticity parameterization in CMAQ

\* https://edgar.jrc.ec.europa.eu/htap\_v2/ \*\* http://www.geiacenter.org/

Table S5.	Summary	of model	configurations	for (	СТМ	simulations.

	PA continental	PA H-CMAQ	EQUATES continental	EQUATES H-CMAQ
CMAQ model version	5.2.1	5.2.1	5.3.2	5.3.2
Chemical mechanism	cb6r3_ae6nvPOA_aq	cb6r3_ae6_aq	cb6r3_ae7_aq	cb6r3m_ae7_kmtbr
Lateral boundary conditions	nested from H-CMAQ to 36 km CMAQ to 12 km CMAQ	clean conditions at equator	Nested from H-CMAQ	clean conditions at equator
Meteorology model version	WRF v3.8	WRF v3.8	WRF v4.1.1	WRF v4.1.1

# Seasonal average O<sub>3</sub> concentrations



Figure S1. Seasonal average O<sub>3</sub> from Policy Assessment CMAQ simulations. Results are shown for 36 km horizontal resolution for winter (DJF), spring (MAM), summer (JJA), and fall (SON). O<sub>3</sub> concentrations include total (BASE) O<sub>3</sub> as well as O<sub>3</sub> components from USA, NAT, LINTL, and CANMEX sources.



Figure S2. Seasonal average O<sub>3</sub> from Policy Assessment CMAQ simulations. Results are shown for 108 km horizontal resolution for winter (DJF), spring (MAM), summer (JJA), and fall (SON). O<sub>3</sub> concentrations include total (BASE) O<sub>3</sub> as well as O<sub>3</sub> components from USA, NAT, LINTL, and CANMEX sources.



Figure S3. Seasonal average O<sub>3</sub> from EQUATES CMAQ simulations. Results are shown for 12 km horizontal resolution for winter (DJF), spring (MAM), summer (JJA), and fall (SON). O<sub>3</sub> concentrations include total (BASE) O<sub>3</sub> as well as O<sub>3</sub> components from USA and USB sources.



Figure S4. Seasonal average O<sub>3</sub> from EQUATES CMAQ simulations. Results are shown for 108 km horizontal resolution for winter (DJF), spring (MAM), summer (JJA), and fall (SON). O<sub>3</sub> concentrations include total (BASE) O<sub>3</sub> as well as O<sub>3</sub> components from USA and USB sources.

## **Regression modelling supplemental information**

The regression variables are normalized to zero mean and unit standard deviation. The means and standard deviations for the 2016, 2017, and combined 2016-2017 observations are provided below.

		mea	n	s	tandard (	deviation
variable	2016	2017	2016-2017	2016	2017	2016-2017
lon	-95.4	-95.0	-95.2	16.0	15.7	15.8
lat	37.5	37.7	37.6	4.80	4.73	4.76
z	401	402	402	566	571	569
sin(d)	-0.017	0.016	0.000	0.718	0.725	0.722
cos(d)	-0.142	-0.128	-0.135	0.681	0.676	0.679

## Table S6. Regression variable means and standard deviations.

In the cross-validation summary tables, spatial and temporal withholding refers to randomly assigning 10% of data to the test set, spatial withholding refers to assigning data from 10% of randomly chosen observation sites to the test set, and temporal withholding refers to assigning data from 10% of randomly chosen days of the year to the test set.  $O_3$  split refers to the  $O_3$  components included in each regression model. The BASE  $O_3$  simulation performance is also provided for comparison to the results of the regression models.

modelling case	O3 split	BASE Simulation RMSE (ppb)	training RMSE spatial and temporal withholding (ppb)	test RMSE spatial and temporal withholding (ppb)	training RMSE spatial withholding (ppb)	test RMSE spatial withholding (ppb)	training RMSE temporal withholding (ppb)	test RMSE temporal withholding (ppb)
	USA + USB		7.25	7.25	7.25	7.22	7.25	7.28
EQUATES 12 km	USA + USB_NOSTRAT + STRAT	8.09	7.12	7.13	7.12	7.14	7.11	7.2
EQUATES 108 km	USA + USB	9.29	8.33	8.34	8.33	8.40	8.35	8.24
	USA + USB		7.04	7.10	7.07	6.79	7.04	7.04
PA 12 km	USA + NAT + INTL	8.18	7.14	7.18	7.17	6.86	7.14	7.17
	USA + NAT + LINTL + CANMEX		7.09	7.13	7.12	6.82	7.09	7.09
	USA + USB		7.96	7.97	8.01	7.47	7.97	7.89
PA 36 km	USA + NAT + INTL	10.04	7.98	7.98	8.02	7.55	7.98	7.93
	USA + NAT + LINTL + CANMEX		7.89	7.89	7.93	7.52	7.9	7.87
	USA + USB		8.67	8.69	8.71	8.33	8.68	8.63
PA 108 km	USA + NAT + INTL	12.05	8.65	8.69	8.68	8.45	8.66	8.64
	USA + NAT + LINTL + CANMEX		8.52	8.56	8.54	8.42	8.54	8.47
Average	n/a	9.53	7.80	7.83	7.83	7.58	7.81	7.79

Table S7. Summary of linear regression model cross-validation root mean square error (RMSE).

modelling		BASE Simulation MB	training MB random split	test MB random split	training MB site split	test MB site split	training MB time split	test MB time split
case	O3 split	(ppb)	(ppb)	(ppb)	(ppb)	(ppb)	(ppb)	(ppb)
	USA + USB		-0.08	-0.07	-0.07	-0.4	-0.08	0.4
EQUATES 12 km	USA + USB_NOSTRAT + STRAT	-1.83	-0.12	-0.12	-0.11	-0.12	-0.12	0.38
EQUATES 108 km	USA + USB	0.66	-0.1	-0.07	-0.1	-0.28	-0.1	0.31
	USA + USB		-0.09	-0.1	-0.09	-0.55	-0.09	0.54
PA 12 km	USA + NAT + INTL	0.49	-0.16	-0.15	-0.16	-0.62	-0.16	0.47
TA 12 KII	USA + NAT + LINTL + CANMEX	0.49	-0.15	-0.14	-0.15	-0.62	-0.15	0.52
	USA + USB		-0.24	-0.28	-0.25	-0.74	-0.24	0.31
PA 36 km	USA + NAT + INTL	2.16	-0.29	-0.31	-0.29	-0.83	-0.29	0.23
	USA + NAT + LINTL + CANMEX	2	-0.26	-0.28	-0.26	-0.79	-0.26	0.31
	USA + USB		-0.26	-0.33	-0.26	-0.83	-0.26	0.38
PA 108 km	USA + NAT + INTL	4.16	-0.26	-0.31	-0.26	-0.9	-0.26	0.33
	USA + NAT + LINTL + CANMEX		-0.23	-0.28	-0.22	-0.86	-0.23	0.39
Average	n/a	1.13	-0.19	-0.20	-0.19	-0.63	-0.19	0.38

Table S8. Summary of linear regression model cross-validation mean biases (MB).

	EQUATES 12 km	EQUATES 108 km	PA 12 km	PA 36 km	PA 108 km
$\alpha_{0,USA}$	$1.093 \pm 0.0021$	$0.951 \pm 0.0026$	$0.86\pm0.0014$	$0.762\pm0.0016$	$0.658\pm0.0017$
$\alpha_{x,USA}$	$-0.119 \pm 0.0015$	$-0.108 \pm 0.0023$	$-0.054 \pm 0.0011$	$-0.061 \pm 0.0011$	$-0.037 \pm 0.0013$
$\alpha_{y,USA}$	$0.075 \pm 0.0016$	$0.006\pm0.002$	$-0.006 \pm 0.0011$	$-0.028 \pm 0.0011$	$0.005\pm0.001$
A <sub>e</sub>	$0.01 \pm 0.0023$	$0.064 \pm 0.0028$	$0.044 \pm 0.0016$	$0.078\pm0.0016$	$0.141 \pm 0.002$
$\alpha_{sin,USA}$	$0.094 \pm 0.0017$	$0.109\pm0.002$	$0.024 \pm 0.0011$	$0.018\pm0.0011$	$-0.016 \pm 0.0012$
$\alpha_{\cos,USA}$	$0.085 \pm 0.0018$	$0.184\pm0.0022$	$0.005 \pm 0.0012$	$0.043 \pm 0.0013$	$0.074 \pm 0.0014$
$\alpha_{0,USB}$	$1.05 \pm 0.0006$	$1.027\pm0.0008$	$1.053 \pm 0.0007$	$1.062 \pm 0.0008$	$1.061 \pm 0.0008$
$\alpha_{x,USB}$	$-0.02 \pm 0.0006$	$-0.008 \pm 0.0007$	$0.008 \pm 0.0006$	$0.029 \pm 0.0007$	$0.02\pm0.0007$
$\alpha_{y,USB}$	$-0.016 \pm 0.0005$	$-0.01 \pm 0.0006$	$0.022\pm0.0006$	$0.016\pm0.0007$	$0.009\pm0.0007$
$\alpha_{z,USB}$	$0.002 \pm 0.0005$	$-0.001 \pm 0.0007$	$0.005 \pm 0.0006$	$0.004 \pm 0.0006$	$-0.014 \pm 0.0007$
$\alpha_{sin,USB}$	$0.044 \pm 0.0006$	$0.036\pm0.0007$	$0.078\pm0.0006$	$0.078\pm0.0007$	$0.089\pm0.0007$
$\alpha_{\cos,USB}$	$0.001 \pm 0.0005$	$-0.041 \pm 0.0006$	$0.028\pm0.0006$	$0.001 \pm 0.0006$	$-0.016 \pm 0.0007$

Table S9. Regression model coefficients and standard errors for USA + USB formulation models.

Table S10. Regression model coefficients and standard errors for USA + NAT + INTL formulation models.

	PA 12 km	PA 36 km	PA 108 km
$\alpha_{0,USA}$	$0.943 \pm 0.0016$	$0.835 \pm 0.0018$	$0.74\pm0.002$
$\alpha_{x,USA}$	$-0.028 \pm 0.0012$	$-0.031 \pm 0.0013$	$-0.051 \pm 0.0014$
$\alpha_{y,USA}$	$0.024 \pm 0.0012$	$-0.032 \pm 0.0012$	$0.046 \pm 0.0012$
$\alpha_{z,USA}$	$0.077 \pm 0.0017$	$0.134 \pm 0.0018$	$0.178\pm0.0022$
$\alpha_{sin,USA}$	$0.066 \pm 0.0013$	$0.066 \pm 0.0013$	$0.026\pm0.0015$
$\alpha_{\cos,USA}$	$-0.014 \pm 0.0014$	$0.062 \pm 0.0015$	$0.118 \pm 0.0017$
$\alpha_{0,NAT}$	$1.065 \pm 0.0022$	$1.107 \pm 0.0025$	$1.1 \pm 0.0027$
$\alpha_{x,NAT}$	$-0.044 \pm 0.0019$	$-0.012 \pm 0.002$	$0.051 \pm 0.0021$
$\alpha_{y,NAT}$	$-0.067 \pm 0.0019$	$-0.022 \pm 0.002$	$-0.102 \pm 0.002$
$\alpha_{z,NAT}$	$-0.041 \pm 0.0019$	$-0.104 \pm 0.0021$	$-0.085 \pm 0.0021$
$\alpha_{sin,NAT}$	$0.009 \pm 0.002$	$-0.01 \pm 0.0022$	$0.06 \pm 0.0022$
$\alpha_{\cos,NAT}$	$0.103 \pm 0.0022$	$-0.016 \pm 0.0026$	$-0.071 \pm 0.0027$
$\alpha_{0,INTL}$	$1.332 \pm 0.0051$	$1.248 \pm 0.0056$	$1.238 \pm 0.0063$
$\alpha_{x,INTL}$	$0.15 \pm 0.004$	$0.123 \pm 0.0041$	$-0.014 \pm 0.0045$
$\alpha_{y,INTL}$	$0.197 \pm 0.0038$	$0.114 \pm 0.0037$	$0.243 \pm 0.0043$
$\alpha_{z,INTL}$	$0.09 \pm 0.0042$	$0.203 \pm 0.0045$	$0.141 \pm 0.0047$
$\alpha_{sin,INTL}$	$0.154 \pm 0.0043$	$0.205 \pm 0.0046$	$0.069 \pm 0.005$
$\alpha_{cos,INTL}$	$-0.146 \pm 0.0049$	$0.005 \pm 0.0055$	$0.074 \pm 0.0059$

	PA 12 km	PA 36 km	PA 108 km
$\alpha_{0,\rm USA}$	$0.951 \pm 0.0016$	$0.859 \pm 0.0018$	$0.771 \pm 0.002$
$\alpha_{x,USA}$	$-0.034 \pm 0.0012$	$-0.046 \pm 0.0013$	$-0.054 \pm 0.0014$
$\alpha_{y,USA}$	$0.033 \pm 0.0012$	$-0.008 \pm 0.0012$	$0.055 \pm 0.0012$
$\alpha_{z,USA}$	$0.066 \pm 0.0018$	$0.12\pm0.0018$	$0.187 \pm 0.0022$
$\alpha_{sin,USA}$	$0.063 \pm 0.0013$	$0.062 \pm 0.0013$	$0.009 \pm 0.0014$
$\alpha_{cos,USA}$	$-0.004 \pm 0.0014$	$0.085 \pm 0.0016$	$0.143\pm0.0018$
$\alpha_{0,NAT}$	$1.037 \pm 0.0023$	$1.047 \pm 0.0027$	$1.006 \pm 0.003$
$\alpha_{x,NAT}$	$-0.043 \pm 0.002$	$0.014 \pm 0.0021$	$0.056 \pm 0.0021$
$\alpha_{y,NAT}$	$-0.073 \pm 0.0019$	$-0.065 \pm 0.0021$	$-0.087 \pm 0.002$
$\alpha_{z,NAT}$	$-0.03 \pm 0.002$	$-0.082 \pm 0.0022$	$-0.1 \pm 0.0021$
$\alpha_{sin,NAT}$	$0.013\pm0.002$	$0.006 \pm 0.0022$	$0.083 \pm 0.0022$
$\alpha_{\cos,NAT}$	$0.082 \pm 0.0023$	$-0.056 \pm 0.0027$	$-0.135 \pm 0.0029$
$\alpha_{0,LINTL}$	$1.54\pm0.0068$	$1.601 \pm 0.0077$	$1.822 \pm 0.0085$
$\alpha_{x,LINTL}$	$0.192 \pm 0.0046$	$0.121 \pm 0.005$	$0.095 \pm 0.005$
$\alpha_{y,LINTL}$	$0.224 \pm 0.0047$	$0.264 \pm 0.0051$	$0.151 \pm 0.0052$
$\alpha_{z,LINTL}$	$0.017 \pm 0.0047$	$0.104 \pm 0.0053$	$0.15 \pm 0.0049$
$\alpha_{sin,LINTL}$	$0.148 \pm 0.0052$	$0.117 \pm 0.0058$	$-0.102 \pm 0.0059$
$\alpha_{cos,LINTL}$	$-0.095 \pm 0.0059$	$0.063 \pm 0.0066$	$0.104 \pm 0.0068$
$\alpha_{0,CANMEX}$	$0.943 \pm 0.0079$	$0.803 \pm 0.0081$	$0.667 \pm 0.009$
$\alpha_{x,\;CANMEX}$	$0.191 \pm 0.0079$	$0.135 \pm 0.0068$	$-0.143 \pm 0.0098$
$\alpha_{y, \text{ CANMEX}}$	$0.117 \pm 0.0063$	$0.004 \pm 0.0052$	$0.173 \pm 0.0075$
$\alpha_{z, \text{ CANMEX}}$	$0.295 \pm 0.0071$	$0.352 \pm 0.0071$	$0.248 \pm 0.0085$
$\alpha_{sin, CANMEX}$	$0.007 \pm 0.0075$	$0.056 \pm 0.0074$	$0.021 \pm 0.0082$
$\alpha_{cos, CANMEX}$	$-0.327 \pm 0.0077$	$-0.174 \pm 0.008$	$0.094 \pm 0.0085$

Table S11. Regression model coefficients and standard errors for USA + NAT + LINTL + CANMEX formulation models.

	EQUATES 12 km
$\alpha_{0,\rm USA}$	$1.088 \pm 0.0015$
$\alpha_{x,USA}$	$-0.1 \pm 0.0011$
$\alpha_{y,USA}$	$0.043 \pm 0.0011$
$\alpha_{z,USA}$	$0.006 \pm 0.0016$
$\alpha_{sin,USA}$	$0.066 \pm 0.0011$
$\alpha_{\cos,USA}$	$0.062 \pm 0.0013$
$\alpha_{0,USB\_NOSTRAT}$	$1.058 \pm 0.0017$
$\alpha_{x,USB\_NOSTRAT}$	$0.097 \pm 0.0012$
$\alpha_{y,USB\_NOSTRAT}$	$-0.011 \pm 0.001$
$\alpha_{z,USB\_NOSTRAT}$	$-0.001 \pm 0.0013$
$\alpha_{sin,USB_NOSTRAT}$	$0.028 \pm 0.0012$
$\alpha_{\cos,USB_NOSTRAT}$	$-0.116 \pm 0.0015$
$\alpha_{0,STRAT}$	$1.038 \pm 0.0022$
$\alpha_{x, STRAT}$	$-0.167 \pm 0.0015$
$\alpha_{y, STRAT}$	$-0.035 \pm 0.0013$
$\alpha_{z, STRAT}$	$0.012 \pm 0.0015$
$\alpha_{ m sin, STRAT}$	$0.074 \pm 0.0016$
$\alpha_{\cos, STRAT}$	$0.154 \pm 0.0019$

 Table S12. Regression model coefficients and standard errors for USA + USB\_NOSTRAT + NOSTRAT formulation model.

### **Empirical orthogonal function analysis**

The inferred CMAQ bias fields are further analyzed by performing an empirical orthogonal function (EOF) analysis to explore the spatial and temporal variability of the inferred bias. The EOF analysis is performed using the eofs Python package (Dawson, 2016). EOFs and principal components (PCs) represent the inferred bias time series as follows:

$$f(t, x, y) = \sum_{k} P_{k}(t) \times E_{k}(x, y)$$

Where:

f is the inferred bias timeseries

k is the number of orthogonal basis functions

P are the PCs that represent how the EOFs vary in time

E are the EOFs that show the spatial structure of the influences on the temporal variability of f

The EOFs are scaled by multiplying by the square root of the corresponding eigenvectors. The PCs are scaled by dividing by the square root of the corresponding eigenvectors (which is equivalent to scaling the PCs to unit variance). The leading EOF of each of the inferred bias components are shown in Figures S5 - S6. Results are shown here for the 12 km horizontal resolution Policy Assessment (PA) and EQUATES simulations. Note that the data is normalized to zero mean along the time axis before calculating the EOFs and time series. The EOFs and PCs then represent the variation from the average bias for each component.

In both simulation cases, the leading EOF of BASE  $O_3$  bias is positive and is higher in the eastern US. The corresponding PCs are also similar, showing a seasonal pattern with negative values in the winter and spring and positive values in the summer and fall. The leading EOFs of the USA  $O_3$  bias are also similar in the two cases, with the highest values in the most populated areas. The PCs are also similar with positive values in the summer and fall and slightly negative values during other times. In general, for BASE  $O_3$  and each of the components, the PC of the leading EOF follows the same temporal pattern as the temporal trends of the bias shown in Figure 6 if the EOF is mostly positive and the inverse of the temporal trend of the bias if the EOF is mostly negative.

The information that can be obtained from an EOF analysis of a single year (or two years for the EQUATES data) is limited. Longer timeseries are needed to uncover the structure of variability within the data. The full EQUATES dataset from 2002 - 2019 for total (i.e., BASE) O<sub>3</sub> may provide some opportunity to explore this further in the future.



Figure S5. Leading EOF and PC time series for inferred bias of BASE O<sub>3</sub> and each O<sub>3</sub> component for Policy Assessment (PA) 12 km simulations. The number in parenthesis is the percent of variance explained by the leading EOF.



Figure S6. Leading EOF and PC time series for inferred bias of BASE O<sub>3</sub> and each O<sub>3</sub> component for EQUATES 12 km simulations. The number in parenthesis is the percent of variance explained by the leading EOF.

#### CTM biases by model resolution

As mentioned in previous sections, the inferred CTM bias in USA O<sub>3</sub> tends to increase both with coarser model resolution and with increasing BASE  $O_3$ . The effects of this finding at  $O_3$  monitoring site locations are shown in Figure 9. Here, total O<sub>3</sub> is split only into the most basic two components (USA and USB) for simplicity. The inferred bias in USB O3 is consistent across model resolutions and BASE O3 concentrations. USB O3 bias is also consistent between the PA and EQUATES simulations. USA  $O_3$  for the PA simulations is typically biased high. Across all three model resolutions (12, 36, and 108 km), the inferred bias increases with higher BASE  $O_3$ . The bias increases at all  $O_3$  concentration bins when going from 12 km to 36 km and from 36 km to 108 km. Both the typical biases (as indicated by the median) and the more extreme biases (as indicated by the 5<sup>th</sup> and 95<sup>th</sup> percentiles) increase with coarser model resolution. The EQUATES simulations have lower inferred biases in USA O<sub>3</sub> compared to the corresponding PA simulations of the same model resolution. For the 12 km resolution EQUATES simulation, the USA  $O_3$  biases do not change much at different  $O_3$  concentration bins. The 108 km EQUATES simulation has similar behavior to the PA simulations that the USA O<sub>3</sub> bias gets larger with increasing BASE O<sub>3</sub>. The increasingly high bias with coarser model resolution is likely due to over-dilution of  $NO_x$  in the coarser resolution simulations (e.g., Li et al. (2023)). This can result in NO<sub>x</sub> that is more localized with finer model resolution being spread out across a larger area in the coarser model resolutions and enhancing  $O_3$  production in areas that should in reality have less NO<sub>x</sub> as well as reducing the effect of NO<sub>x</sub> titration in high NO<sub>x</sub> areas.



Figure S7. Inferred biases of USA and USB separated by simulated O<sub>3</sub> concentration at O<sub>3</sub> monitoring sites. Results are shown for the PA (top row) and EQUATES (bottom row) simulations for all available model resolutions. The line shows the median; the box shows the 25th-75th percentiles; the whiskers show the 5th and 95th percentiles.

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