## **Supplemental Material**

## Using Regionalized Air Quality Model Performance and Bayesian Maximum Entropy data fusion to map global surface ozone concentration

Jacob S. Becker<sup>1</sup>, Marissa N. DeLang<sup>1</sup>, Kai-Lan Chang<sup>2,3</sup>, Marc L. Serre<sup>1</sup>, Owen R. Cooper<sup>2,3</sup>,

Hantao Wang<sup>1</sup>, Martin G. Schultz<sup>4</sup>, Sabine Schröder<sup>4</sup>, Xiao Lu<sup>5</sup>, Lin Zhang<sup>6</sup>, Makoto Deushi<sup>7</sup>,

Beatrice Josse<sup>8</sup>, Christoph A. Keller<sup>9,10</sup>, Jean-François Lamarque<sup>11</sup>, Meiyun Lin<sup>12,13</sup>, Junhua

Liu<sup>9,10</sup>, Virginie Marécal<sup>8</sup>, Sarah A. Strode<sup>9,10</sup>, Kengo Sudo<sup>14,15</sup>, Simone Tilmes<sup>11</sup>, Li Zhang<sup>12,13,16</sup>, Michael Brauer<sup>17,18</sup>, J. Jason West<sup>\*,1</sup>

<sup>1</sup>Department of Environmental Sciences and Engineering, University of North Carolina at Chapel Hill, Chapel Hill, NC, USA

<sup>2</sup>Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, USA

<sup>3</sup>NOAA Chemical Sciences Laboratory, Boulder, CO, USA

<sup>4</sup>Jülich Supercomputing Centre (JSC), Forschungszentrum Jülich, Jülich, Germany

<sup>5</sup>School of Atmospheric Sciences, Sun Yat-Sen University, Zhuhai, Guangdong, China

<sup>6</sup>Laboratory for Climate and Ocean-Atmosphere Studies, Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing, China

<sup>7</sup>Meteorological Research Institute (MRI), Tsukuba, Japan

<sup>8</sup>Centre National de Recherches Météorologiques, Université de Toulouse, Météo-France, CNRS, Toulouse, France

<sup>9</sup>NASA Goddard Space Flight Center, Greenbelt, MD, USA

<sup>10</sup>Universities Space Research Association, Columbia, MD, USA

<sup>11</sup>National Center for Atmospheric Research, Boulder, CO, USA

<sup>12</sup>NOAA Geophysical Fluid Dynamics Laboratory, Princeton, NJ, USA

<sup>13</sup>Program in Atmospheric and Oceanic Sciences, Princeton University, Princeton, NJ, USA

<sup>14</sup>Graduate School of Environmental Studies, Nagoya University, Nagoya, Japan

<sup>15</sup>Japan Agency for Marine-Earth Science and Technology (JAMSTEC), Yokosuka, Japan

<sup>16</sup>Department of Meteorology and Atmospheric Science, Pennsylvania State University, University Park, PA. USA

<sup>17</sup>Institute for Health Metrics and Evaluation, University of Washington, Seattle, Washington, USA

<sup>18</sup>School of Population and Public Health, University of British Columbia, Vancouver, British Columbia, Canada

\* Corresponding Author: jasonwest@unc.edu

September 25, 2023

List of Contents:

**Text S1. Covariance description in Bayesian Maximum Entropy.** (page 4)

**Figure S1. Complete annual results for each year from 1990 to 2017.** Shown for each year are observations, estimates from the M<sup>3</sup>Fusion composite, CAMP, RAMP, and wRAMP, and BME estimates with the M<sup>3</sup>Fusion composite, CAMP, and wRAMP (our final estimates) as the global offset. (page 5)

Figure S2. CAMP analysis for all years. (page 15)

**Figure S3. Space/time covariance equations and graphs**. a) is based on the RAMP corrected model, while b) is based solely on M<sup>3</sup>Fusion. Both are used in BME estimation based on RAMP weight at a given location. (page 16)

**Figure S4. The BME variance each year under different configurations.** Shown are the BME variance when the M<sup>3</sup>Fusion composite, CAMP, and wRAMP are used as global offsets. (page 17)

**Figure S5. The likelilhood of exceedance of OSDMA8 at selected levels.** Results are shown for each year 1990 to 2017, based on BME with wRAMP as the global offset. (page 22)

**Figure S6. Ozone trends by region**. Population weighted ozone rose from 1990 in Asia, which drove an overall global rise. North America had a clear downward trend while other regions slightly fluctuated but overall have no clear trend. (page 27)

2

**Table S1. Differences in ozone estimates caused by including RAMP.** Differences between ozone estimates are shown in the final case where weighted RAMP is used before BME data fusion, minus results without using RAMP, in ppb (the results of DeLang et al., 2021). Results are shown for 2000 and 2017, for average difference and the absolute value of differences, for area-weighted and population-weighted results, and for the mean, median, and 5<sup>th</sup> / 95<sup>th</sup> percentiles. A positive value indicates that results using weighted RAMP are higher. (page 28)

**Table S2. Leave one out cross validation results.** Results show that using RAMP decreases the mean square error (MSE) and increases  $R^2$  over the M<sup>3</sup>Fusion multi-model composite. When BME is used, the MSE and  $R^2$  do not differ when using M<sup>3</sup>Fusion as the global offset, or M<sup>3</sup>Fusion bias-corrected using CAMP and weighted RAMP (wRAMP), which is expected because BME matches observations exactly at measurement stations, and many stations are clustered near one another. (page 30)

## Text S1. Covariance description in Bayesian Maximum Entropy

The general knowledge  $Y(\mathbf{p})$  includes the mean function  $m_x(\mathbf{p}) = E[X]$ , which is assumed to be zero due to the removal of the offset, and the covariance function  $c_x(\mathbf{p},\mathbf{p}') = E[X(\mathbf{p})-m(\mathbf{p}))(X(\mathbf{p}')-m(\mathbf{p}))(X(\mathbf{p}'))$ , which uses experimentally determined covariance of  $\mathbf{r}_h$ .

$$\hat{q}(r,\tau) \approx \frac{1}{N(x,\tau)} \sum_{i=1}^{N(x,\tau)} r_{head,i} r_{tail,i} - m_X^2$$

 $N(x, \tau)$  is the number of pairs of points with values ( $r_{head}$ ,  $r_{tail}$ ) separated by spatial distance x and temporal distance  $\tau$ , while  $m_x$  is the mean of  $r_0$ . We use an exponential covariance model for S/TRF Y(p) to fit:

$$c_X(r,\tau) = C \left[ \gamma \exp \frac{-3r}{a_{r1}} \exp \frac{-3\tau}{a_{t1}} + \lambda \exp \frac{-3r}{a_{r2}} \exp \frac{-3\tau}{a_{t2}} + (1 - \gamma - \lambda) \exp \frac{-3r}{a_{r3}} \exp \frac{-3\tau}{a_{t3}} \right]$$

Areas with a weight  $w(\mathbf{p}) \ge 0.2$  used the RAMP derived covariance model (Figure S3a), while areas with  $w(\mathbf{p}) < 0.2$  used the M<sup>3</sup>Fusion derived covariance model, to insure a larger spatial range of influence for observations in areas without substantial RAMP correction (Figure S3b). **Figure S1. Complete annual results for each year from 1990 to 2017.** Shown for each year are observations, estimates from the M<sup>3</sup>Fusion composite, CAMP, RAMP, and wRAMP, and BME estimates with the M<sup>3</sup>Fusion composite, CAMP, and wRAMP (our final estimates) as the global offset.























Figure S2. CAMP analysis for all years.



**Figure S3. Space/time covariance equations and graphs**. a) is based on the RAMP corrected model, while b) is based solely on M<sup>3</sup>Fusion. Both are used in BME estimation based on RAMP weight at a given location.

**Figure S4. The BME variance each year under different configurations.** Shown are the BME variance when the M<sup>3</sup>Fusion composite, CAMP, and wRAMP are used as global offsets.











**Figure S5. The likelilhood of exceedance of OSDMA8 at selected levels.** Results are shown for each year 1990 to 2017, based on BME with wRAMP as the global offset.















**Figure S6. Ozone trends by region**. Population weighted ozone rose from 1990 in Asia, which drove an overall global rise. North America had a clear downward trend while other regions slightly fluctuated but overall have no clear trend.

**Table S1. Differences in ozone estimates caused by including RAMP.** Differences between ozone estimates are shown in the final case where weighted RAMP is used before BME data fusion, minus results without using RAMP, in ppb (the results of DeLang et al., 2021). Results are shown for 2000 and 2017, for average difference and the absolute value of differences, for area-weighted and population-weighted results, and for the mean, median, and 5<sup>th</sup> / 95<sup>th</sup> percentiles. A positive value indicates that results using weighted RAMP are higher.

## **Year 2000**

Region	Difference			Absolute Difference			
	5%ile	Mean	Median	95%ile	Mean	Median	95%ile
Africa	-0.018	0.004	0.001	0.048	0.012	0.002	0.061
East Asia	-0.029	0.199	0.036	0.869	0.233	0.051	0.908
Europe	-2.425	-0.273	-0.105	1.114	0.613	0.190	2.704
North America	-0.118	0.032	0.015	0.363	0.192	0.030	1.066
Oceania	-0.008	-0.000	-0.001	0.020	0.005	0.003	0.020
Russia	-0.042	0.012	0.000	0.131	0.026	0.003	0.141
South Central Asia	-0.018	0.006	0.007	0.026	0.012	0.008	0.035
South America	-0.007	0.012	0.006	0.057	0.014	0.007	0.057

Region	Population Weighted Difference			Population Weighted Absolute Difference			
	5%ile	Mean	Median	95%ile	Mean	Median	95%ile
Africa	-0.009	-0.000	0.000	0.009	0.010	0.001	0.050
East Asia	-0.163	0.294	0.078	0.913	0.337	0.166	0.934
Europe	-2.630	-0.205	-0.102	2.148	0.863	0.304	3.038
North America	-0.410	0.258	0.018	2.151	0.446	0.058	2.320
Oceania	-0.003	0.001	-0.000	0.017	0.002	0.001	0.017
Russia	-0.148	-0.033	-0.019	0.029	0.050	0.032	0.157

South Central Asia	-0.005	0.008	0.008	0.021	0.012	0.009	0.036
South America	-0.014	0.022	0.010	0.120	0.025	0.013	0.120

Year 2017

Region	Difference			Absolute Difference			
	5%ile	Mean	Median	95%ile	Mean	Median	95%ile
Africa	-0.642	-0.102	-0.000	0.018	0.114	0.013	0.652
East Asia	-1.868	0.140	0.043	2.776	0.855	0.420	3.057
Europe	-1.350	0.002	-0.005	1.469	0.488	0.192	2.024
North America	-0.088	0.013	0.007	0.118	0.042	0.014	0.176
Oceania	-0.002	0.011	0.001	0.063	0.012	0.001	0.062
Russia	-0.310	-0.044	-0.008	0.071	0.066	0.017	0.319
South Central Asia	-0.084	0.027	0.015	0.177	0.052	0.025	0.215
South America	-0.019	-0.001	0.002	0.011	0.008	0.003	0.020

Region	Population Weighted Difference			Population Weighted Absolute Difference			
	5%ile	Mean	Median	95%ile	Mean	Median	95%ile
Africa	-0.854	-0.101	0.001	0.017	0.111	0.007	0.854
East Asia	-1.916	0.217	0.148	2.560	0.897	0.563	2.736
Europe	-1.189	0.120	0.006	1.967	0.589	0.344	2.197
North America	-0.286	0.074	0.017	0.574	0.163	0.069	0.653
Oceania	0.000	0.033	0.024	0.091	0.033	0.024	0.091
Russia	-0.091	0.058	0.048	0.288	0.112	0.073	0.386
South Central Asia	-0.064	0.032	0.031	0.128	0.061	0.040	0.161
South America	-0.003	0.011	0.003	0.106	0.015	0.004	0.126

**Table S2. Leave one out cross validation results.** Results show that using RAMP decreases the mean square error (MSE) and increases  $R^2$  over the M<sup>3</sup>Fusion multi-model composite. When BME is used, the MSE and  $R^2$  do not differ when using M<sup>3</sup>Fusion as the global offset, or M<sup>3</sup>Fusion bias-corrected using CAMP and weighted RAMP (wRAMP), which is expected because BME matches observations exactly at measurement stations, and many stations are clustered near one another.

Scenario	MSE (ppb <sup>2</sup> )	$\mathbb{R}^2$
Simple multi-model mean	189.23	0.28
M <sup>3</sup> Fusion	61.14	0.30
CAMP	53.54	0.35
wRAMP	46.79	0.43
BME using M <sup>3</sup> Fusion as offset (DeLang et al., 2021)	14.5	0.83
BME using CAMP as offset	14.5	0.83
BME using wRAMP as offset	14.5	0.83