

# Intercomparison of global ground-level ozone datasets for health-relevant metrics

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## SUPPLEMENTARY INFORMATION

### Text S1. Grouping method used in pairwise spatial similarity comparison

The Grouping method is based on the pairwise correlation between each dataset. Initially, all datasets are re-gridded to a resolution of  $0.1^\circ$  by  $0.1^\circ$ . Subsequently, the pairwise correlation coefficient (R) is computed for each pair of datasets spanning the period from 2006 to 2016. The mean pairwise correlation for each grouping over the same timeframe is then calculated to construct Figure 5(a). Then, we generate a random groups table. For this comparison, involving 6 datasets, there are 203 possible combinations for grouping. For each grouping combination, 4 variables are computed: the sum of pairwise correlations within groups ( $C_i$ ), the sum of pairwise correlations outside the groups ( $C_o$ ), the number of dataset pairs within groups ( $N_i$ ), and the number of dataset pairs outside the groups ( $N_o$ ).

The objective is to ascertain the grouping combination that maximizes the difference between  $\frac{C_i}{N_i}$  and  $\frac{C_o}{N_o}$ .

For example, consider a grouping scenario where:

Group A: BME, NJML, UKML

Group B: CAMS, GEOS, TCR-2.

“pwcorr” denotes the operation to compute the pairwise correlation between two datasets. The calculations proceed as follows:

$C_i = \text{pwcorr}(\text{BME, NJML}) + \text{pwcorr}(\text{BME, UKML}) + \text{pwcorr}(\text{NJML, UKML}) + \text{pwcorr}(\text{CAMS, GEOS}) + \text{pwcorr}(\text{CAMS, TCR-2}) + \text{pwcorr}(\text{TCR-2, GEOS})$

$C_o = \text{pwcorr}(\text{BME, GEOS}) + \text{pwcorr}(\text{BME, TCR-2}) + \text{pwcorr}(\text{BME, CAMS}) + \text{pwcorr}(\text{NJML, GEOS}) + \text{pwcorr}(\text{NJML, TCR-2}) + \text{pwcorr}(\text{NJML, CAMS}) + \text{pwcorr}(\text{UKML, GEOS}) + \text{pwcorr}(\text{UKML, TCR-2}) + \text{pwcorr}(\text{UKML, CAMS})$

$N_i = 6$

$N_o = 9$

$\text{Difference} = \frac{C_i}{N_i} - \frac{C_o}{N_o}$

This analysis is conducted for all 203 combinations, identifying the optimal grouping that maximizes the difference.

Group A: BME, UKML, CAMS, GEOS, TCR-2

Group B: NJML

We also use the hierarchical clustering method to group 6 datasets based on pairwise correlation ( $R$ ) and get the same result. The hierarchical clustering is typically used with a dissimilarity (distance) matrix, where the elements represent the distances or dissimilarities between data (Bishop, 2006).

**Table S1. Inputs to the BME dataset include nine chemistry models and observations from TOAR-I and CNEMC. Table based on DeLang et al. (2021)**

| Model/Observation | Years     | Resolution                         | Experiment   | Reference                                    |
|-------------------|-----------|------------------------------------|--|--|
| TOAR-I            | 1990-2017 | 7269 monitoring stations           | Tropospheric Ozone Assessment Report                         | (Schultz et al., 2017)                       |
| CNEMC             | 2014-2017 | 1565 monitoring stations           | the Chinese National Environmental Monitoring Center Network | (Lu et al., 2018)                            |
| CESM1 CAM4-CHEM   | 1990-2010 | $1.9^{\circ} \times 2.5^{\circ}$   | CCMI REF-C1SD  | (Tilmes et al., 2015)                        |
| CESM1 WACCM       | 1990-2010 | $1.9^{\circ} \times 2.5^{\circ}$   | CCMI REF-C1SD  | (Marsh et al., 2013; Garcia et al., 2017)    |
| CHASER            | 1990-2010 | $2.8^{\circ} \times 2.8^{\circ}$   | CCMI REF-C1SD  | (Sudo et al., 2002; Watanabe et al., 2011)   |
| GFDL AM3          | 1990-2014 | $2^{\circ} \times 2.5^{\circ}$     | CCMI REF-C1SD  | (Lin et al., 2017)                           |
| GFDL AM4          | 2010-2016 | $1^{\circ} \times 1.25^{\circ}$    | CMIP6 nudged to NCEP wind                                    | (Zhang et al., 2020; Horowitz et al., 2020)  |
| MERRA2-GMI        | 1990-2017 | $0.5^{\circ} \times 0.625^{\circ}$ | MACCity and GFED-4s emissions                                | (Ziemke et al., 2019; Strode et al., 2019)   |
| MOCAGE            | 1990-2016 | $2^{\circ} \times 2^{\circ}$       | CCMI REF-C1SD  | (Josse et al., 2004; Teyssèdre et al., 2007) |
| MRI-ESM1r1        | 1990-2010 | $2.8^{\circ} \times 2.8^{\circ}$   | CCMI REF-C1SD  | (Adachi et al., 2013)                        |
| MRI-ESM2.0        | 2011-2017 | $2.8^{\circ} \times 2.8^{\circ}$   | CMIP6 historical and ssp370                                  | (Yukimoto et al., 2019c)                     |

**Table S2. Inputs to the NJML dataset include meteorology terms, chemical models, landcover and satellite observations. Ground observation from TOAR-I (2003–2019) are used as labels for training. Table based on Liu et al. (2022)**

| Variable  | Years     | Resolution   | Source | Reference               |
|---|-----------|--------------|--------|-------------------------|
| 10m wind speed                                  | 2003-2019 | 30 km/0.25 ° | ERA5   | (Hersbach et al., 2020) |
| 2m temperature                                  | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Boundary layer height                           | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Forecast surface roughness                      | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Mean sea level pressure                         | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Mean surface downward short-wave radiation flux | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Mean surface downward UV radiation flux         | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Total precipitation                             | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Soil temperature level 1                        | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Surface latent heat flux                        | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Surface sensible heat flux                      | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Total cloud cover                               | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Total column ozone                              | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Forecast albedo                                 | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Evaporation                                     | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| 2m dewpoint temperature                         | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Volumetric soil water layer 1                   | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| 2m temperature one month lag                    | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| 2m dewpoint temperature one month lag           | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Total precipitation one month lag               | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Mean sea level pressure one month lag           | 2003-2019 | 30 km/0.25 ° | ERA5   |                         |
| Total aerosol optical depth at 550 nm           | 2003-2019 | 80 km/0.75 ° | CAMS   | (Inness et al., 2019)   |
| Total column hydroxyl radical                   | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Total column methane                            | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Total column nitrogen dioxide                   | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Total column ozone/GEMS                         | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Total column ozone                              | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Particulate matter d < 2.5 µm (PM2.5)           | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Total column carbon monoxide                    | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Total column formaldehyde                       | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Total column nitrogen monoxide                  | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Carbon monoxide                                 | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Dust aerosol (0.55 - 0.9 µm) mixing ratio       | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Dust aerosol (0.03 - 0.55 µm) mixing ratio      | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Dust aerosol (0.9 - 20 µm) mixing ratio         | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Hydrophilic organic matter aerosol mixing ratio | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |
| Hydrophobic organic matter aerosol mixing ratio | 2003-2019 | 80 km/0.75 ° | CAMS   |                         |

|   |           |               |                   |   |
|---|-----------|---------------|-------------------|---|
| Nitrogen monoxide                             | 2003-2019 | 80 km/0.75 °  | CAMS              | Global Modeling and Assimilation Office (GMAO) (2015)   |
| SO2 precursor mixing ratio                    | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Sea salt aerosol (0.5 - 5 µm) mixing ratio    | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Sulphur dioxide                               | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Formaldehyde                                  | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Hydrophilic black carbon aerosol mixing ratio | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Hydrophobic black carbon aerosol mixing ratio | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Hydroxyl radical                              | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Methane (chemistry)                           | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Nitrogen dioxide                              | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Ozone/GEMS Ozone                              | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Sea salt aerosol (0.03 - 0.5 µm) mixing ratio | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Sea salt aerosol (5 - 20 µm) mixing ratio     | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Sulphate aerosol mixing ratio                 | 2003-2019 | 80 km/0.75 °  | CAMS              |   |
| Black Carbon Surface Mass Concentration       | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| Dust Surface Mass Concentration               | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| Organic Carbon Surface Mass Concentration     | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| Sea Salt Surface Mass Concentration           | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| SO4 Surface Mass Concentration                | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| SO2 Surface Mass Concentration                | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| Total Aerosol Extinction AOT [550 nm]         | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| CO Column Burden                              | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| CO Surface Concentration in ppbv              | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| total_column_ozone                            | 2003-2019 | 0.5 °×0.625 ° | MERRA2            |   |
| Nitrogen dioxide                              | 2003-2019 | 0.9 °×1.25 °  | CAM_CHEM          | Atmospheric Chemistry Observations & Modeling, National Center for Atmospheric Research, University Corporation for Atmospheric Research (2020) |
| Ozone   | 2003-2019 | 0.9 °×1.25 °  | CAM_CHEM          |   |
| Nitrogen monoxide                             | 2003-2019 | 0.9 °×1.25 °  | CAM_CHEM          |   |
| Population                                    | 2003-2019 | 1 km          | Worldpop          | (Lloyd et al., 2019)  |
| Elevation mean                                | 2003-2019 | 1 arc-minute  | NOAA/NGDC/E TOPO1 | (Amante and Eakins, 2009)   |
| Elevation sd                                  | 2003-2019 | 1 arc-minute  | NOAA/NGDC/E TOPO1 |   |
| Water percentage                              | 2003-2019 | 0.05 °        | MODIS             | (Friedl, 2015)  |
| Shrublands percentage                         | 2003-2019 | 0.05 °        | MODIS             |   |

|  |           |            |          |                                |
|--|-----------|------------|----------|--------------------------------|
| Savannas percentage  | 2003-2019 | 0.05 °     | MODIS    |                                |
| Non Vegetated percentage   | 2003-2019 | 0.05 °     | MODIS    |                                |
| Grasslands percentage  | 2003-2019 | 0.05 °     | MODIS    |                                |
| Croplands percentage   | 2003-2019 | 0.05 °     | MODIS    |                                |
| Forests percentage   | 2003-2019 | 0.05 °     | MODIS    |                                |
| Urban percentage   | 2003-2019 | 0.05 °     | MODIS    |                                |
| Dark Target and Deep Blue AOD @ 0.55 micron                      | 2003-2019 | 1 °        | CERES    | (Doelling et al., 2016)        |
| Tropospheric vertical column densities (VCDs) of NO <sub>2</sub> | 2004-2019 | 0.25 °     | OMI/Aura | (Krotkov et al., 2017)         |
| tropospheric ozone   | 2004-2019 | 1 °        | OMI/Aura |                                |
| Total ozone column   | 2003-2019 | 1.25 °×1 ° | TCO      | (Greg E. Bodeker, 2022)        |
| Total ozone column uncertainty                                   | 2003-2019 | 1.25 °×1 ° | TCO      |                                |
| Leaf area index  | 2003-2019 | 0.5 °      | MODIS    | (Lin et al., 2023)             |
| bc emission  | 2003-2019 | 0.5 °      | CEDS     | DOI:<br>10.5281/zenodo.3592072 |
| nox emission   | 2003-2019 | 0.5 °      | CEDS     |                                |
| co emission  | 2003-2019 | 0.5 °      | CEDS     |                                |
| nmvoc emission   | 2003-2019 | 0.5 °      | CEDS     |                                |
| ch4 emission   | 2003-2019 | 0.5 °      | CEDS     |                                |
| so2 emission   | 2003-2019 | 0.5 °      | CEDS     |                                |
| oc emission  | 2003-2019 | 0.5 °      | CEDS     |                                |

**Table S3. Inputs to the UKML dataset include multiple CMIP6 Chemistry–Climate Models. In situ observations from TOAR-I (1990–2014) and CNEMC (2015–2019) are used as labels for supervised learning. Table based on Sun et al. (2022)**

| Model <sup>a</sup> | Grids <sup>b</sup> | Hist <sup>c</sup> | Ssp245 <sup>d</sup> | Reference   |
|--------------------|--------------------|-------------------|---------------------|---|
| BCC-ESM1           | 128×64             | 3                 |                     | (Wu et al., 2020; Zhang et al., 2018)   |
| MPI-ESM1.2-HAM     | 192×96             | 3                 |                     | (Neubauer et al., 2019)   |
| MPI-ESM1.2-HR      | 384×192            | 19                | 3                   | (Von Storch et al., 2017; Gutjahr et al., 2019; Schupfner et al., 2019)   |
| UKESM1-0-LL        | 192×144            | 3                 |                     | (Tang et al., 2019; Good et al., 2019; Yool et al., 2020; Sellar et al., 2020; Sellar et al., 2019; Mulcahy et al., 2018; Archibald et al., 2020) |
| MRI-ESM2.0         | 128×64             | 5                 | 5                   | (Yukimoto et al., 2019a; Yukimoto et al., 2012; Yukimoto et al., 2019b)   |
| NASA-GISS-E2.1-G   | 144×90             | 19                | 20                  | (Studies, 2018b, a, 2020; Shindell et al., 2013)  |
| NASA-GISS-E2.1-H   | 144×90             | 10                |                     |   |
| NCAR-CESM2-WACCM   | 288×192            | 3                 |                     | (Danabasoglu, 2019; Gettelman et al., 2019)   |
| NCC-NorESM-MM      | 288×192            | 3                 | 2                   | (Seland et al., 2019; Bentsen et al., 2019)   |
| NOAA-GFDL-ESM4     | 288×180            | 1                 | 1                   | (John et al., 2018; Horowitz et al., 2018; Krasting et al., 2018)   |
| EC-Earth3          | 120×90             | 2                 |                     | (Consortium, 2020)  |
| BCC-CSM2           | 320×160            | 1                 |                     | (Hegglin et al., 2016; Wu et al., 2021)   |

<sup>a</sup>The names of coupled earth system models are abbreviated.

<sup>b</sup>The planar dimensional spatial resolutions are presented in longitudinal latitudinal grids.

<sup>c</sup>Numbers of simulation ensembles for historical period (1990–2014, using historical experiments)

<sup>d</sup>Numbers of simulation ensembles for recent years (2015–2019, using ssp245 experiments)

**Table S4. Satellite observations used for ozone assimilation in CAMS (CAMS global ECMWF Atmospheric Composition Reanalysis 4). Table based on Inness et al. (2019)**

| Species                | Instrument               | Period    | Data type <sup>a</sup>                | Reference                                     |
|------------------------|--------------------------|-----------|---------------------------------------|---|
| Ozone, NO <sub>2</sub> | SCIAMACHY (Envisat)      | 2003-2012 | TC for Ozone, TRC for NO <sub>2</sub> | (Lerot et al., 2009)                          |
| Ozone                  | MIPAS (Envisat)          | 2003-2012 | PROF                                  | (Von Clarmann et al., 2009)                   |
| Ozone                  | MLS (Aura)               | 2004-2016 | PROF                                  | (Schwartz, 2015)                              |
| Ozone, NO <sub>2</sub> | OMI (Aura)               | 2004-2020 | TC for Ozone, TRC for NO <sub>2</sub> | (Liu et al., 2010)                            |
| Ozone, NO <sub>2</sub> | GOME-2 (Metop-A)         | 2007-2020 | TC for Ozone, TRC for NO <sub>2</sub> | (Hao et al., 2014)                            |
| Ozone, NO <sub>2</sub> | GOME-2 (Metop-B)         | 2013-2020 | TC for Ozone, TRC for NO <sub>2</sub> |   |
| Ozone                  | SBUV/2 (NOAA-14–NOAA-19) | 2003-2020 | PC                                    | (Bhartia et al., 1996; Mcpeters et al., 2013) |
| CO                     | MOPITT (Terra)           | 2002-2020 | TC                                    | (Deeter et al., 2014)                         |

<sup>a</sup>Satellite retrievals of atmospheric composition that were assimilated in the chemistry reanalysis. TC is total column, TRC is tropospheric column, PROF is profiles, PC is partial columns.



**Table S5. Inputs used for ozone assimilation in GEOS. Table based on Qu et al. (2020)**

| Variable                                | Years     | Source                        | Reference   |
|---|-----------|-------------------------------|---|
| NO <sub>2</sub> tropospheric column     | 2005-2016 | OMNO2                         | (Qu et al., 2020b)                                    |
| NO <sub>2</sub> tropospheric column     | 2005-2016 | DOMINO                        | (Qu et al., 2020a)                                    |
| Meteorological fields                   | 2005-2016 | MERRA-2                       | Global Modeling and Assimilation Office (GMAO) (2015) |
| Anthropogenic emissions <sup>a</sup>    | 2005-2016 | HTAP 2010 inventory version 2 | (Janssens-Maenhout et al., 2015)                      |
| Anthropogenic emissions                 | 2005-2016 | CEDS                          | (Hoesly et al., 2018)                                 |
| Nonanthropogenic emissions <sup>b</sup> | 2005-2016 | Follows Qu et al. 2017        | (Qu et al., 2017)                                     |

<sup>a</sup>Anthropogenic emissions include NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, CO, NMVOC<sub>s</sub> (non-methane volatile organic compounds), and primary aerosols.

<sup>b</sup>Nonanthropogenic emissions include biomass burning emissions, NO<sub>x</sub> emissions from lightning, and soil NO<sub>x</sub> emissions.

**Table S6. Satellite observations used for ozone assimilation in TCR-2. Table based on Miyazaki et al. (2020)**

| Species                           | Instrument          | Period    | Data type <sup>a</sup>                            | Reference   |
|-----------------------------------|---------------------|-----------|---|---|
| NO <sub>2</sub> , SO <sub>2</sub> | OMI (Aura)          | 2005-2020 | TrC for NO <sub>2</sub> , PBL for SO <sub>2</sub> | (Boersma et al., 2011; Boersma et al., 2017; Krotkov et al., 2016; Li et al., 2013) |
| NO <sub>2</sub>                   | SCIAMACHY (Envisat) | 2005-2012 | TrC   | (Boersma et al., 2004)  |
| NO <sub>2</sub>                   | GOME-2 (Metop-A)    | 2007-2020 | TrC   |   |
| Ozone                             | TES (Aura)          | 2005-2011 | PROF  | (Bowman et al., 2006; Herman and Kulawik, 2013)                                     |
| Ozone, HNO <sub>3</sub>           | MLS (Aura)          | 2005-2020 | PROF  | (Livesey et al., 2017)  |
| CO                                | MOPITT (Terra)      | 2005-2020 | PROF  | (Deeter et al., 2017; Deeter et al., 2013)  |

<sup>a</sup>Satellite retrievals of atmospheric composition that were assimilated in the chemistry reanalysis. TC is total column, TRC is tropospheric column, PROF is profiles, PC is partial columns. PBL is planetary boundary layer.

**Table S7. The regions defined by Hemispheric Transport Air Pollution(HTAP)2 (Koffi et al., 2016)**

|    | Tier 1      |   | Tier 2  |
|----|-------------|---|---|
| 01 | <b>GLO*</b> | World   |   |
| 02 | <b>OCN</b>  | Non-arctic/Antarctic Ocean                                | 020 Baltic Sea<br>021 North Atlantic<br>022 South Atlantic<br>023 North Pacific<br>024 South Pacific<br>025 Indian Ocean<br>026 Hudson Bay<br>027 Mediterranean Sea<br>028 Black and Caspian Sea  |
| 03 | <b>NAM*</b> | US+Canada (upto 66 N; polar circle)                       | 031 NE US (all divided on state or provincial lines)<br>032 SE US<br>033 NW US<br>034 SW US<br>035 E. Canada<br>036 W. Canada + Alaska up to 66 N.  |
| 04 | <b>EUR*</b> | Western + Eastern EU+Turkey (upto 66 N polar circle)      | 041 NW Europe<br>042 SW Europe (France follows provinces level at ca. 46 N).<br>043 Eastern Europe<br>044 Greece+Turkey+Cyprus  |
| 05 | <b>SAS*</b> | South Asia: India, Pakistan, Nepal, Bangladesh, Sri Lanka | 051 North India+Pakistan+Nepal+Bangladesh<br>052 South India+Sri Lanka<br>053 Indian Himalaya (above an elevation of 1500 m)  |
| 06 | <b>EAS*</b> | E Asia: China, Korea, Japan                               | 061 North East China<br>062 South East China<br>063 West China +Mongolia (excl. Himalaya)<br>064 North/South Korea<br>065 Japan<br>066 China/Tibet Himalaya (above an elevation of 1500 m)  |
| 07 | <b>SEA*</b> | South East Asia   | 071 Indonesia+Malaysia+Singapore<br>072 Thailand+Myanmar+Vietnam  |
| 08 | <b>PAN</b>  | Pacific, Australia+ New Zealand                           | 081 Pacific<br>082 Australia<br>083 New Zealand   |
| 09 | <b>NAF</b>  | Northern Africa   | 091 Egypt<br>092 Rest of Northern Africa  |
| 10 | <b>SAF*</b> | Sub Saharan Africa  | 101 West and Central Africa: Côte d'Ivoire, Angola, Benin, Burkina Faso, Cameroon, Cape Verde, Chad, Congo Brazzaville, Democratic Republic of Congo, Equatorial Guinea, Gambia, Ghana, Guinea, Guinea Bissau, Liberia, Mali, Niger, Nigeria, Senegal, Sierra Leone and Togo<br>102 East Africa: Burundi, Djibouti, Eritrea, Ethiopia, Kenya, Sudan, Rwanda, Uganda, Somalia and Tanzania.<br>103 Southern Africa: Angola, Botswana, Lesotho, Madagascar, Malawi, Mauritius, Mozambique, Namibia, South Africa, Swaziland, Tanzania, Zambia, and Zimbabwe |
| 11 | <b>MDE*</b> | Middle East; S. Arabia etc, Iran, Iraq                    | 111 Middle East<br>112 S.Arabia; Yemen; Oman; etc<br>113 Iran, Iraq   |
| 12 | <b>MCA</b>  | Mexico, Central America, Caribbean,                       | 121 Mexico<br>122 Central America<br>123 Caribbean  |

|    |            |  |  |
|----|------------|--|--|
|    |            | <b>Guyanas, Venezuela, Columbia</b>              | 124 Guyanas, Columbia, Venezuela   |
| 13 | <b>SAM</b> | <b>S. America</b>                                | 131 South Brazil<br>132 Rest of Brazil<br>133 Uruguay, Paraguay, Argentina, Chile<br>134 Peru, Ecuador |
| 14 | <b>RBU</b> | <b>Russia, Belarussia, Ukraine, Central Asia</b> | 141 Russia West<br>142 Russia East<br>143 Belarussia+Ukraine   |
| 15 | <b>CAS</b> | <b>Central Asia</b>                              | 144 Uzbekistan, Kazakhstan, Kyrgyzstan, Tajikistan, Turkmenistan                                       |
| 16 | <b>NPO</b> | <b>Arctic Circle (North of 66 N)+Greenland</b>   | 150 Arctic (includes ocean and all of Greenland)   |
| 17 | <b>SPO</b> | <b>Antarctic</b>                                 | 160 Antarctic<br>161 Southern Ocean, south of 60S  |

\*Regions in bold characters defined as priority regions in this paper.

**Table S8. The share of population exposed to ozone above particular thresholds in each world region for each year (from 2006 to 2016). The unit of thresholds is ppb and unit of share is percentage (%).**

| 2006  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
|-------|--------|------|------|------|------|------|------|------|------|
| BME   | >30    | 100  | 99.5 | 100  | 99.8 | 92.5 | 100  | 77.3 | 95.1 |
|       | >50    | 57.7 | 18.6 | 73.8 | 47.3 | 10   | 79.8 | 0.5  | 42.2 |
|       | >70    | 0    | 0.3  | 0    | 0.8  | 0    | 0    | 0    | 0.1  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 93.5 | 99.4 |
|       | >50    | 79.3 | 92.3 | 99.5 | 91.1 | 35.2 | 98.5 | 31.8 | 73.7 |
|       | >70    | 3    | 2.5  | 17.4 | 9.5  | 0    | 13.7 | 0    | 6    |
| UKML  | >30    | 100  | 100  | 100  | 100  | 95.8 | 100  | 97.7 | 97.2 |
|       | >50    | 99.7 | 87.7 | 90.9 | 89.4 | 11.4 | 96   | 29.5 | 68.7 |
|       | >70    | 20.5 | 0    | 0    | 0    | 0    | 6.8  | 0    | 6    |
| CAMS  | >30    | 100  | 97.7 | 100  | 99.9 | 85.7 | 100  | 90.8 | 93.5 |
|       | >50    | 71   | 20.8 | 91.4 | 61.7 | 7.3  | 94.8 | 28.7 | 51.9 |
|       | >70    | 3.3  | 0    | 9.4  | 3.6  | 0.1  | 13.2 | 7.1  | 5    |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 98.7 | 100  | 93.7 | 97.8 |
|       | >50    | 95.4 | 54.6 | 99.5 | 71.4 | 13.2 | 92   | 5.5  | 60.6 |
|       | >70    | 4.8  | 0    | 4.2  | 0    | 0    | 0.2  | 0    | 1.3  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 97.5 | 100  | 86.5 | 97.1 |
|       | >50    | 94.1 | 78.9 | 92.5 | 88.6 | 18.3 | 87.3 | 19.5 | 64.9 |
|       | >70    | 37.2 | 0.1  | 33.5 | 3.1  | 1.3  | 2.8  | 0    | 11.1 |
| 2007  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 100  | 97.2 | 100  | 99.4 | 96.8 | 100  | 86.1 | 96.4 |
|       | >50    | 76.7 | 19.9 | 80.5 | 42.1 | 12.7 | 85.8 | 0.7  | 48.5 |
|       | >70    | 17.4 | 0    | 0    | 0.6  | 0    | 0    | 0    | 3.7  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 85.3 | 99   |
|       | >50    | 78.8 | 79.6 | 99.5 | 91.4 | 38.7 | 98.5 | 31.3 | 73.2 |
|       | >70    | 3.6  | 0    | 17.9 | 12.2 | 0    | 10.2 | 0    | 5    |
| UKML  | >30    | 100  | 100  | 100  | 100  | 97.6 | 100  | 96.5 | 97.8 |
|       | >50    | 99.9 | 81.4 | 92.2 | 90.1 | 10.7 | 97.3 | 35.9 | 69.3 |
|       | >70    | 23.8 | 0    | 0    | 0    | 0    | 14.9 | 0    | 8.6  |
| CAMS  | >30    | 100  | 92.5 | 100  | 100  | 91.5 | 100  | 86.9 | 94.6 |
|       | >50    | 77.2 | 19.6 | 88.2 | 52.2 | 12.6 | 94.7 | 28.3 | 53   |
|       | >70    | 0    | 0    | 8.1  | 4.2  | 0    | 8.4  | 6.6  | 3.1  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 98.5 | 100  | 83.6 | 97.1 |
|       | >50    | 97.1 | 40.3 | 99.5 | 58.3 | 11   | 94.3 | 2.6  | 59.5 |
|       | >70    | 2.9  | 0    | 3.6  | 0    | 0    | 0.3  | 0    | 0.8  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 98.7 | 100  | 78.9 | 96.9 |
|       | >50    | 93.2 | 70.7 | 93.4 | 86.9 | 20.3 | 85.3 | 6.5  | 62.8 |
|       | >70    | 37.5 | 0.2  | 34.1 | 1.7  | 0.3  | 2.8  | 0    | 10.7 |
| 2008  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 100  | 100  | 100  | 99.5 | 88.6 | 100  | 76.1 | 93.9 |
|       | >50    | 76.5 | 77.7 | 84   | 30.3 | 4    | 87.5 | 0.1  | 46.4 |
|       | >70    | 0.6  | 0    | 4.5  | 0.9  | 0    | 0.9  | 0    | 0.5  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 85.6 | 99   |
|       | >50    | 76.8 | 75.8 | 99.5 | 89.3 | 38.6 | 98.4 | 25.1 | 71.4 |
|       | >70    | 3.2  | 0    | 9.1  | 4    | 0    | 8.4  | 0    | 3.8  |
| UKML  | >30    | 100  | 97.4 | 100  | 100  | 97.8 | 100  | 96.5 | 97.9 |
|       | >50    | 99.9 | 1.9  | 89.3 | 87.4 | 10.2 | 98.4 | 29.9 | 68.5 |
|       | >70    | 32.6 | 0    | 0    | 0    | 0    | 28.3 | 0    | 13.6 |
| CAMS  | >30    | 100  | 100  | 100  | 100  | 77.7 | 100  | 84.4 | 90.6 |
|       | >50    | 66.1 | 36   | 79.9 | 14   | 9.1  | 93.8 | 21   | 44.5 |
|       | >70    | 0.1  | 0    | 5.1  | 0    | 0    | 4    | 5.5  | 1.6  |

|       |        |      |      |      |      |      |      |      |      |
|-------|--------|------|------|------|------|------|------|------|------|
| GEOS  | >30    | 100  | 100  | 100  | 100  | 98.3 | 100  | 83.6 | 97   |
|       | >50    | 93.9 | 70.4 | 99.4 | 46.5 | 15.8 | 93.1 | 0.2  | 57.7 |
|       | >70    | 2.1  | 0.1  | 0.4  | 0    | 0    | 0    | 0    | 0.5  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 96   | 100  | 76.2 | 95.5 |
|       | >50    | 89.3 | 77.7 | 93.2 | 81.8 | 19.5 | 86.7 | 3.7  | 61.3 |
|       | >70    | 31.5 | 0    | 32.5 | 0.8  | 0.3  | 5.5  | 0    | 9.7  |
| 2009  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 100  | 98.3 | 99.8 | 99.8 | 69.7 | 99.4 | 80.2 | 90.5 |
|       | >50    | 76.8 | 18.3 | 79.3 | 13.4 | 0.8  | 84.7 | 0.4  | 44.7 |
|       | >70    | 6.2  | 0    | 0.1  | 0.5  | 0    | 0    | 0    | 1.3  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 99.9 | 100  | 89.9 | 99.3 |
|       | >50    | 79.2 | 77.2 | 99.2 | 88.5 | 36.2 | 99.1 | 32.8 | 71.8 |
|       | >70    | 3.5  | 0    | 5.4  | 3.4  | 0    | 19   | 0    | 6.6  |
| UKML  | >30    | 100  | 100  | 100  | 100  | 98   | 100  | 97   | 97.4 |
|       | >50    | 99.8 | 79   | 92   | 85.5 | 9.9  | 98.2 | 34   | 68.4 |
|       | >70    | 20.4 | 0    | 0    | 0    | 0    | 29.6 | 0    | 11.3 |
| CAMS  | >30    | 100  | 91.6 | 100  | 100  | 90.8 | 100  | 88.1 | 93.7 |
|       | >50    | 72.9 | 15.4 | 87.8 | 28.7 | 6.8  | 95.2 | 30.9 | 50   |
|       | >70    | 0.1  | 0    | 6.5  | 3.6  | 0.1  | 9.5  | 6.6  | 3.4  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 97.9 | 100  | 91.7 | 97.3 |
|       | >50    | 96.4 | 44.3 | 97.9 | 36.9 | 11.4 | 97.1 | 2    | 58.8 |
|       | >70    | 3.5  | 0    | 2.7  | 0    | 0    | 1.4  | 0    | 1.2  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 97.5 | 100  | 82.9 | 97.1 |
|       | >50    | 93   | 74.9 | 93.3 | 85.6 | 15.9 | 88.9 | 13   | 63.5 |
|       | >70    | 36.1 | 0.1  | 34.3 | 0    | 0.6  | 9.5  | 0    | 11.6 |
| 2010  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 100  | 97.4 | 98.7 | 99.2 | 80.8 | 99.1 | 71.8 | 91.7 |
|       | >50    | 65.6 | 11.1 | 62.4 | 27.3 | 1.4  | 74.1 | 0    | 38.5 |
|       | >70    | 0    | 0    | 0    | 0.3  | 0    | 0    | 0    | 0    |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 82   | 98.9 |
|       | >50    | 62.9 | 73.1 | 99.5 | 87.7 | 34.1 | 98.3 | 21.4 | 67.8 |
|       | >70    | 3.1  | 0    | 8.2  | 2.5  | 0    | 8.9  | 0    | 3.6  |
| UKML  | >30    | 100  | 100  | 100  | 100  | 97.3 | 100  | 96.4 | 97.8 |
|       | >50    | 98.4 | 79.8 | 93.4 | 81.9 | 8.3  | 98.2 | 27.9 | 67.3 |
|       | >70    | 31.6 | 0    | 0    | 0    | 0    | 29.3 | 0    | 13.7 |
| CAMS  | >30    | 100  | 88.2 | 100  | 100  | 73.5 | 100  | 84.4 | 89.5 |
|       | >50    | 32.2 | 2    | 76.2 | 15   | 4.5  | 95.9 | 23.5 | 37.4 |
|       | >70    | 0    | 0    | 5.6  | 0    | 0    | 24.7 | 4.9  | 6.4  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 99.1 | 100  | 81.3 | 97.1 |
|       | >50    | 88.3 | 35.2 | 99.7 | 50.6 | 17.5 | 94.8 | 1.9  | 57.9 |
|       | >70    | 3.5  | 0    | 4.4  | 0    | 0    | 0.2  | 0    | 0.9  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 98   | 99.8 | 75.7 | 96.3 |
|       | >50    | 93.5 | 64.5 | 92.4 | 84.7 | 18.2 | 86.1 | 14.2 | 62.9 |
|       | >70    | 37.7 | 0.3  | 40.7 | 0    | 1.1  | 6.6  | 0    | 11.9 |
| 2011  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 100  | 98.9 | 99.3 | 99.4 | 94.1 | 100  | 81.3 | 95.8 |
|       | >50    | 68.5 | 15.5 | 72.6 | 26.6 | 8.6  | 89.8 | 7.9  | 46   |
|       | >70    | 0.3  | 0    | 0    | 0.4  | 0    | 2.1  | 0    | 0.6  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 88.1 | 99   |
|       | >50    | 73.5 | 75.6 | 99.4 | 87.6 | 37.6 | 98.6 | 24.6 | 70.2 |
|       | >70    | 3.4  | 0.3  | 5    | 4.5  | 0    | 9.3  | 0    | 4    |
| UKML  | >30    | 100  | 100  | 100  | 100  | 97.3 | 100  | 96.6 | 97.6 |
|       | >50    | 99.9 | 75.5 | 93.6 | 85.2 | 12.5 | 98.9 | 43.5 | 69.5 |

|       |        |      |      |      |      |      |      |      |      |
|-------|--------|------|------|------|------|------|------|------|------|
|       | >70    | 29.1 | 0    | 0    | 0    | 0    | 35.5 | 0    | 14.6 |
| CAMS  | >30    | 100  | 92   | 100  | 99.9 | 92.3 | 100  | 88.6 | 94.3 |
|       | >50    | 66.5 | 14.5 | 91.7 | 54.6 | 11.7 | 97.2 | 27.7 | 51   |
|       | >70    | 2.9  | 0    | 10.4 | 0    | 0.2  | 9.2  | 6.6  | 3.8  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 99.2 | 100  | 90   | 97.6 |
|       | >50    | 97   | 38.6 | 99.7 | 55.2 | 15   | 95.1 | 0.1  | 59.4 |
|       | >70    | 2.4  | 0    | 2.3  | 0    | 0    | 0.6  | 0    | 0.7  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 97.6 | 100  | 81.7 | 96.5 |
|       | >50    | 94.1 | 69.4 | 96.1 | 86.1 | 19.6 | 89.7 | 11.3 | 64.2 |
|       | >70    | 40.2 | 0.1  | 35.3 | 0    | 0.4  | 17.4 | 0    | 14.4 |
| 2012  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 99.8 | 97.7 | 100  | 99.5 | 92.9 | 100  | 82.6 | 96.3 |
|       | >50    | 60.8 | 21.4 | 80.9 | 35.5 | 0.8  | 92.8 | 9    | 45.1 |
|       | >70    | 0.1  | 0    | 0    | 0.4  | 0    | 1.3  | 0    | 0.3  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 88.7 | 99.1 |
|       | >50    | 69.5 | 73.1 | 99.5 | 90.6 | 30.2 | 98.7 | 31.4 | 68.7 |
|       | >70    | 3.4  | 0    | 8.4  | 3.2  | 0    | 8.1  | 0    | 3.5  |
| UKML  | >30    | 100  | 100  | 100  | 100  | 95.8 | 100  | 97.6 | 97.5 |
|       | >50    | 99.9 | 74.6 | 93.2 | 83.1 | 10   | 98.5 | 42.8 | 68.7 |
|       | >70    | 33.3 | 0    | 0    | 0    | 0    | 35   | 0    | 15.4 |
| CAMS  | >30    | 100  | 90.1 | 100  | 100  | 76.3 | 100  | 84.1 | 89.5 |
|       | >50    | 33.7 | 1    | 83.1 | 13.9 | 5.3  | 96.3 | 22.1 | 38.1 |
|       | >70    | 0.1  | 0    | 5.1  | 0    | 0    | 15.2 | 5.4  | 4.2  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 97.9 | 100  | 89.3 | 97.6 |
|       | >50    | 96   | 46.9 | 100  | 79.8 | 13.8 | 97.5 | 1    | 62   |
|       | >70    | 6    | 0    | 13.4 | 0    | 0    | 2.6  | 0    | 2.4  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 96.9 | 100  | 86.6 | 97.3 |
|       | >50    | 94.6 | 67.7 | 96.4 | 83.7 | 16.8 | 92.5 | 15.8 | 64.6 |
|       | >70    | 42.7 | 0.1  | 45.1 | 0    | 0.4  | 15.4 | 0    | 15.1 |
| 2013  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 98.6 | 99.6 | 99.5 | 99.1 | 93.2 | 100  | 79.1 | 96   |
|       | >50    | 35.2 | 22.1 | 74.2 | 12.4 | 1.9  | 84.6 | 0.2  | 35.7 |
|       | >70    | 0.1  | 0    | 0    | 0.3  | 0    | 0    | 0    | 0    |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 88.8 | 99.1 |
|       | >50    | 72.6 | 73.5 | 98.9 | 85   | 32.8 | 98.5 | 26.1 | 68.6 |
|       | >70    | 3    | 0    | 4.7  | 2.5  | 0    | 4.7  | 0    | 2.4  |
| UKML  | >30    | 100  | 100  | 100  | 100  | 96.1 | 100  | 96.9 | 97.5 |
|       | >50    | 99.9 | 69.5 | 92.4 | 84.7 | 12.7 | 99.8 | 43.5 | 69.3 |
|       | >70    | 34.8 | 0    | 0    | 0    | 0    | 34.8 | 0    | 15.6 |
| CAMS  | >30    | 100  | 99.6 | 100  | 100  | 86.2 | 100  | 88.6 | 93.5 |
|       | >50    | 73.9 | 11.5 | 86.8 | 45   | 6.3  | 97.4 | 19.4 | 50.3 |
|       | >70    | 1    | 0    | 10.4 | 0    | 0.1  | 14.3 | 5.9  | 4.4  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 98.7 | 100  | 88.3 | 97.5 |
|       | >50    | 92.2 | 42.8 | 99.7 | 45.9 | 14.5 | 94.9 | 0.5  | 58.5 |
|       | >70    | 1.2  | 0    | 6.6  | 0    | 0    | 0.3  | 0    | 0.6  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 97.1 | 100  | 86.7 | 97.3 |
|       | >50    | 94.5 | 71.3 | 96.4 | 83.4 | 17.3 | 90.4 | 19   | 64.8 |
|       | >70    | 39.8 | 0.1  | 40.2 | 0    | 1.2  | 13.2 | 0    | 13.4 |
| 2014  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 98.9 | 99.3 | 99.8 | 99.4 | 100  | 100  | 86.8 | 96   |
|       | >50    | 45.5 | 20.4 | 79.8 | 11.4 | 27.1 | 96.3 | 7.5  | 45.7 |
|       | >70    | 0.6  | 0    | 0    | 0.2  | 0    | 8.1  | 0    | 2    |
| NJML  | >30    | 100  | 100  | 100  | 100  | 99.9 | 100  | 91.5 | 99.2 |

|       |        |      |      |      |      |      |      |      |      |
|-------|--------|------|------|------|------|------|------|------|------|
|       | >50    | 64.8 | 65.1 | 98.8 | 83.1 | 28.9 | 98.7 | 31.2 | 65.9 |
|       | >70    | 3.7  | 0    | 3.9  | 2.5  | 0    | 8.8  | 0    | 3.5  |
| UKML  | >30    | 100  | 100  | 100  | 100  | 98.6 | 100  | 97.7 | 98.3 |
|       | >50    | 99.9 | 71.9 | 94.5 | 83.3 | 10.8 | 99.6 | 46.1 | 69.1 |
|       | >70    | 31.6 | 0    | 0    | 0    | 0    | 41.4 | 0    | 16.5 |
| CAMS  | >30    | 100  | 99.2 | 100  | 100  | 83.1 | 100  | 91.3 | 93.2 |
|       | >50    | 68.9 | 9    | 89   | 39.3 | 6.8  | 92   | 26.8 | 48   |
|       | >70    | 0.1  | 0    | 9.5  | 0    | 0.1  | 12.4 | 6.3  | 3.8  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 98.5 | 100  | 90   | 97.6 |
|       | >50    | 94.5 | 47.1 | 99.9 | 44   | 13.2 | 96.1 | 1.6  | 59   |
|       | >70    | 23.5 | 0    | 5.6  | 0    | 0    | 0.3  | 0    | 5.2  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 97.2 | 100  | 91.3 | 97.7 |
|       | >50    | 94.7 | 69.8 | 93.8 | 81.4 | 16.3 | 91.4 | 21.7 | 64.4 |
|       | >70    | 48.4 | 0    | 41.1 | 0    | 0.9  | 14.9 | 0    | 15.7 |
| 2015  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 99.4 | 97.2 | 100  | 99.5 | 91.1 | 99.9 | 84.6 | 95.6 |
|       | >50    | 46.7 | 19.5 | 81.7 | 13.9 | 2.4  | 92.9 | 7.1  | 40.5 |
|       | >70    | 0.3  | 0    | 0    | 0    | 0    | 0.1  | 0.7  | 0.1  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 95   | 99.6 |
|       | >50    | 64.8 | 70.4 | 99   | 80.9 | 38   | 96.8 | 29.6 | 67.1 |
|       | >70    | 3.2  | 0.1  | 3.5  | 1.9  | 0    | 6.2  | 0    | 2.6  |
| UKML  | >30    | 100  | 100  | 100  | 100  | 99.1 | 100  | 97.6 | 98.1 |
|       | >50    | 99.9 | 81.2 | 96.4 | 63.2 | 12.9 | 99   | 46   | 69.5 |
|       | >70    | 33.7 | 0    | 0.9  | 0    | 0.8  | 44.5 | 0    | 17.8 |
| CAMS  | >30    | 100  | 100  | 100  | 100  | 85.2 | 100  | 91.3 | 93.4 |
|       | >50    | 72.1 | 13.6 | 90.8 | 40.1 | 5.1  | 94.6 | 34.6 | 50.1 |
|       | >70    | 1.2  | 0    | 12.2 | 0    | 0.1  | 12.4 | 7.9  | 4.4  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 98.8 | 100  | 90.6 | 97.6 |
|       | >50    | 92.3 | 50.5 | 99.9 | 43.2 | 15.7 | 92.1 | 0.9  | 58.1 |
|       | >70    | 14.1 | 0    | 2.5  | 0    | 0    | 0.5  | 0    | 3.2  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 98.5 | 100  | 90.5 | 97.9 |
|       | >50    | 94.5 | 71.2 | 93.4 | 80.2 | 20.7 | 91.2 | 20.2 | 64.7 |
|       | >70    | 42.3 | 0.1  | 37.1 | 0    | 1.5  | 29.9 | 0.6  | 17.7 |
| 2016  | Region | EAS  | EUR  | MDE  | NAM  | SAF  | SAS  | SEA  | GLO  |
| BME   | >30    | 100  | 97.4 | 100  | 99.5 | 95.7 | 100  | 67.3 | 95.1 |
|       | >50    | 53.9 | 13.5 | 82.3 | 12.4 | 7.9  | 97.2 | 2.5  | 43.5 |
|       | >70    | 0.6  | 0    | 0.2  | 0    | 0    | 2.3  | 0    | 0.7  |
| NJML  | >30    | 100  | 100  | 100  | 100  | 100  | 100  | 84.3 | 99   |
|       | >50    | 53.9 | 64.2 | 97.7 | 15.5 | 39.7 | 97.1 | 19.1 | 63.2 |
|       | >70    | 0.6  | 0    | 3.1  | 0    | 0    | 7.1  | 0    | 2.8  |
| UKML  | >30    | 100  | 100  | 100  | 100  | 98.9 | 100  | 97.6 | 98.1 |
|       | >50    | 99.9 | 81.1 | 95.9 | 62.8 | 13.2 | 98.4 | 47.6 | 69.5 |
|       | >70    | 33.3 | 0    | 0.5  | 0    | 0.9  | 50.6 | 0    | 19.2 |
| CAMS  | >30    | 100  | 98.8 | 100  | 100  | 84.3 | 100  | 85.6 | 93.2 |
|       | >50    | 68.3 | 11.5 | 88.9 | 42.3 | 12.7 | 96.7 | 20.9 | 50   |
|       | >70    | 1.2  | 0    | 14.7 | 0.4  | 0    | 26.4 | 5.6  | 7.5  |
| GEOS  | >30    | 100  | 100  | 100  | 100  | 99.1 | 100  | 82.9 | 96.8 |
|       | >50    | 86.5 | 49.9 | 99.9 | 46.2 | 13.9 | 97.4 | 0.2  | 58.1 |
|       | >70    | 1.6  | 0    | 9    | 0    | 0    | 0.4  | 0    | 0.8  |
| TCR-2 | >30    | 100  | 100  | 100  | 100  | 98.2 | 100  | 84.1 | 97.3 |
|       | >50    | 93.5 | 67.4 | 96.4 | 84.1 | 22.7 | 93.8 | 18.4 | 65.9 |
|       | >70    | 40.2 | 0    | 40.6 | 0    | 1    | 33.4 | 0    | 18.6 |



**Table S9. Different quantiles of TOAR-II observations for 2006 to 2015. The unit of TOAR-II observations is ppb.**

| Year | 2006  | 2007  | 2008  | 2009  | 2010  | 2011  | 2012  | 2013  | 2014  | 2015  |
|------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| 0%   | 12.52 | 15.74 | 15.32 | 13.25 | 12.28 | 8.25  | 12.01 | 9.07  | 8.76  | 12.24 |
| 10%  | 37.73 | 36.42 | 37.29 | 37.16 | 36.35 | 36.00 | 36.01 | 37.00 | 36.52 | 36.85 |
| 20%  | 41.58 | 39.46 | 40.32 | 40.14 | 39.35 | 38.83 | 38.79 | 39.52 | 39.25 | 39.92 |
| 30%  | 43.69 | 41.75 | 42.32 | 42.19 | 41.16 | 40.77 | 40.67 | 41.42 | 41.09 | 42.02 |
| 40%  | 45.56 | 43.96 | 44.13 | 44.06 | 42.64 | 42.49 | 42.29 | 42.96 | 42.74 | 43.89 |
| 50%  | 47.24 | 46.03 | 45.93 | 45.92 | 43.97 | 43.92 | 43.90 | 44.43 | 44.20 | 45.63 |
| 60%  | 48.87 | 48.08 | 47.76 | 48.13 | 45.28 | 45.28 | 45.53 | 45.71 | 45.59 | 47.44 |
| 70%  | 50.63 | 50.40 | 49.78 | 50.10 | 46.92 | 46.99 | 47.58 | 47.29 | 47.14 | 49.60 |
| 80%  | 52.81 | 52.96 | 52.18 | 52.45 | 49.07 | 49.61 | 49.91 | 49.33 | 48.87 | 52.58 |
| 90%  | 55.82 | 56.31 | 55.51 | 55.50 | 52.57 | 53.50 | 53.29 | 52.60 | 51.98 | 57.07 |
| 100% | 78.83 | 79.95 | 79.33 | 80.38 | 72.17 | 82.24 | 80.63 | 83.21 | 75.16 | 92.03 |

**Table S10. Performance evaluation of six datasets for countries (unions) with the most monitors from 2006 to 2015 against TOAR-II observations of OSDMA8. Number is the number of the TOAR-II monitor stations in each country. Density (per km<sup>2</sup>) is the density of the TOAR-II monitors in each country based on land area. Estimate is the average of the grid estimates for each dataset at the TOAR-II monitor locations in each country. Linear regression R<sup>2</sup> and root mean squared error (RMSE) against TOAR-II observations in each country are presented. Country names are in ISO 3166-1 alpha-3, United States of America (USA), Japan (JPN), China (CHN), South Korea (KOR), Canada (CAN), United Kingdom of Great Britain and Northern Ireland (GBR). EU-27 includes Austria, Belgium, Bulgaria, Croatia, Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain, Sweden. “Others” is all other countries apart from those listed. The order of countries or regions is determined based on the number of ground monitoring stations.**

|       |                |          |          |          |          |          |          |          |
|-------|----------------|----------|----------|----------|----------|----------|----------|----------|
| 2006  | Country        | EU-27    | USA      | JPN      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 1755     | 717      | 643      | 226      | 198      | 82       | 94       |
|       | Density        | 4.39E-04 | 7.84E-05 | 1.76E-03 | 2.32E-03 | 2.25E-05 | 3.39E-04 | 4.53E-06 |
|       | TOAR           | 46.66    | 51.15    | 49.33    | 41.21    | 39.16    | 39.38    | 44.56    |
| BME   | Estimate       | 46.36    | 51.31    | 47.92    | 41.52    | 39.05    | 38.99    | 44.06    |
|       | R <sup>2</sup> | 0.55     | 0.77     | 0.21     | 0.11     | 0.73     | 0.46     | 0.63     |
|       | RMSE           | 4.90     | 3.67     | 5.00     | 6.97     | 2.67     | 4.38     | 6.24     |
| NJML  | Estimate       | 58.69    | 63.74    | 50.52    | 46.10    | 50.44    | 49.54    | 54.29    |
|       | R <sup>2</sup> | 0.33     | 0.47     | 0.01     | 0.02     | 0.45     | 0.16     | 0.46     |
|       | RMSE           | 13.47    | 13.88    | 6.35     | 8.64     | 11.98    | 11.51    | 12.45    |
| UKML  | Estimate       | 55.43    | 57.53    | 62.36    | 65.09    | 48.08    | 48.86    | 50.29    |
|       | R <sup>2</sup> | 0.11     | 0.26     | 0.09     | 0.00     | 0.27     | 0.09     | 0.47     |
|       | RMSE           | 11.29    | 9.19     | 13.94    | 24.87    | 10.34    | 11.12    | 9.94     |
| CAMS  | Estimate       | 44.14    | 54.13    | 42.56    | 49.61    | 40.28    | 31.65    | 43.49    |
|       | R <sup>2</sup> | 0.17     | 0.31     | 0.00     | 0.00     | 0.20     | 0.00     | 0.30     |
|       | RMSE           | 7.59     | 7.93     | 9.41     | 11.00    | 4.93     | 10.03    | 8.79     |
| GEOS  | Estimate       | 50.17    | 53.00    | 56.77    | 62.23    | 45.57    | 43.95    | 46.08    |
|       | R <sup>2</sup> | 0.15     | 0.28     | 0.03     | 0.01     | 0.24     | 0.04     | 0.44     |
|       | RMSE           | 7.62     | 6.70     | 9.21     | 22.12    | 7.83     | 7.34     | 7.89     |
| TCR-2 | Estimate       | 54.17    | 59.30    | 58.89    | 63.89    | 47.52    | 45.29    | 47.86    |
|       | R <sup>2</sup> | 0.16     | 0.23     | 0.01     | 0.03     | 0.23     | 0.04     | 0.35     |
|       | RMSE           | 10.20    | 10.95    | 11.94    | 24.02    | 9.83     | 8.37     | 10.07    |
| 2007  | Country        | EU-27    | JPN      | USA      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 1840     | 751      | 745      | 237      | 197      | 50       | 95       |
|       | Density        | 4.67E-04 | 2.06E-03 | 8.14E-05 | 2.43E-03 | 2.24E-05 | 2.07E-04 | 4.18E-06 |
|       | TOAR           | 44.01    | 51.22    | 50.23    | 42.83    | 38.13    | 35.36    | 42.73    |
| BME   | Estimate       | 43.80    | 50.07    | 50.31    | 41.63    | 37.87    | 34.81    | 41.00    |
|       | R <sup>2</sup> | 0.58     | 0.28     | 0.81     | 0.26     | 0.83     | 0.30     | 0.70     |
|       | RMSE           | 4.46     | 5.28     | 3.40     | 6.58     | 2.32     | 4.65     | 6.17     |
| NJML  | Estimate       | 56.16    | 53.14    | 63.01    | 46.84    | 50.59    | 43.61    | 53.50    |
|       | R <sup>2</sup> | 0.43     | 0.01     | 0.54     | 0.06     | 0.60     | 0.11     | 0.52     |
|       | RMSE           | 13.28    | 7.11     | 13.99    | 8.24     | 13.10    | 9.80     | 13.29    |
| UKML  | Estimate       | 54.81    | 61.35    | 57.68    | 65.75    | 48.38    | 47.98    | 49.42    |
|       | R <sup>2</sup> | 0.21     | 0.03     | 0.40     | 0.01     | 0.42     | 0.03     | 0.51     |
|       | RMSE           | 12.57    | 11.77    | 9.64     | 24.18    | 11.27    | 13.93    | 10.55    |
| CAMS  | Estimate       | 41.22    | 44.96    | 52.41    | 51.29    | 39.42    | 30.88    | 41.48    |
|       | R <sup>2</sup> | 0.31     | 0.02     | 0.45     | 0.00     | 0.42     | 0.02     | 0.48     |
|       | RMSE           | 6.84     | 9.08     | 7.21     | 11.38    | 4.41     | 7.55     | 7.92     |
| GEOS  | Estimate       | 47.89    | 57.09    | 51.39    | 62.07    | 44.93    | 41.19    | 44.32    |
|       | R <sup>2</sup> | 0.24     | 0.08     | 0.36     | 0.04     | 0.55     | 0.02     | 0.50     |
|       | RMSE           | 7.18     | 8.30     | 6.40     | 20.59    | 7.89     | 8.01     | 7.82     |
| TCR-2 | Estimate       | 52.38    | 58.75    | 58.43    | 61.19    | 47.30    | 42.86    | 47.46    |

|       |                |          |          |          |          |          |          |          |
|-------|----------------|----------|----------|----------|----------|----------|----------|----------|
|       | R <sup>2</sup> | 0.29     | 0.03     | 0.42     | 0.05     | 0.43     | 0.00     | 0.40     |
|       | RMSE           | 10.25    | 10.59    | 10.34    | 20.01    | 10.35    | 9.45     | 10.20    |
| 2008  | Country        | EU-27    | JPN      | USA      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 1962     | 808      | 746      | 255      | 197      | 87       | 113      |
|       | Density        | 4.91E-04 | 2.22E-03 | 8.16E-05 | 2.61E-03 | 2.24E-05 | 3.60E-04 | 4.74E-06 |
|       | TOAR           | 44.20    | 51.58    | 49.18    | 45.17    | 39.11    | 37.61    | 41.39    |
| BME   | Estimate       | 43.89    | 51.27    | 49.31    | 44.21    | 38.81    | 37.08    | 40.51    |
|       | R <sup>2</sup> | 0.47     | 0.34     | 0.85     | 0.19     | 0.80     | 0.42     | 0.75     |
|       | RMSE           | 4.56     | 4.66     | 3.16     | 6.37     | 2.28     | 4.38     | 5.47     |
| NJML  | Estimate       | 54.64    | 52.46    | 61.23    | 49.09    | 49.21    | 45.07    | 50.87    |
|       | R <sup>2</sup> | 0.33     | 0.04     | 0.59     | 0.03     | 0.46     | 0.15     | 0.56     |
|       | RMSE           | 11.70    | 6.09     | 13.17    | 7.84     | 10.81    | 9.12     | 11.92    |
| UKML  | Estimate       | 54.40    | 63.81    | 57.53    | 69.97    | 48.23    | 47.27    | 47.53    |
|       | R <sup>2</sup> | 0.17     | 0.13     | 0.45     | 0.02     | 0.25     | 0.10     | 0.53     |
|       | RMSE           | 11.92    | 13.32    | 10.28    | 25.90    | 10.49    | 11.15    | 9.94     |
| CAMS  | Estimate       | 38.41    | 44.82    | 46.32    | 51.48    | 37.82    | 32.06    | 38.30    |
|       | R <sup>2</sup> | 0.19     | 0.02     | 0.47     | 0.01     | 0.12     | 0.02     | 0.41     |
|       | RMSE           | 8.15     | 9.22     | 6.58     | 9.70     | 4.94     | 8.21     | 8.83     |
| GEOS  | Estimate       | 48.38    | 57.33    | 50.83    | 63.49    | 44.64    | 43.31    | 42.95    |
|       | R <sup>2</sup> | 0.17     | 0.06     | 0.42     | 0.00     | 0.26     | 0.04     | 0.57     |
|       | RMSE           | 7.10     | 7.96     | 6.41     | 19.55    | 7.07     | 7.97     | 7.24     |
| TCR-2 | Estimate       | 52.12    | 55.24    | 57.08    | 62.36    | 45.28    | 43.78    | 44.57    |
|       | R <sup>2</sup> | 0.18     | 0.03     | 0.30     | 0.07     | 0.29     | 0.00     | 0.41     |
|       | RMSE           | 10.11    | 8.26     | 10.62    | 18.97    | 8.05     | 8.83     | 9.75     |
| 2009  | Country        | EU-27    | JPN      | USA      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 2056     | 1115     | 767      | 262      | 195      | 87       | 128      |
|       | Density        | 5.14E-04 | 3.06E-03 | 8.38E-05 | 2.68E-03 | 2.22E-05 | 3.60E-04 | 5.15E-06 |
|       | TOAR           | 44.34    | 51.37    | 46.77    | 48.14    | 38.08    | 35.65    | 40.39    |
| BME   | Estimate       | 44.10    | 50.16    | 47.07    | 47.77    | 37.87    | 34.82    | 39.21    |
|       | R <sup>2</sup> | 0.54     | 0.11     | 0.81     | 0.18     | 0.70     | 0.50     | 0.66     |
|       | RMSE           | 4.55     | 6.08     | 3.11     | 6.01     | 2.30     | 4.48     | 6.04     |
| NJML  | Estimate       | 55.24    | 51.31    | 58.31    | 51.09    | 48.47    | 42.54    | 50.48    |
|       | R <sup>2</sup> | 0.39     | 0.00     | 0.58     | 0.02     | 0.34     | 0.28     | 0.53     |
|       | RMSE           | 12.14    | 6.21     | 12.54    | 7.17     | 11.02    | 8.65     | 12.34    |
| UKML  | Estimate       | 54.28    | 60.79    | 56.41    | 64.69    | 46.28    | 47.72    | 45.93    |
|       | R <sup>2</sup> | 0.27     | 0.02     | 0.39     | 0.08     | 0.15     | 0.15     | 0.57     |
|       | RMSE           | 11.52    | 10.89    | 11.15    | 17.72    | 9.77     | 13.35    | 9.13     |
| CAMS  | Estimate       | 42.58    | 43.14    | 48.94    | 58.00    | 38.04    | 40.35    | 29.96    |
|       | R <sup>2</sup> | 0.31     | 0.08     | 0.46     | 0.01     | 0.01     | 0.41     | 0.02     |
|       | RMSE           | 6.41     | 10.11    | 6.38     | 13.47    | 4.46     | 7.88     | 8.55     |
| GEOS  | Estimate       | 49.32    | 57.59    | 49.88    | 64.30    | 45.04    | 42.38    | 42.62    |
|       | R <sup>2</sup> | 0.27     | 0.10     | 0.32     | 0.06     | 0.25     | 0.09     | 0.54     |
|       | RMSE           | 7.58     | 8.19     | 6.59     | 17.38    | 7.84     | 8.91     | 7.44     |
| TCR-2 | Estimate       | 53.34    | 58.42    | 56.27    | 64.85    | 47.27    | 43.53    | 46.77    |
|       | R <sup>2</sup> | 0.26     | 0.00     | 0.19     | 0.02     | 0.19     | 0.01     | 0.40     |
|       | RMSE           | 10.78    | 10.20    | 11.74    | 18.28    | 10.34    | 10.06    | 10.89    |
| 2010  | Country        | EU-27    | JPN      | USA      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 2119     | 1057     | 799      | 265      | 200      | 87       | 121      |
|       | Density        | 5.30E-04 | 2.90E-03 | 8.73E-05 | 2.72E-03 | 2.28E-05 | 3.60E-04 | 5.07E-06 |
|       | TOAR           | 44.06    | 43.52    | 48.31    | 43.13    | 38.60    | 34.09    | 40.30    |
| BME   | Estimate       | 44.09    | 48.23    | 48.27    | 44.43    | 38.04    | 33.73    | 39.33    |
|       | R <sup>2</sup> | 0.57     | 0.13     | 0.79     | 0.14     | 0.76     | 0.44     | 0.57     |
|       | RMSE           | 4.26     | 6.81     | 3.30     | 6.09     | 2.42     | 3.88     | 6.12     |

|       |                |          |          |          |          |          |          |          |
|-------|----------------|----------|----------|----------|----------|----------|----------|----------|
| NJML  | Estimate       | 55.51    | 50.45    | 59.06    | 46.96    | 48.53    | 41.92    | 51.02    |
|       | R <sup>2</sup> | 0.40     | 0.01     | 0.54     | 0.05     | 0.56     | 0.15     | 0.49     |
|       | RMSE           | 12.59    | 9.06     | 11.87    | 7.20     | 10.48    | 9.18     | 12.81    |
| UKML  | Estimate       | 54.16    | 58.93    | 55.80    | 63.83    | 46.29    | 47.68    | 46.64    |
|       | R <sup>2</sup> | 0.27     | 0.02     | 0.43     | 0.00     | 0.29     | 0.26     | 0.46     |
|       | RMSE           | 11.58    | 16.08    | 9.22     | 21.64    | 9.15     | 14.30    | 9.99     |
| CAMS  | Estimate       | 37.68    | 43.72    | 45.81    | 49.13    | 38.65    | 29.04    | 37.66    |
|       | R <sup>2</sup> | 0.18     | 0.00     | 0.43     | 0.00     | 0.23     | 0.05     | 0.17     |
|       | RMSE           | 8.85     | 5.19     | 5.98     | 9.28     | 4.24     | 7.25     | 9.26     |
| GEOS  | Estimate       | 48.87    | 55.80    | 50.48    | 60.66    | 46.28    | 41.62    | 42.99    |
|       | R <sup>2</sup> | 0.21     | 0.00     | 0.26     | 0.00     | 0.39     | 0.06     | 0.50     |
|       | RMSE           | 7.56     | 13.17    | 6.54     | 18.63    | 8.57     | 9.05     | 7.06     |
| TCR-2 | Estimate       | 51.63    | 56.58    | 57.16    | 60.62    | 46.53    | 40.62    | 44.89    |
|       | R <sup>2</sup> | 0.22     | 0.00     | 0.27     | 0.08     | 0.44     | 0.00     | 0.40     |
|       | RMSE           | 9.79     | 14.95    | 11.28    | 18.95    | 9.07     | 8.58     | 9.61     |
| 2011  | Country        | EU-27    | JPN      | USA      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 2142     | 822      | 741      | 277      | 205      | 93       | 127      |
|       | Density        | 5.36E-04 | 2.26E-03 | 8.10E-05 | 2.84E-03 | 2.33E-05 | 3.84E-04 | 3.16E-06 |
|       | TOAR           | 44.57    | 41.89    | 49.27    | 43.93    | 38.31    | 34.76    | 39.92    |
| BME   | Estimate       | 44.56    | 46.05    | 49.10    | 44.47    | 38.04    | 34.69    | 39.90    |
|       | R <sup>2</sup> | 0.60     | 0.15     | 0.86     | 0.19     | 0.79     | 0.38     | 0.64     |
|       | RMSE           | 4.36     | 6.24     | 2.82     | 5.92     | 1.98     | 3.80     | 5.69     |
| NJML  | Estimate       | 55.86    | 47.81    | 59.91    | 46.71    | 47.61    | 43.14    | 50.90    |
|       | R <sup>2</sup> | 0.43     | 0.02     | 0.55     | 0.04     | 0.44     | 0.07     | 0.55     |
|       | RMSE           | 12.46    | 8.26     | 11.83    | 7.01     | 9.94     | 9.60     | 12.97    |
| UKML  | Estimate       | 53.73    | 62.96    | 55.48    | 65.20    | 45.66    | 48.90    | 45.91    |
|       | R <sup>2</sup> | 0.31     | 0.00     | 0.42     | 0.04     | 0.12     | 0.07     | 0.60     |
|       | RMSE           | 10.84    | 21.66    | 8.44     | 22.19    | 8.90     | 14.93    | 8.93     |
| CAMS  | Estimate       | 41.55    | 44.45    | 50.60    | 47.45    | 38.45    | 29.95    | 39.94    |
|       | R <sup>2</sup> | 0.30     | 0.02     | 0.38     | 0.01     | 0.21     | 0.04     | 0.37     |
|       | RMSE           | 6.98     | 5.42     | 6.33     | 7.34     | 4.09     | 6.90     | 7.60     |
| GEOS  | Estimate       | 48.72    | 54.45    | 51.75    | 59.52    | 45.27    | 41.83    | 42.37    |
|       | R <sup>2</sup> | 0.26     | 0.01     | 0.29     | 0.01     | 0.23     | 0.06     | 0.62     |
|       | RMSE           | 7.24     | 13.40    | 6.78     | 16.86    | 7.91     | 8.46     | 6.39     |
| TCR-2 | Estimate       | 52.46    | 56.63    | 56.65    | 61.01    | 46.94    | 42.77    | 44.52    |
|       | R <sup>2</sup> | 0.27     | 0.01     | 0.23     | 0.06     | 0.22     | 0.03     | 0.57     |
|       | RMSE           | 9.93     | 16.34    | 10.22    | 18.56    | 10.04    | 9.35     | 8.53     |
| 2012  | Country        | EU-27    | JPN      | KOR      | USA      | CAN      | GBR      | Others   |
|       | Number         | 2527     | 1094     | 286      | 262      | 207      | 91       | 129      |
|       | Density        | 6.32E-04 | 3.00E-03 | 2.93E-03 | 2.86E-05 | 2.36E-05 | 3.76E-04 | 3.14E-06 |
|       | TOAR           | 44.30    | 43.63    | 47.65    | 53.11    | 39.05    | 33.77    | 39.36    |
| BME   | Estimate       | 44.52    | 46.43    | 48.28    | 52.68    | 38.99    | 33.99    | 38.97    |
|       | R <sup>2</sup> | 0.63     | 0.07     | 0.21     | 0.83     | 0.82     | 0.48     | 0.62     |
|       | RMSE           | 4.13     | 5.98     | 5.87     | 4.16     | 2.16     | 3.18     | 5.61     |
| NJML  | Estimate       | 54.85    | 49.44    | 50.26    | 62.71    | 49.42    | 41.59    | 49.85    |
|       | R <sup>2</sup> | 0.43     | 0.01     | 0.00     | 0.56     | 0.55     | 0.13     | 0.53     |
|       | RMSE           | 11.76    | 8.14     | 7.18     | 11.81    | 11.08    | 8.83     | 12.45    |
| UKML  | Estimate       | 54.25    | 61.31    | 65.08    | 57.96    | 45.89    | 46.96    | 45.09    |
|       | R <sup>2</sup> | 0.25     | 0.07     | 0.00     | 0.43     | 0.32     | 0.07     | 0.58     |
|       | RMSE           | 11.60    | 18.31    | 18.65    | 9.01     | 8.09     | 13.96    | 8.69     |
| CAMS  | Estimate       | 38.48    | 43.20    | 48.55    | 47.90    | 38.33    | 30.19    | 37.29    |
|       | R <sup>2</sup> | 0.29     | 0.03     | 0.01     | 0.52     | 0.48     | 0.08     | 0.25     |
|       | RMSE           | 8.20     | 6.10     | 7.16     | 8.96     | 3.92     | 5.73     | 8.55     |

|       |                |          |          |          |          |          |          |          |
|-------|----------------|----------|----------|----------|----------|----------|----------|----------|
| GEOS  | Estimate       | 49.59    | 56.63    | 63.73    | 53.23    | 46.85    | 42.57    | 42.22    |
|       | R <sup>2</sup> | 0.29     | 0.00     | 0.00     | 0.37     | 0.50     | 0.11     | 0.64     |
|       | RMSE           | 7.79     | 14.03    | 17.36    | 7.96     | 8.62     | 9.71     | 6.35     |
| TCR-2 | Estimate       | 52.09    | 55.53    | 63.19    | 56.59    | 46.95    | 42.45    | 44.13    |
|       | R <sup>2</sup> | 0.30     | 0.00     | 0.01     | 0.41     | 0.48     | 0.01     | 0.57     |
|       | RMSE           | 9.76     | 13.85    | 17.19    | 8.45     | 8.94     | 9.82     | 8.58     |
| 2013  | Country        | EU-27    | JPN      | USA      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 2120     | 1046     | 866      | 291      | 197      | 94       | 172      |
|       | Density        | 5.30E-04 | 2.87E-03 | 9.47E-05 | 2.98E-03 | 2.24E-05 | 3.89E-04 | 4.19E-06 |
|       | TOAR           | 44.07    | 44.37    | 47.09    | 49.23    | 38.14    | 36.01    | 39.78    |
| BME   | Estimate       | 45.21    | 48.59    | 46.73    | 49.60    | 37.85    | 35.70    | 40.81    |
|       | R <sup>2</sup> | 0.45     | 0.11     | 0.84     | 0.20     | 0.73     | 0.45     | 0.47     |
|       | RMSE           | 5.19     | 6.14     | 2.81     | 5.67     | 2.38     | 3.33     | 6.92     |
| NJML  | Estimate       | 54.25    | 50.42    | 56.60    | 52.29    | 47.37    | 45.22    | 51.44    |
|       | R <sup>2</sup> | 0.36     | 0.09     | 0.55     | 0.01     | 0.41     | 0.03     | 0.41     |
|       | RMSE           | 11.53    | 7.85     | 10.69    | 6.94     | 9.93     | 10.33    | 13.89    |
| UKML  | Estimate       | 52.83    | 59.96    | 55.72    | 64.72    | 46.44    | 46.55    | 46.94    |
|       | R <sup>2</sup> | 0.24     | 0.04     | 0.40     | 0.01     | 0.22     | 0.07     | 0.51     |
|       | RMSE           | 10.58    | 16.20    | 10.19    | 16.63    | 9.64     | 11.52    | 10.22    |
| CAMS  | Estimate       | 42.93    | 49.30    | 49.82    | 55.17    | 42.08    | 33.99    | 42.04    |
|       | R <sup>2</sup> | 0.28     | 0.03     | 0.35     | 0.01     | 0.11     | 0.00     | 0.27     |
|       | RMSE           | 6.04     | 6.49     | 6.38     | 8.88     | 5.85     | 5.39     | 8.45     |
| GEOS  | Estimate       | 49.32    | 58.12    | 51.02    | 64.31    | 46.28    | 43.42    | 43.98    |
|       | R <sup>2</sup> | 0.21     | 0.02     | 0.39     | 0.00     | 0.31     | 0.01     | 0.54     |
|       | RMSE           | 7.94     | 14.46    | 6.71     | 16.28    | 8.96     | 8.69     | 7.79     |
| TCR-2 | Estimate       | 51.72    | 59.99    | 55.76    | 65.60    | 47.50    | 44.61    | 46.37    |
|       | R <sup>2</sup> | 0.21     | 0.00     | 0.19     | 0.03     | 0.34     | 0.00     | 0.48     |
|       | RMSE           | 9.78     | 17.16    | 11.05    | 17.90    | 10.56    | 9.93     | 10.20    |
| 2014  | Country        | EU-27    | JPN      | USA      | KOR      | CAN      | GBR      | Others   |
|       | Number         | 1787     | 1127     | 856      | 224      | 204      | 94       | 114      |
|       | Density        | 4.47E-04 | 3.09E-03 | 9.36E-05 | 2.30E-03 | 2.32E-05 | 3.89E-04 | 2.76E-06 |
|       | TOAR           | 43.00    | 45.16    | 46.44    | 51.09    | 37.37    | 35.51    | 36.99    |
| BME   | Estimate       | 45.41    | 49.16    | 46.65    | 52.75    | 38.82    | 35.16    | 37.69    |
|       | R <sup>2</sup> | 0.45     | 0.05     | 0.86     | 0.29     | 0.39     | 0.34     | 0.62     |
|       | RMSE           | 5.40     | 6.46     | 2.54     | 6.32     | 3.81     | 3.68     | 5.88     |
| NJML  | Estimate       | 53.42    | 51.01    | 55.75    | 54.87    | 46.45    | 43.07    | 48.33    |
|       | R <sup>2</sup> | 0.40     | 0.04     | 0.61     | 0.01     | 0.28     | 0.11     | 0.38     |
|       | RMSE           | 11.54    | 7.79     | 10.30    | 8.11     | 10.01    | 8.69     | 14.22    |
| UKML  | Estimate       | 53.14    | 62.07    | 55.15    | 67.10    | 46.29    | 46.36    | 41.97    |
|       | R <sup>2</sup> | 0.21     | 0.07     | 0.49     | 0.02     | 0.17     | 0.12     | 0.42     |
|       | RMSE           | 11.65    | 17.76    | 9.97     | 17.75    | 10.02    | 11.83    | 9.23     |
| CAMS  | Estimate       | 42.16    | 46.48    | 49.13    | 56.05    | 40.62    | 32.84    | 40.66    |
|       | R <sup>2</sup> | 0.21     | 0.01     | 0.33     | 0.00     | 0.01     | 0.00     | 0.15     |
|       | RMSE           | 5.87     | 5.10     | 6.45     | 9.88     | 5.76     | 5.66     | 10.70    |
| GEOS  | Estimate       | 49.95    | 57.55    | 50.50    | 65.63    | 44.76    | 43.84    | 40.80    |
|       | R <sup>2</sup> | 0.19     | 0.00     | 0.38     | 0.04     | 0.23     | 0.01     | 0.49     |
|       | RMSE           | 8.97     | 13.52    | 6.76     | 16.16    | 8.27     | 9.61     | 8.09     |
| TCR-2 | Estimate       | 51.78    | 59.36    | 55.35    | 64.89    | 45.87    | 43.89    | 42.19    |
|       | R <sup>2</sup> | 0.17     | 0.00     | 0.23     | 0.09     | 0.27     | 0.00     | 0.41     |
|       | RMSE           | 10.73    | 16.27    | 11.03    | 16.21    | 9.89     | 10.13    | 9.79     |
| 2015  | Country        | EU-27    | CHN      | JPN      | USA      | KOR      | CAN      | Others   |
|       | Number         | 1849     | 1427     | 1123     | 875      | 312      | 204      | 267      |
|       | Density        | 4.63E-04 | 1.52E-04 | 3.08E-03 | 9.57E-05 | 3.20E-03 | 2.32E-05 | 6.10E-06 |

|       |                |       |       |       |       |       |       |       |
|-------|----------------|-------|-------|-------|-------|-------|-------|-------|
|       | TOAR           | 45.64 | 50.39 | 44.03 | 46.41 | 50.40 | 38.58 | 38.24 |
| BME   | Estimate       | 45.41 | 48.66 | 46.34 | 45.50 | 50.26 | 37.28 | 38.36 |
|       | R <sup>2</sup> | 0.71  | 0.52  | 0.02  | 0.86  | 0.13  | 0.50  | 0.49  |
|       | RMSE           | 3.79  | 7.73  | 5.38  | 2.76  | 7.91  | 3.25  | 6.89  |
| NJML  | Estimate       | 55.88 | 53.08 | 49.63 | 55.51 | 54.78 | 47.03 | 47.10 |
|       | R <sup>2</sup> | 0.52  | 0.41  | 0.01  | 0.61  | 0.02  | 0.19  | 0.31  |
|       | RMSE           | 11.41 | 8.81  | 7.49  | 10.11 | 9.38  | 9.52  | 12.24 |
| UKML  | Estimate       | 53.68 | 68.02 | 58.93 | 52.81 | 64.40 | 47.63 | 47.45 |
|       | R <sup>2</sup> | 0.22  | 0.21  | 0.02  | 0.45  | 0.00  | 0.20  | 0.33  |
|       | RMSE           | 10.17 | 20.67 | 15.46 | 8.22  | 16.38 | 9.86  | 12.44 |
| CAMS  | Estimate       | 44.02 | 55.71 | 45.43 | 49.21 | 60.94 | 40.91 | 38.57 |
|       | R <sup>2</sup> | 0.37  | 0.07  | 0.01  | 0.28  | 0.04  | 0.06  | 0.23  |
|       | RMSE           | 5.92  | 12.49 | 4.65  | 6.73  | 15.17 | 4.75  | 8.92  |
| GEOS  | Estimate       | 50.14 | 61.94 | 57.88 | 50.85 | 65.42 | 45.52 | 43.15 |
|       | R <sup>2</sup> | 0.30  | 0.20  | 0.00  | 0.44  | 0.00  | 0.27  | 0.39  |
|       | RMSE           | 7.39  | 15.31 | 14.71 | 6.83  | 17.21 | 7.74  | 9.12  |
| TCR-2 | Estimate       | 52.27 | 67.50 | 59.42 | 54.73 | 67.55 | 46.64 | 45.51 |
|       | R <sup>2</sup> | 0.39  | 0.16  | 0.02  | 0.24  | 0.08  | 0.26  | 0.38  |
|       | RMSE           | 8.66  | 20.33 | 17.55 | 10.50 | 19.65 | 9.35  | 11.02 |

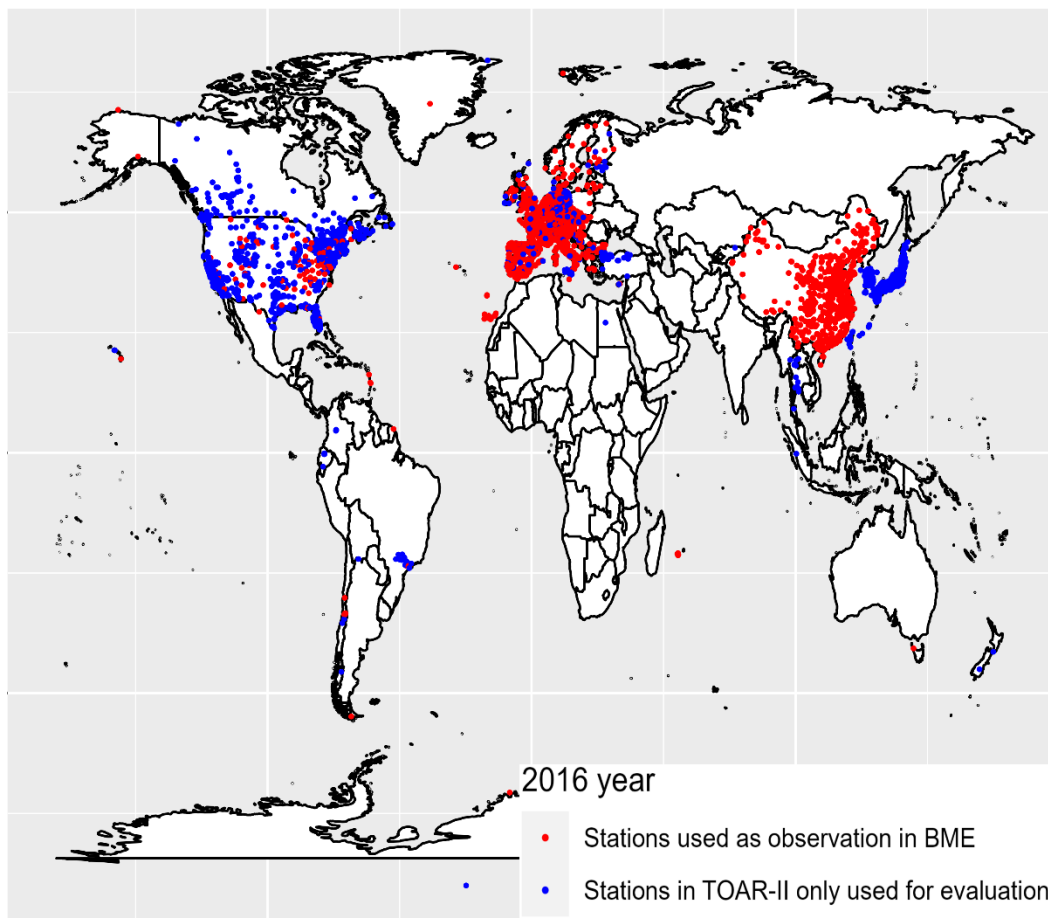
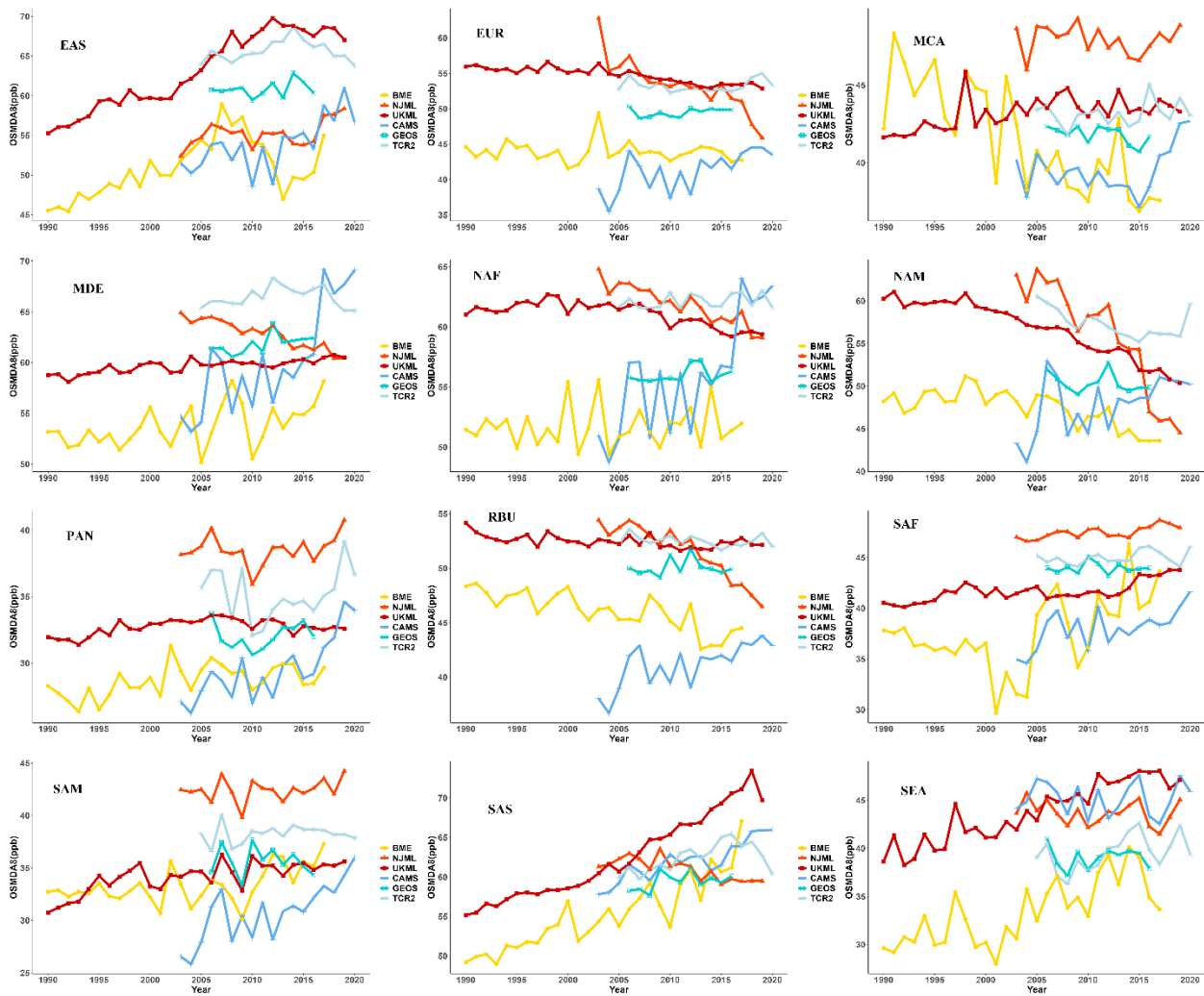


Figure S1. Blue and red dots are TOAR-II January 2016 to March 2017 monitor stations with valid monthly data for more than 11 months (which used for evaluation), red dots are BME inputs from TOAR-I and CNEMC in 2016, blue dots are new TOAR-II stations compared to TOAR-I in 2016 which used for evaluation in Figure S13



**Figure S2. The Yearly trends for six datasets for different regions, where ozone is expressed as population-weighted OSMDA8.**



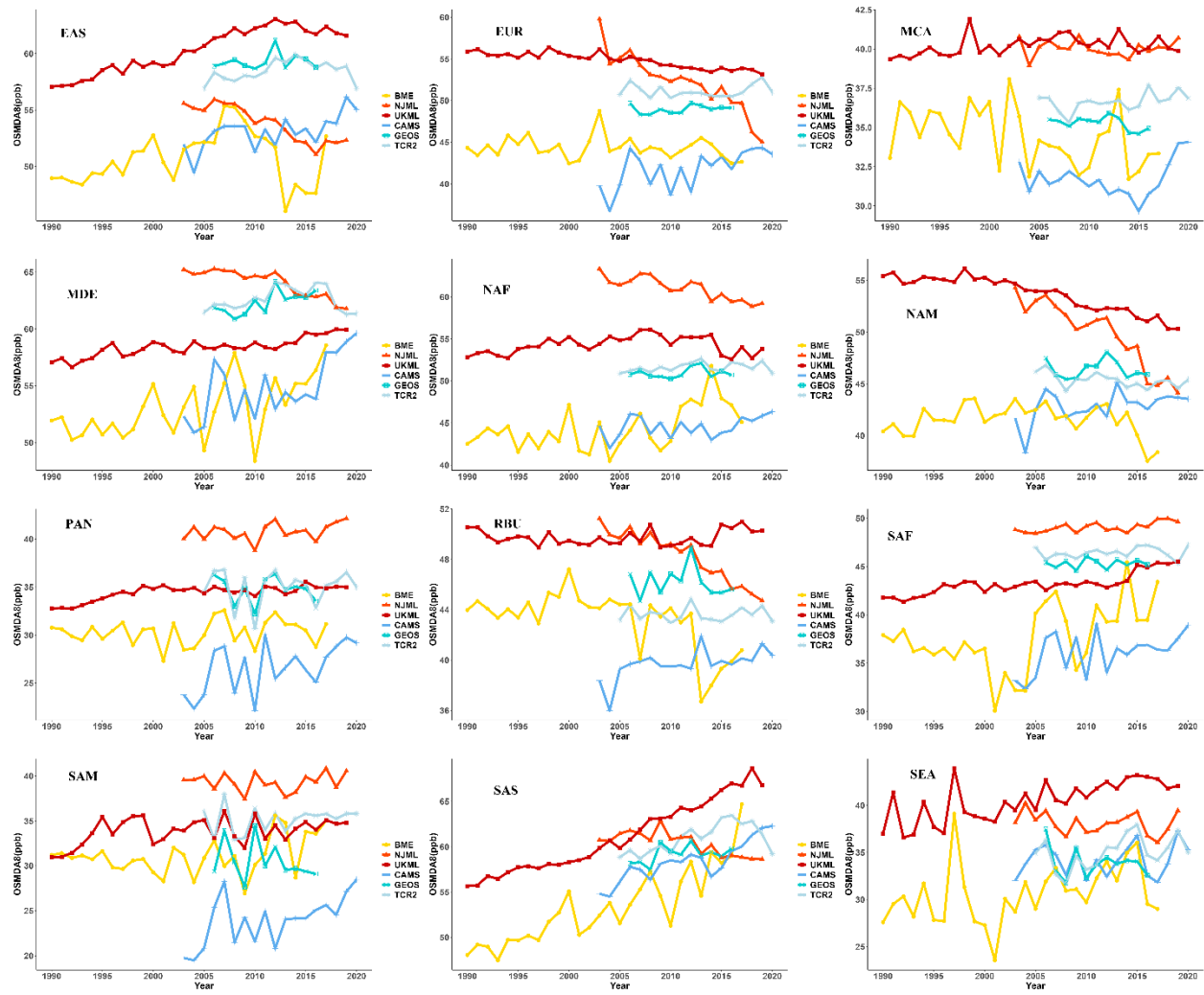


Figure S3. The Yearly trends for six datasets for different regions where ozone is expressed as area-weighted OSMDA8.

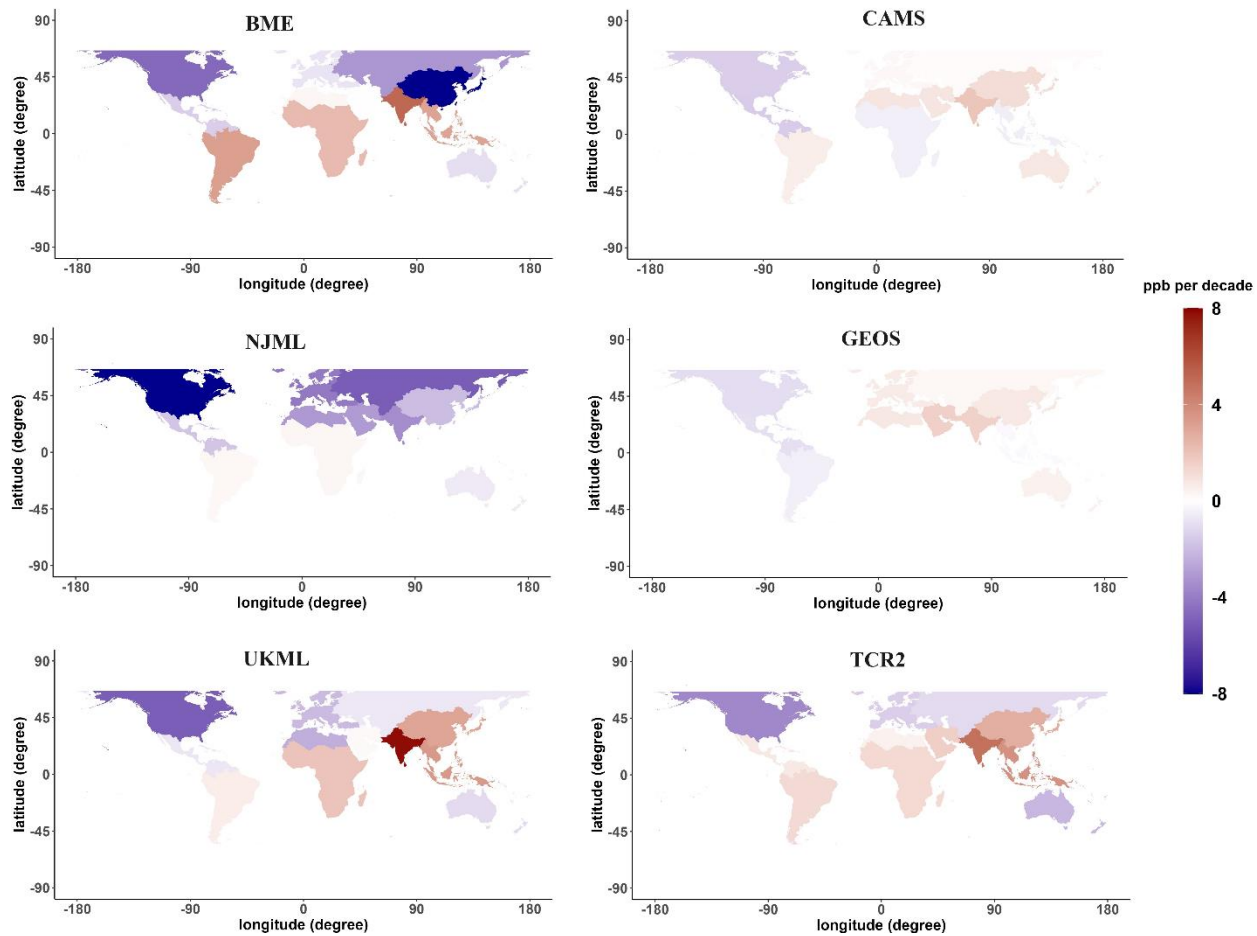
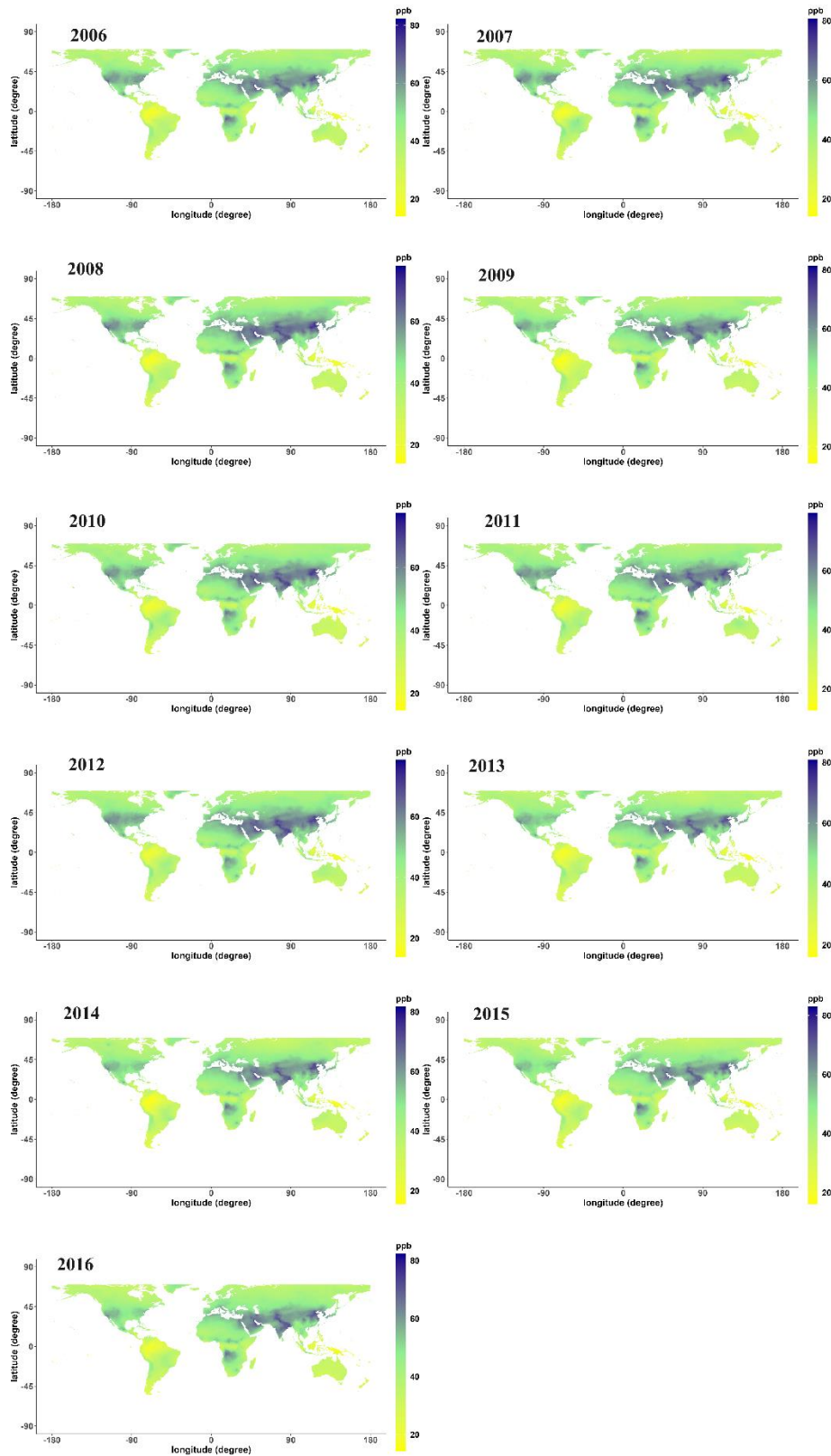
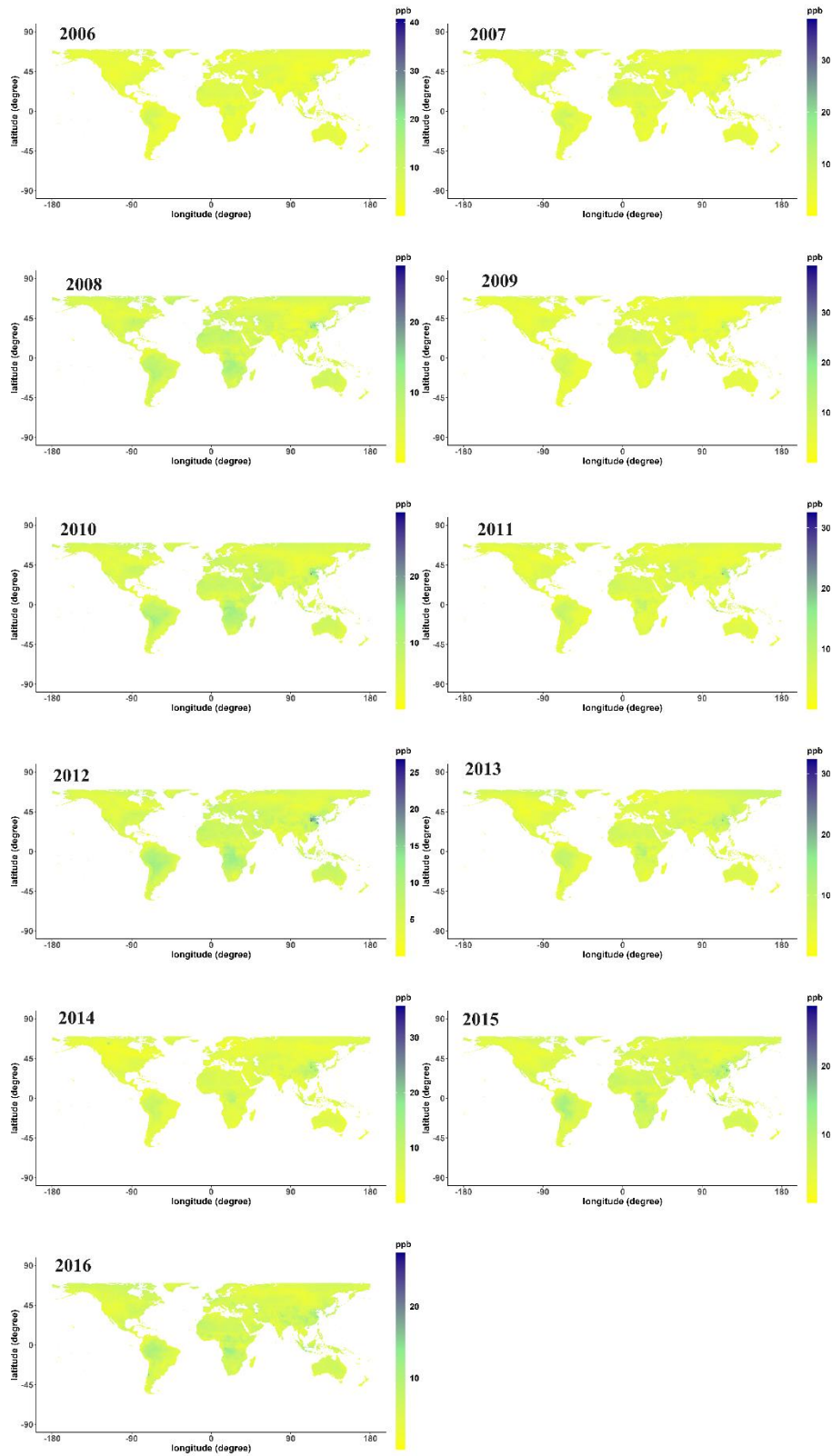


Figure S4. Population weighted ozone (OSMDA8) trends per decade for six datasets, calculated over 2006 to 2016.

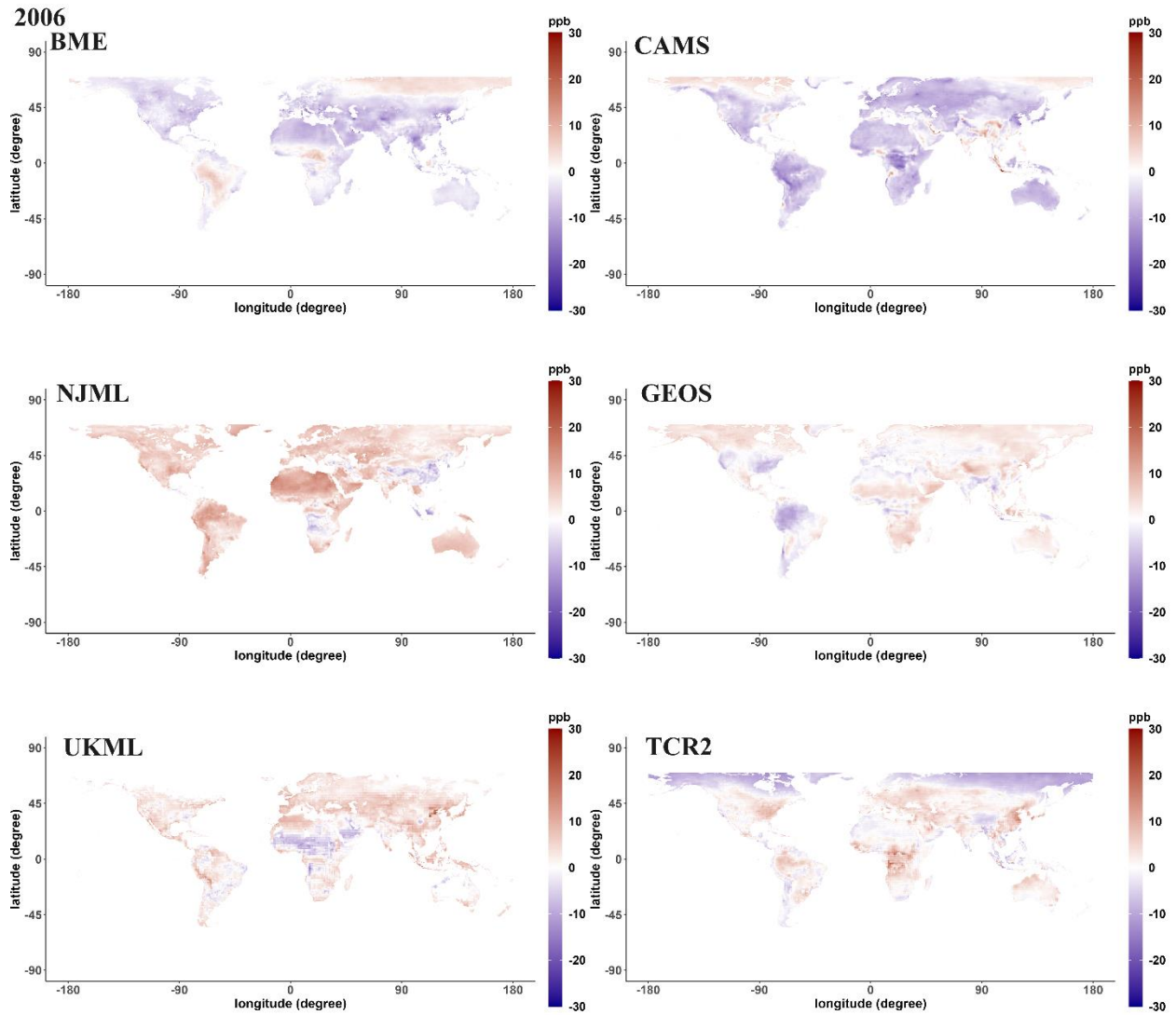


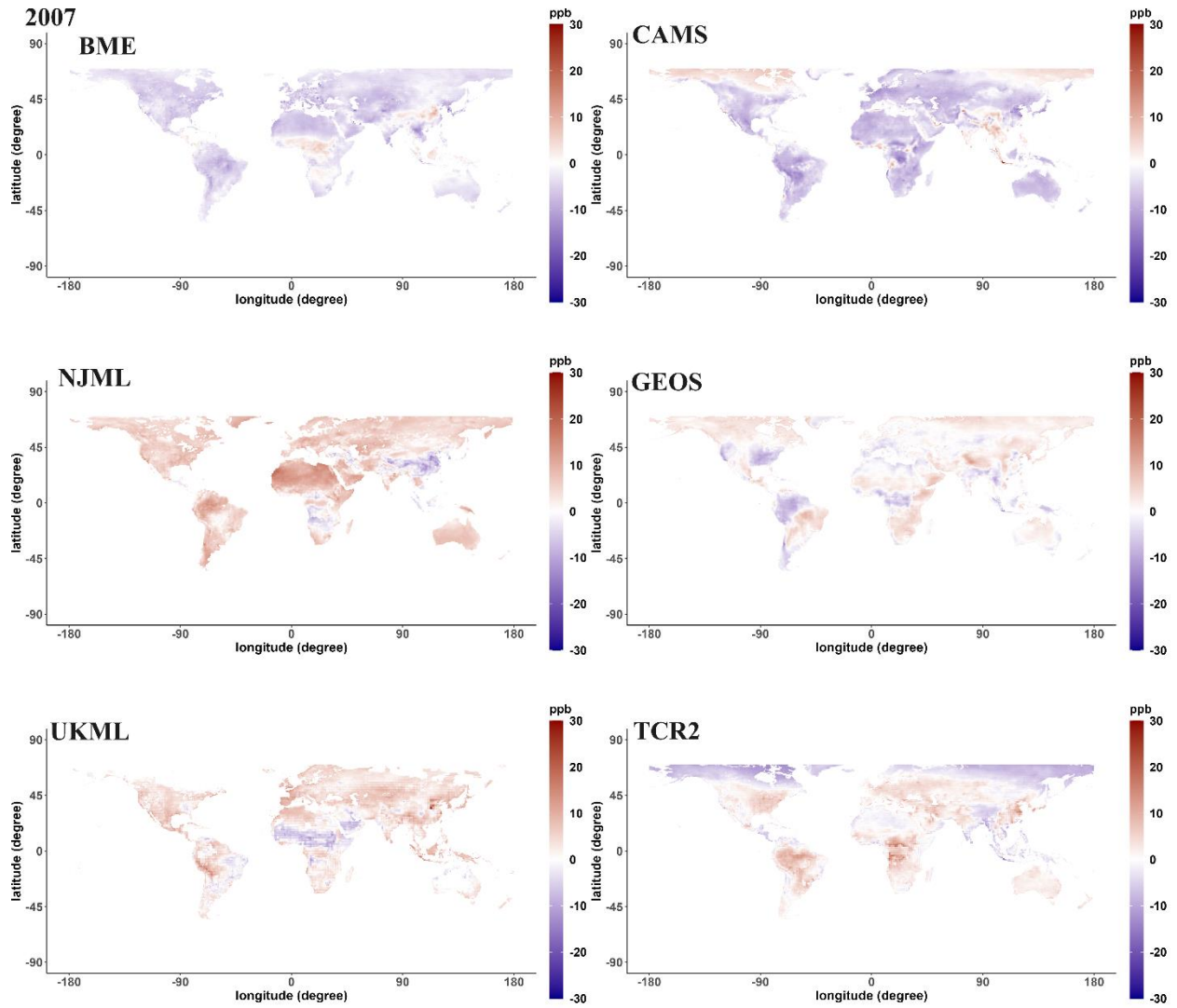
**Figure S5.** Each year ensemble means of six datasets from 2006 to 2016



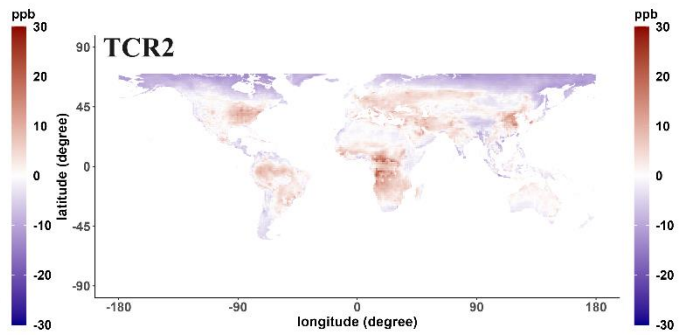
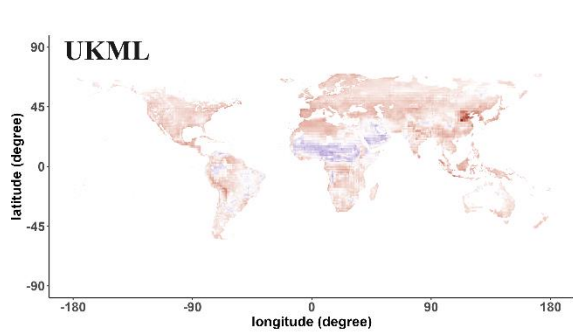
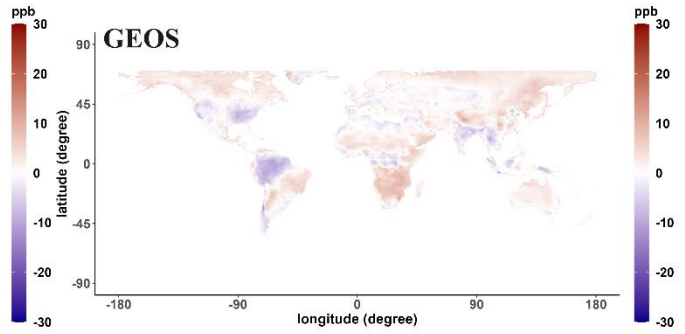
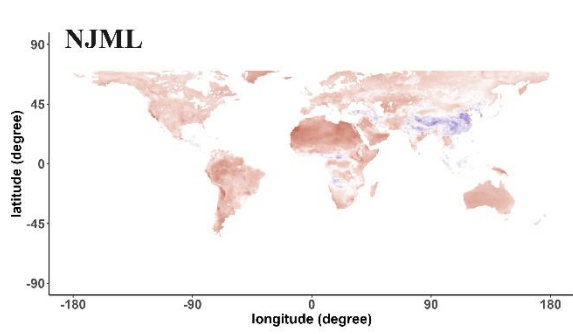
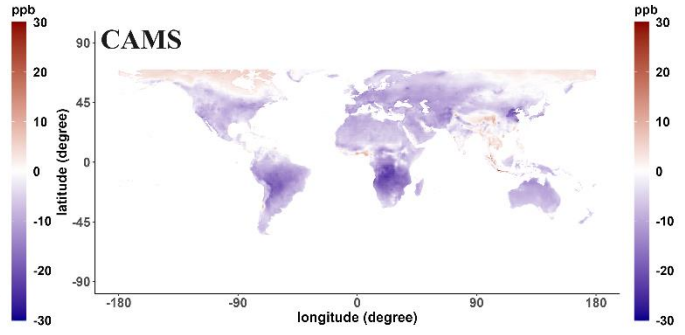
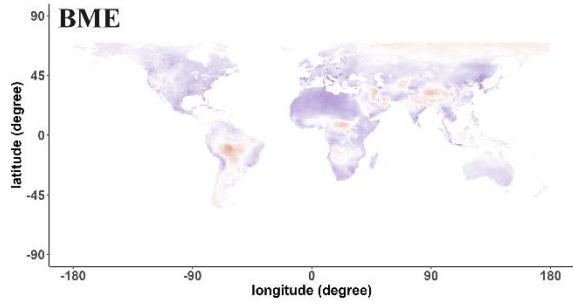
**Figure S6. Each year standard deviations of six datasets from 2006 to 2016**

Figure S7. Each year the difference in each grid cell between the six datasets and the ensemble mean (Figure S5) from 2006 to 2016. Positive values indicate that the estimate of the dataset is higher than the ensemble mean. Negative values indicate that the estimate of the dataset is lower than the ensemble mean of the six datasets.

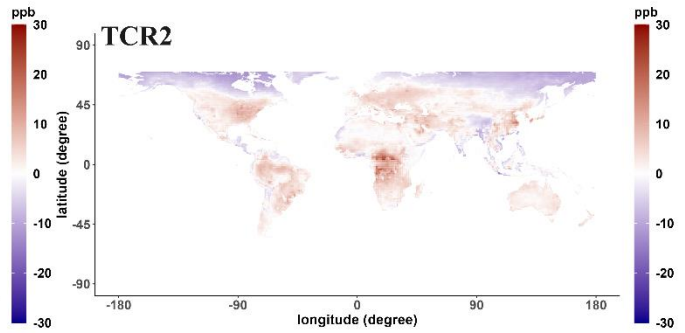
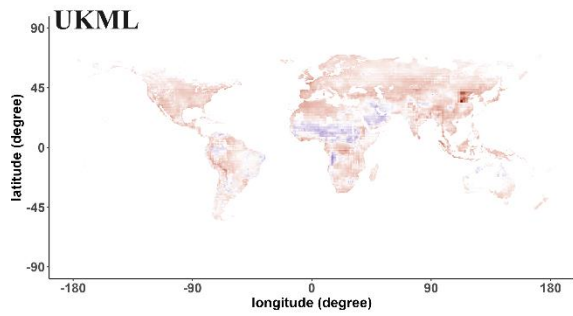
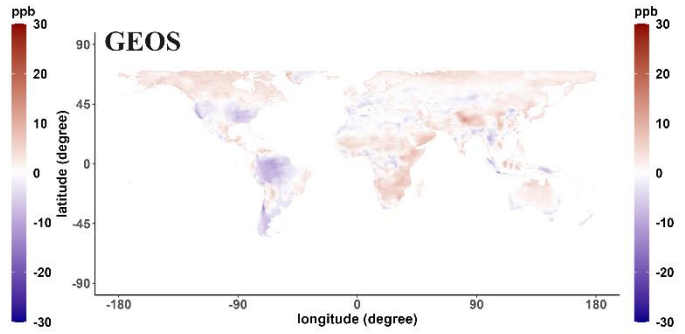
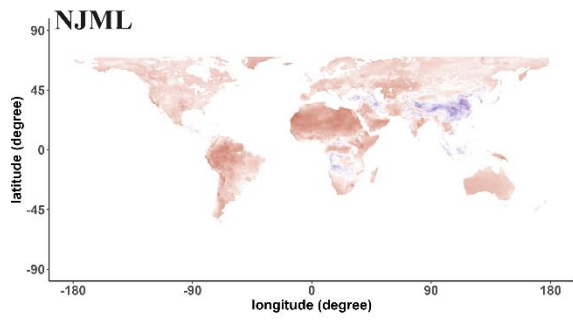
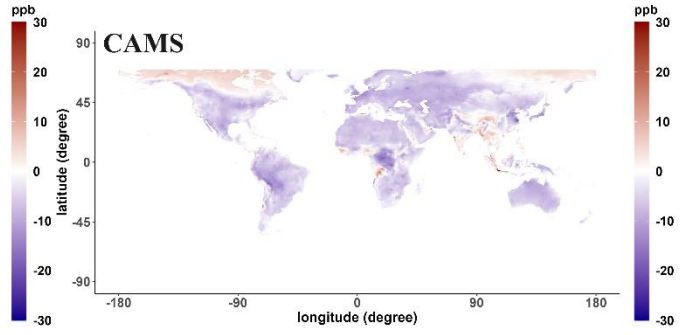
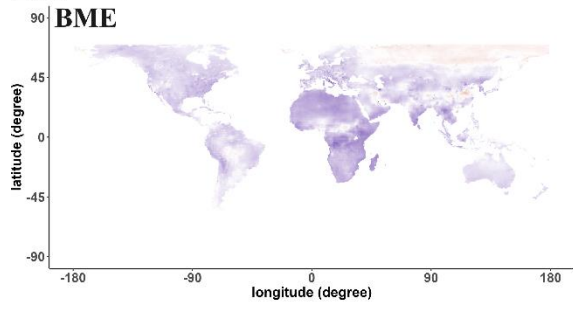




2008

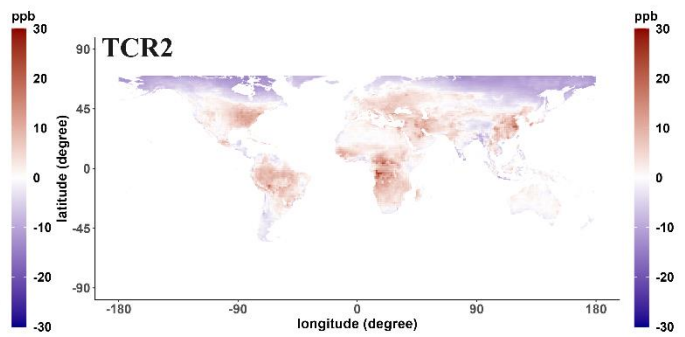
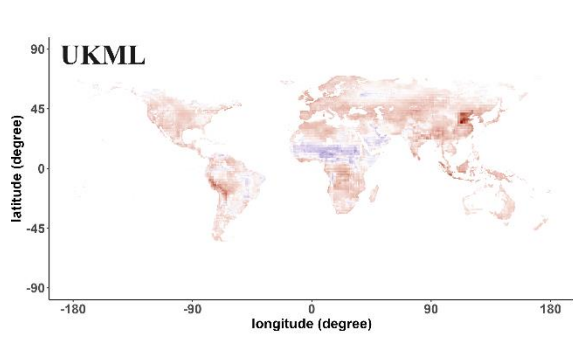
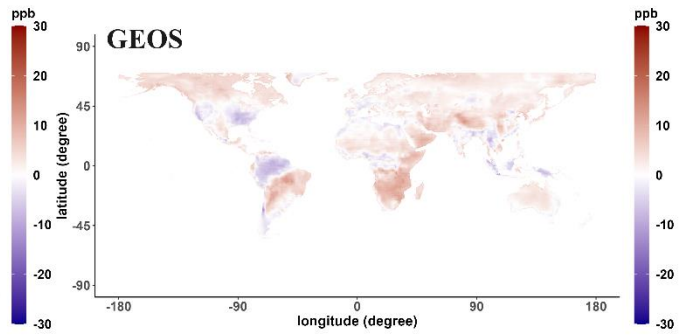
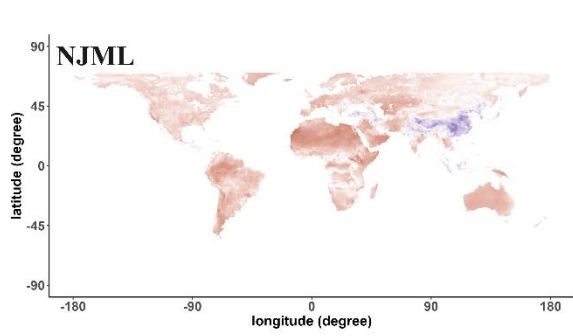
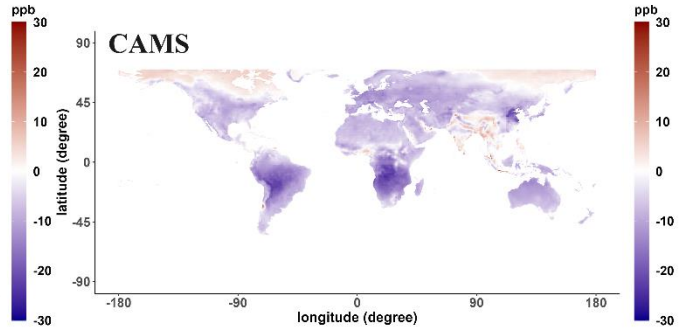
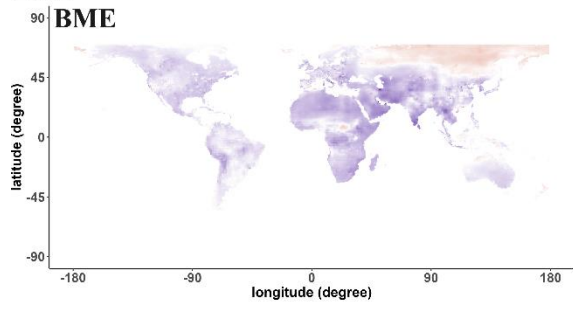


2009

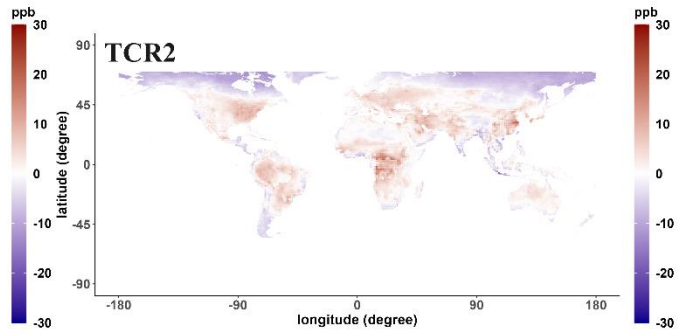
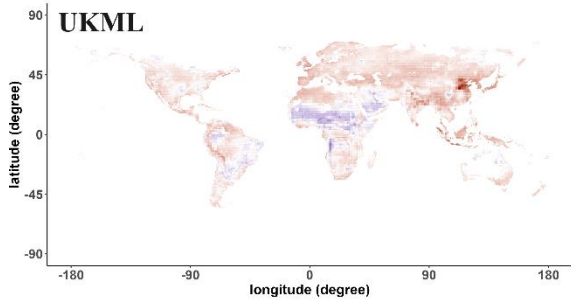
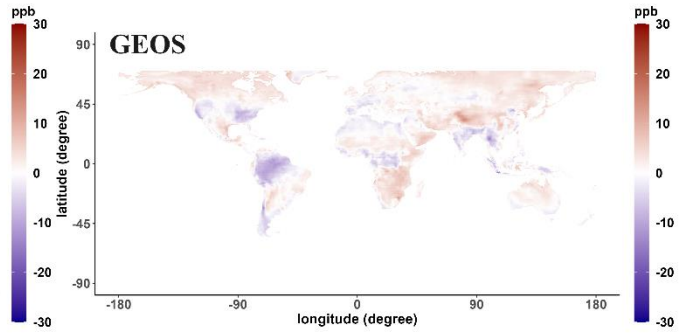
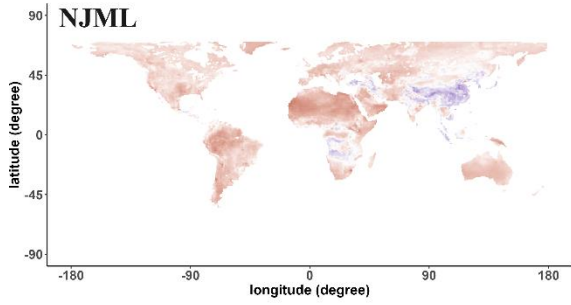
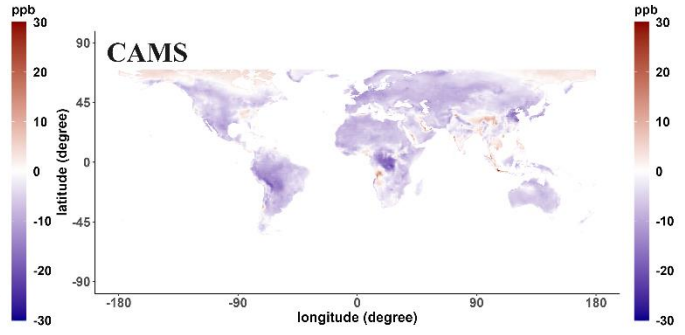
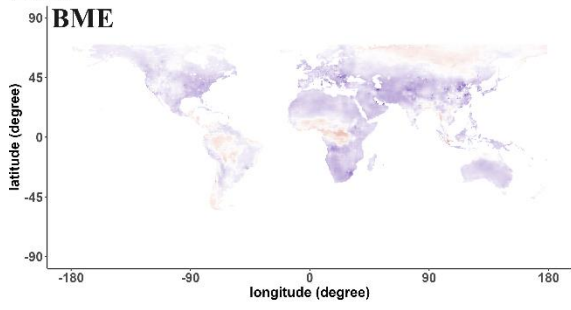




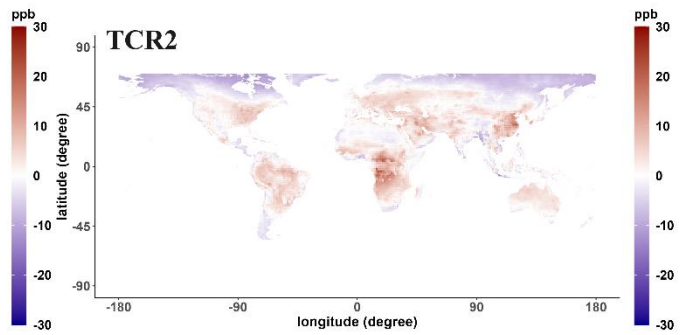
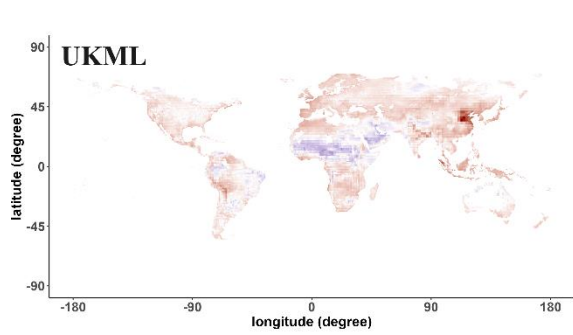
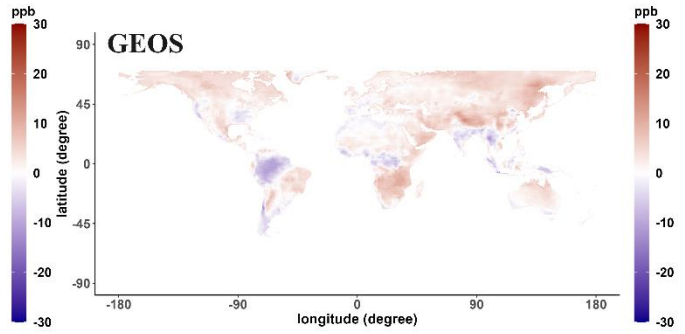
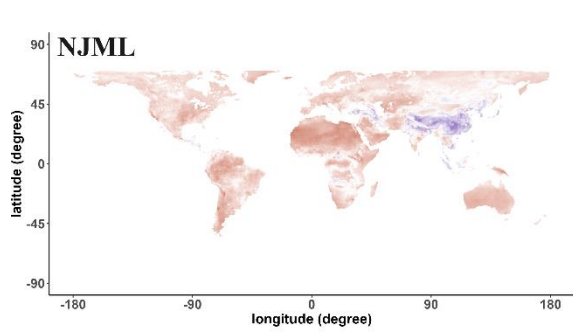
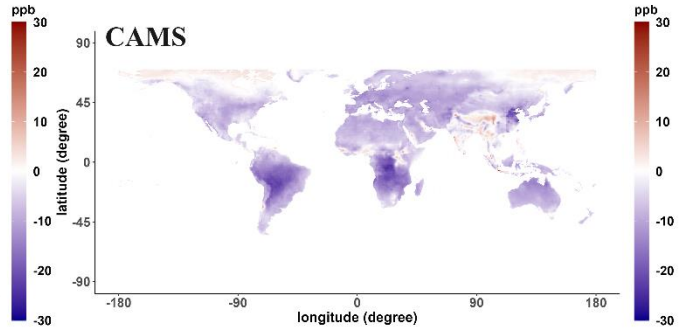
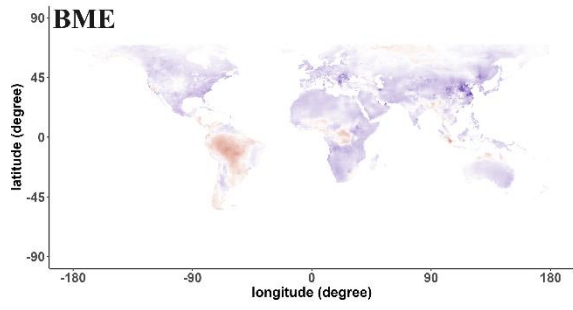
2010



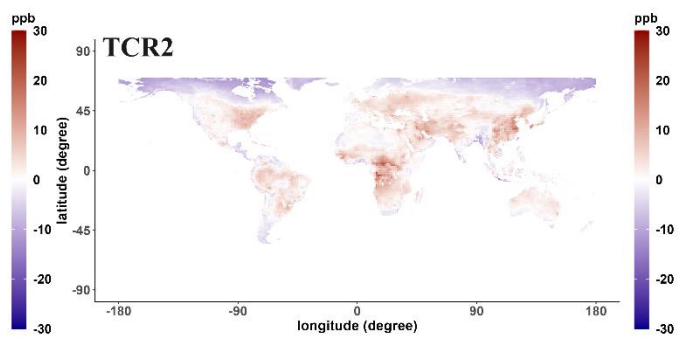
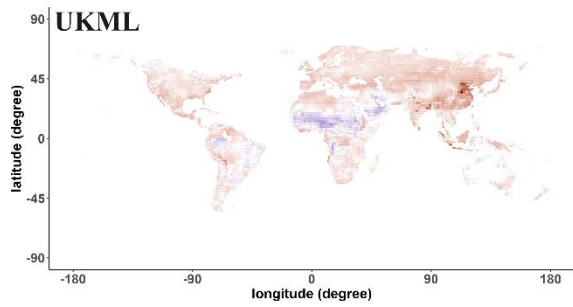
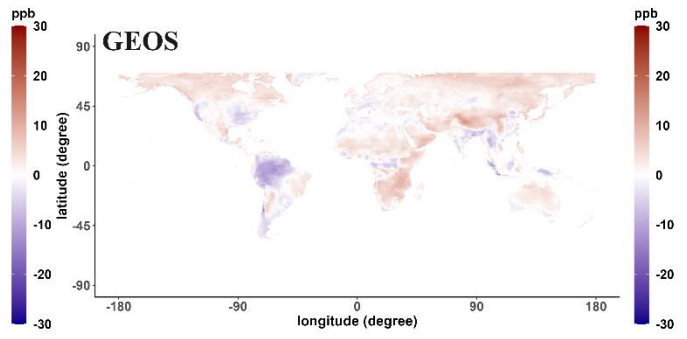
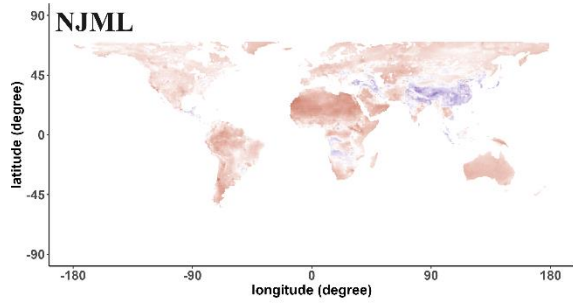
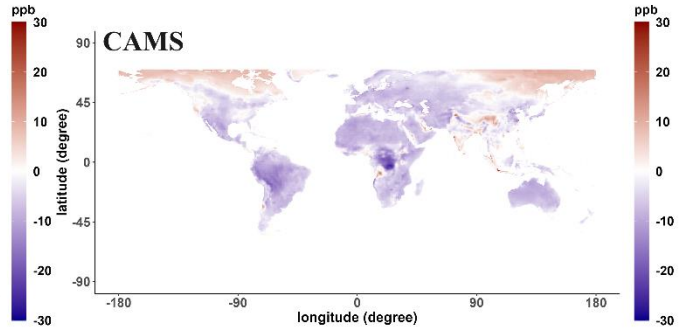
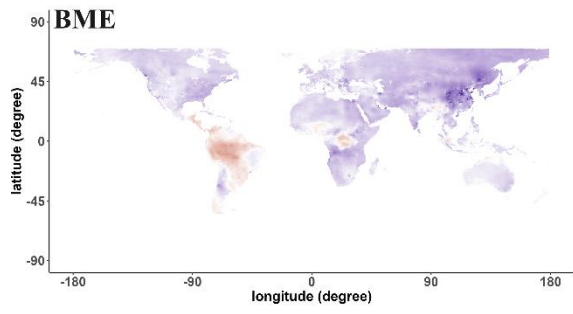
2011



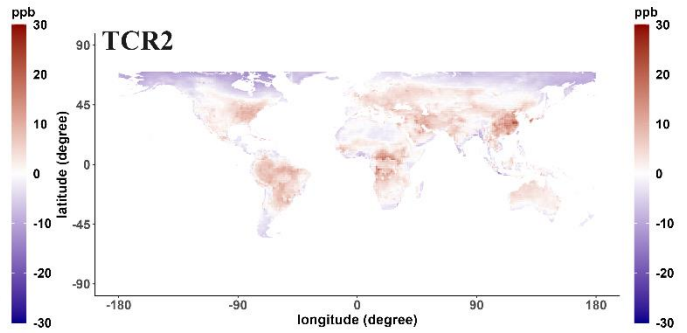
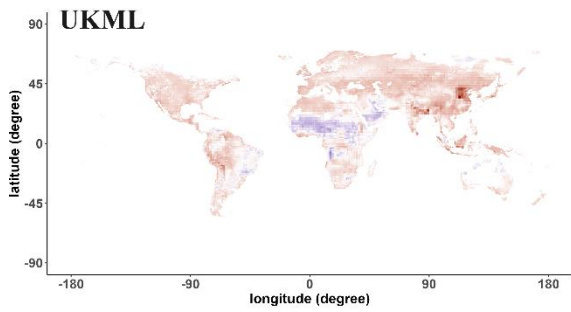
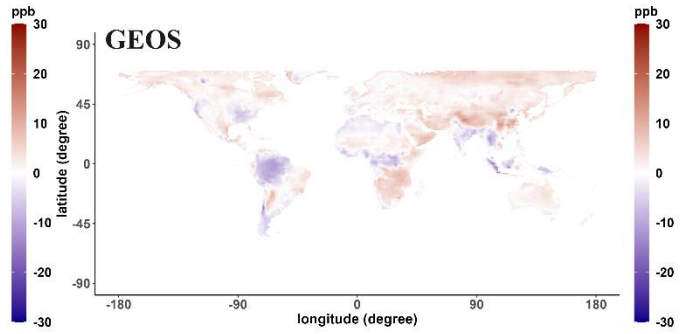
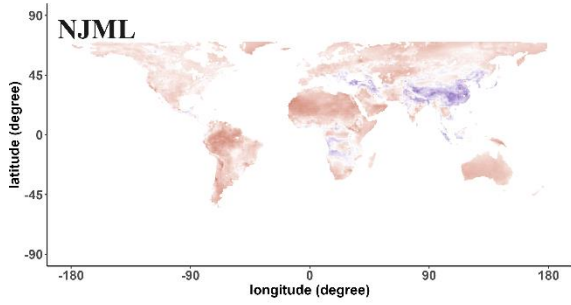
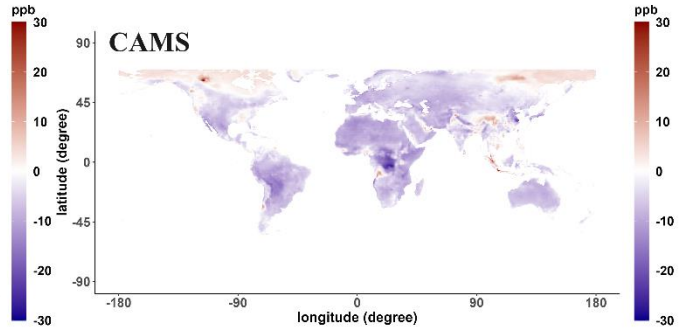
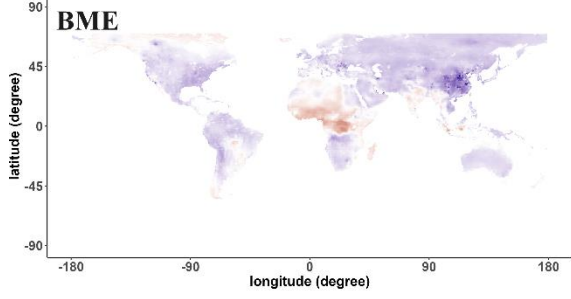
2012



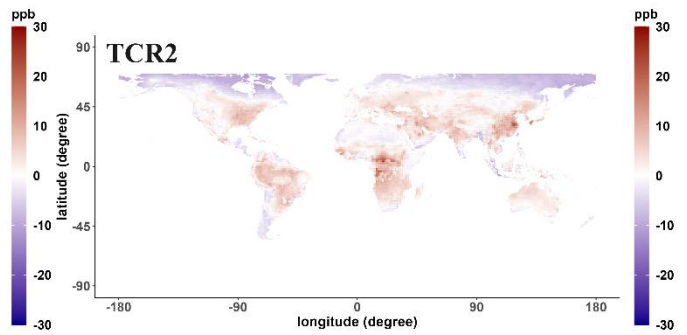
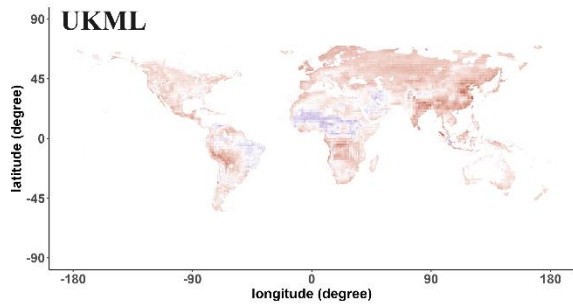
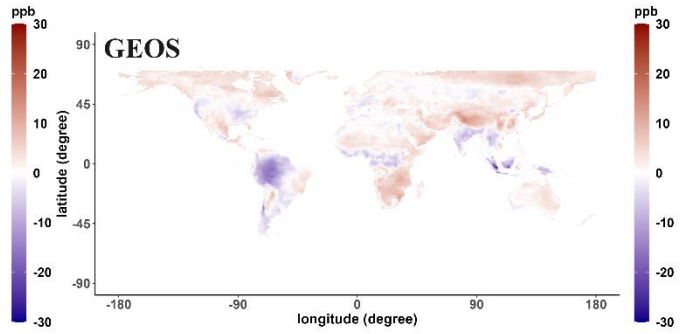
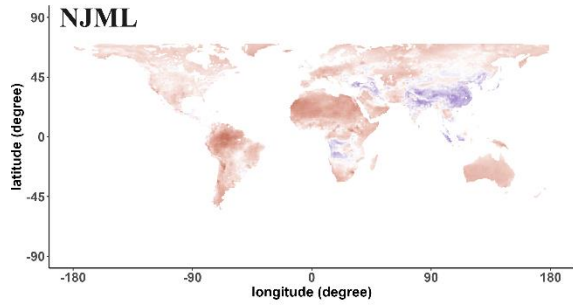
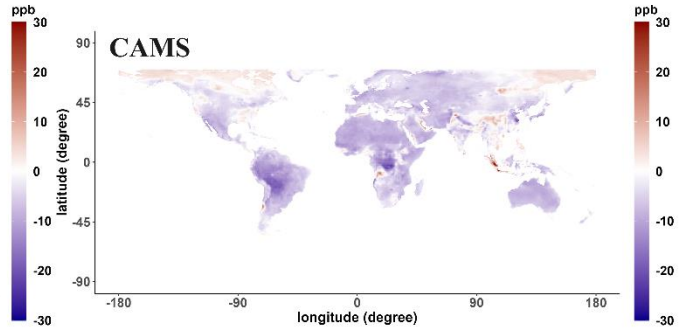
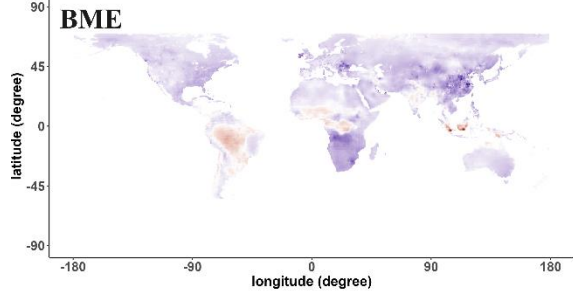
2013



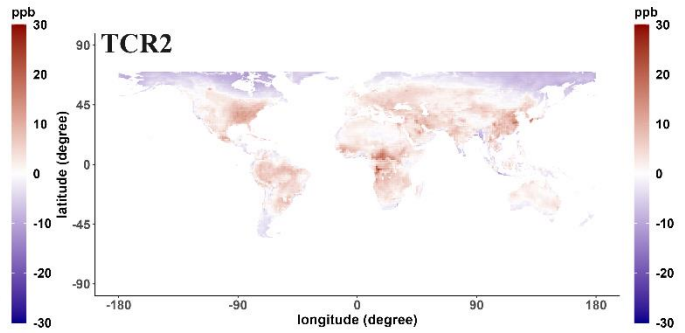
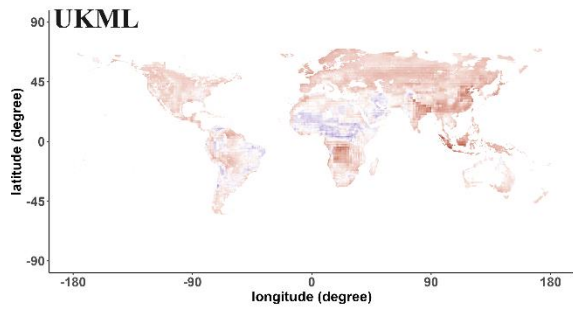
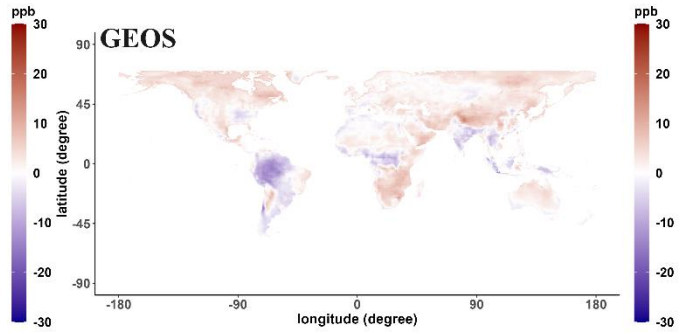
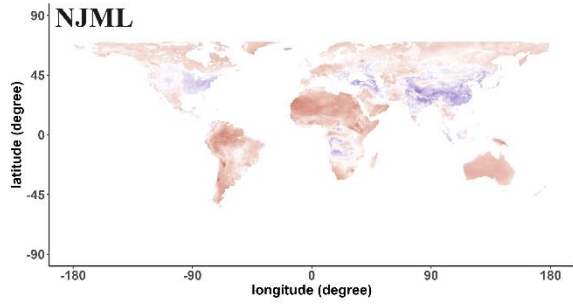
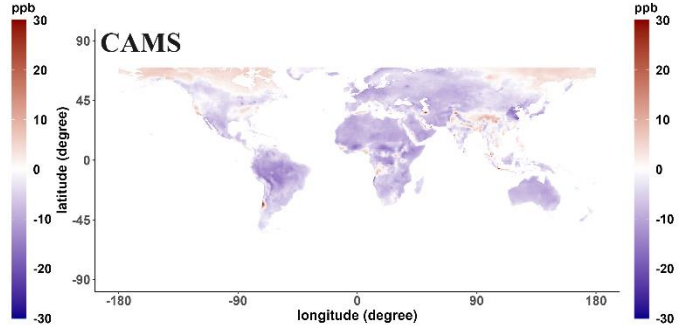
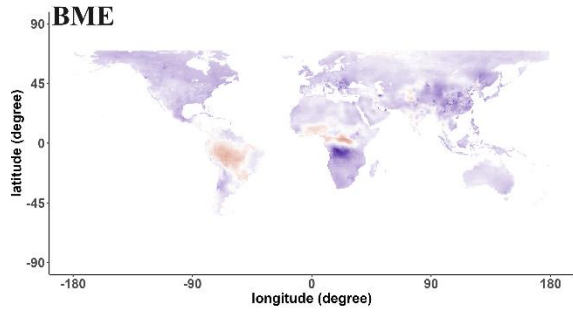
2014



2015



2016



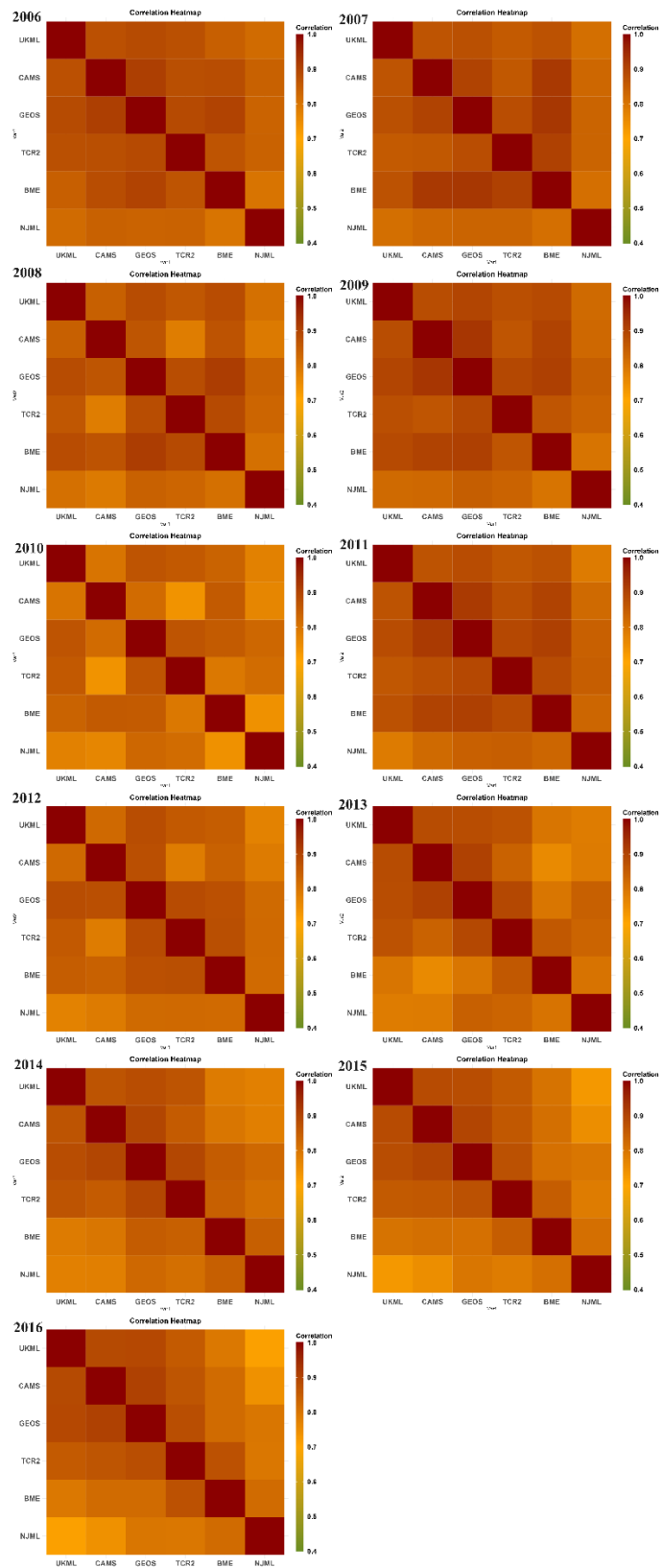


Figure S8. Heatmaps of pairwise correlation (Pearson R) between each dataset from 2006 to 2016.



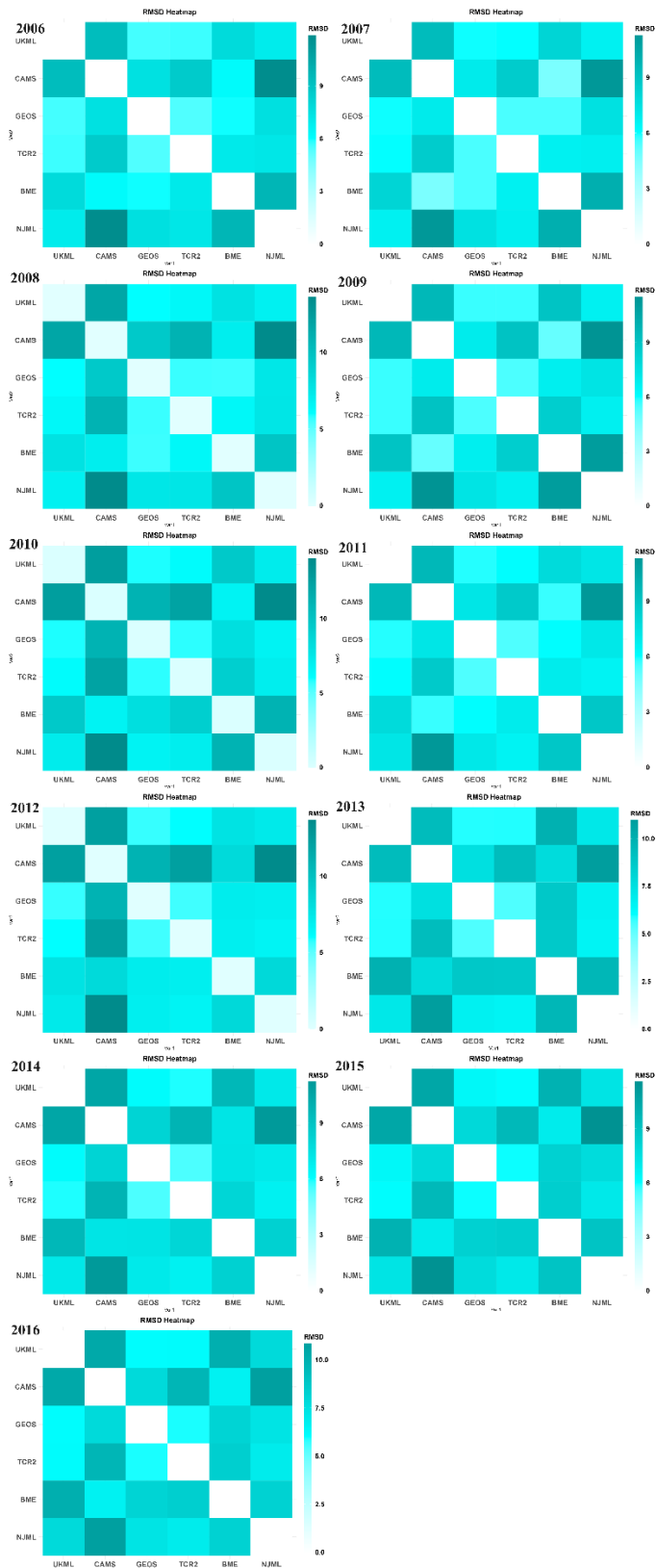
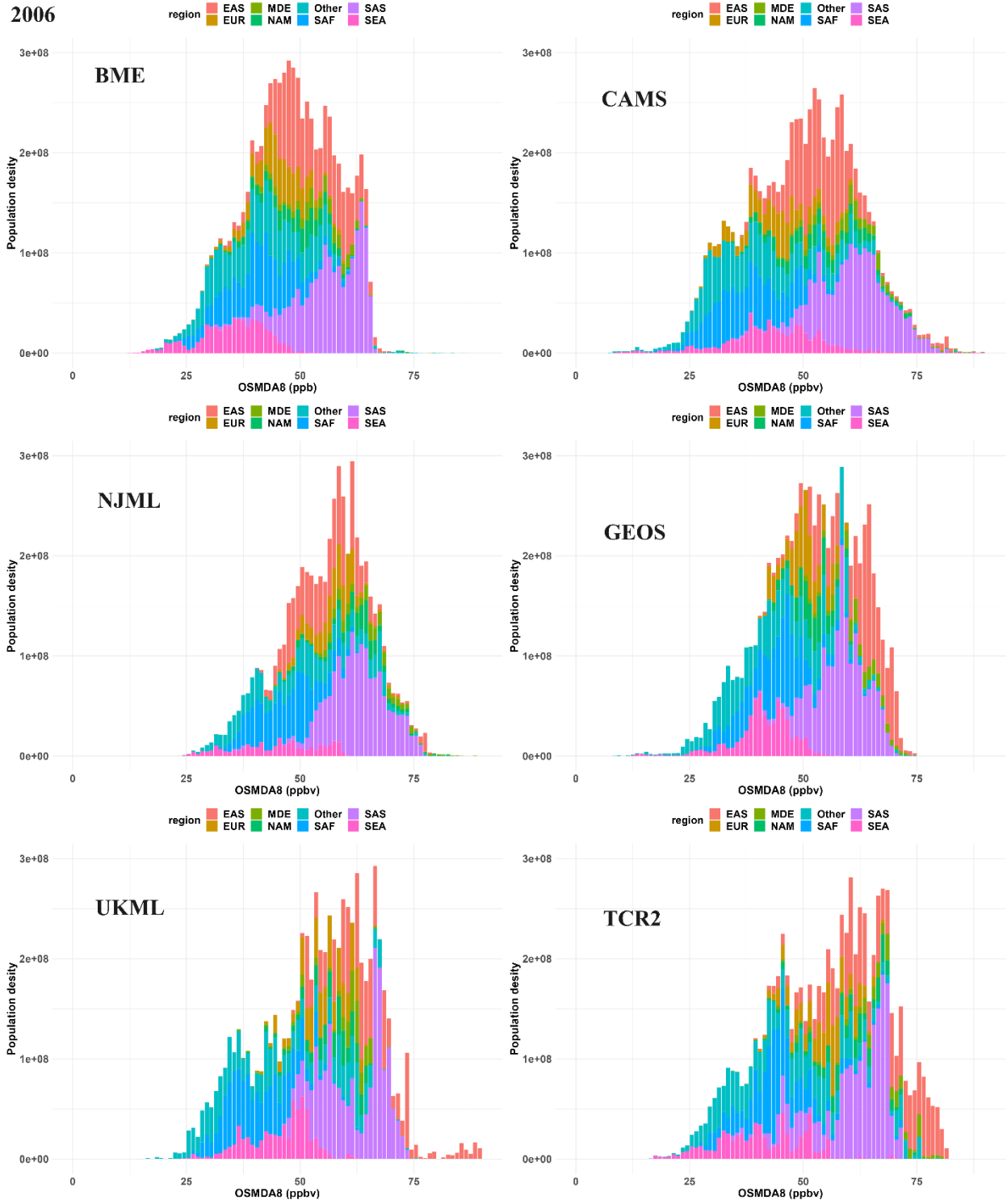
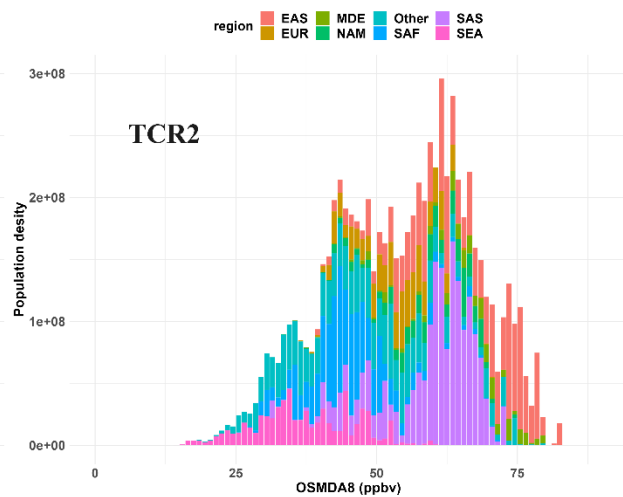
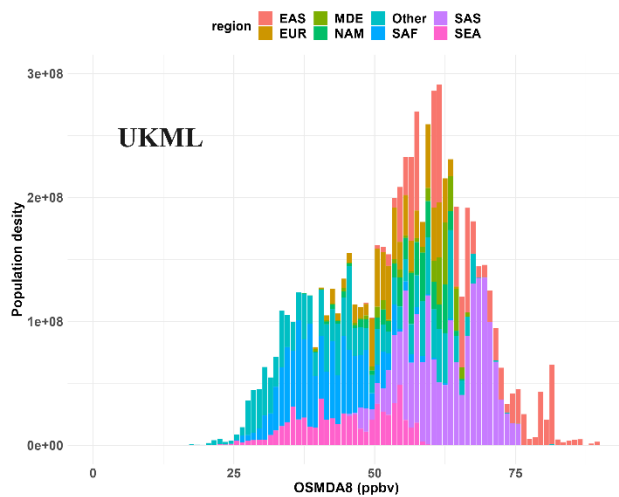
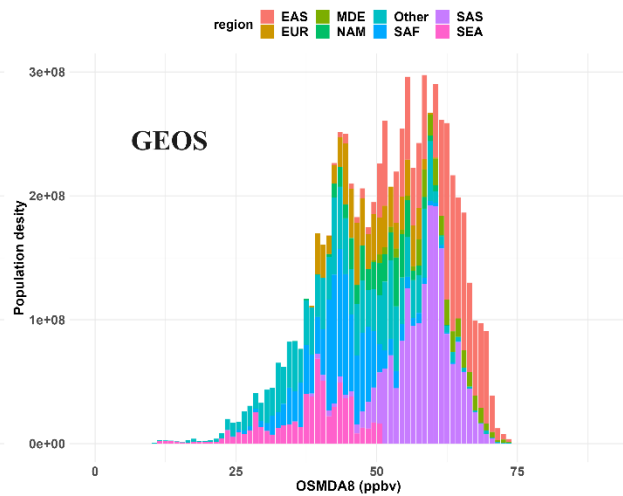
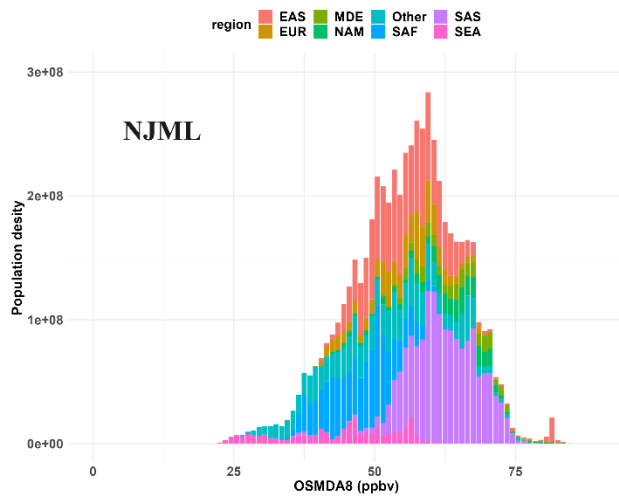
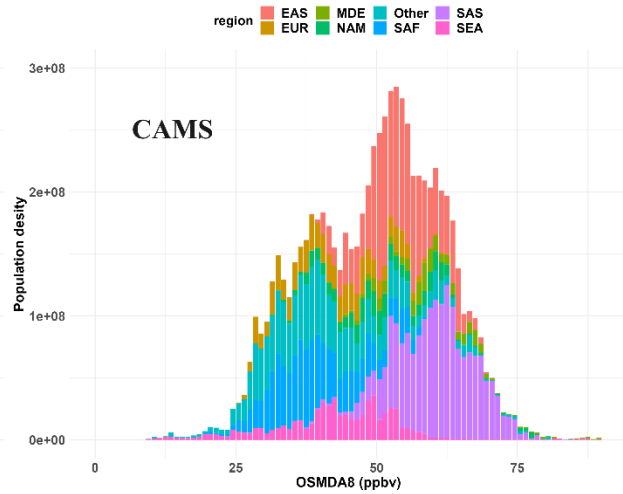
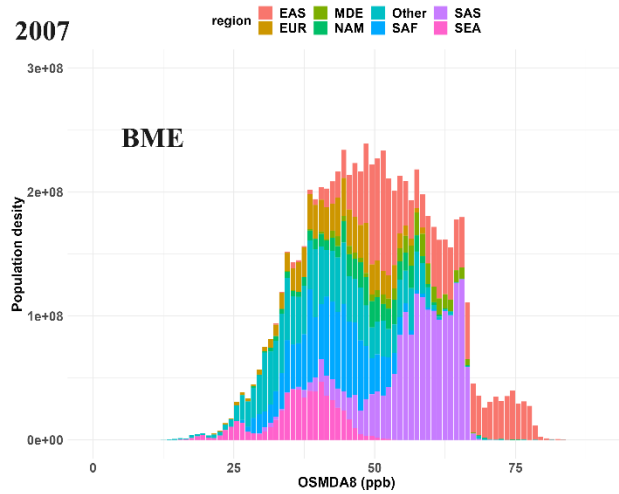


Figure S9. Heatmaps of pairwise Root mean square difference (RMSD) between each dataset from 2006 to 2016.

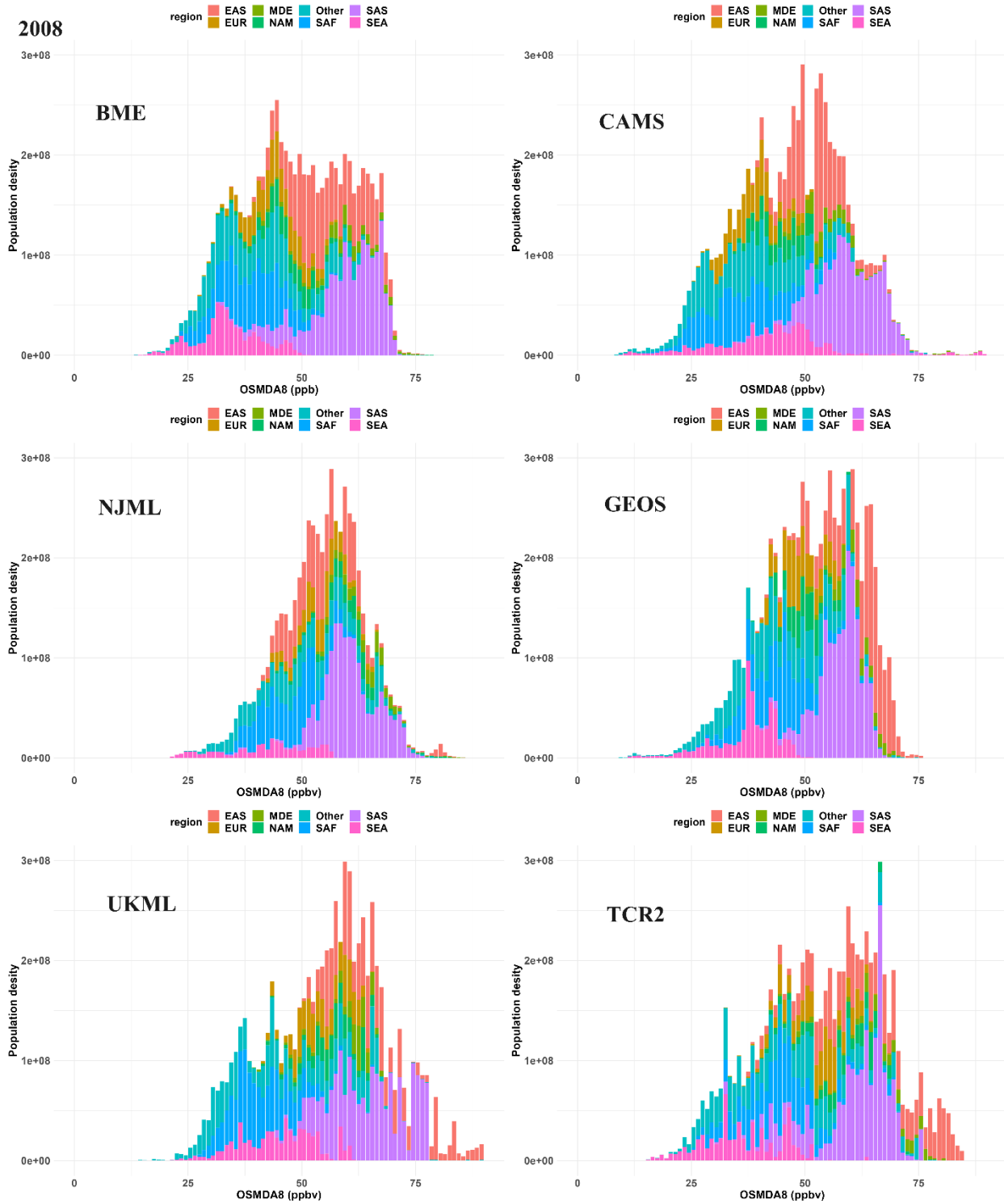
**Figure S10. Population-weighted ozone (OSMDA8) for each year from 2006 to 2016 in different regions. The horizontal axis represents ozone exposure concentrations, and the vertical axis represents population size.**



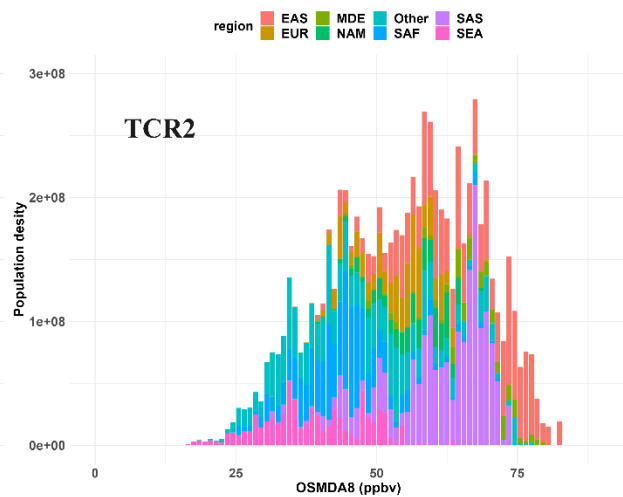
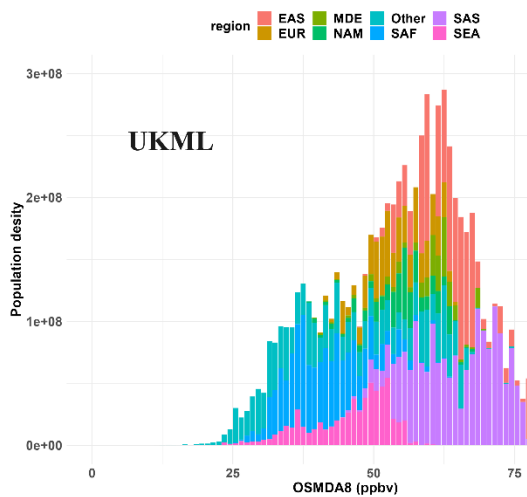
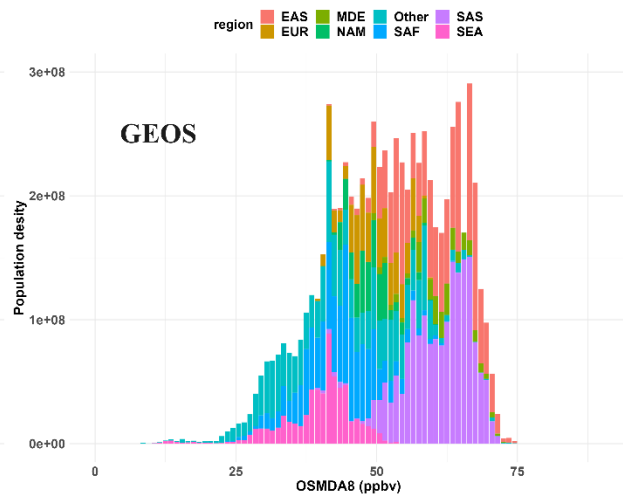
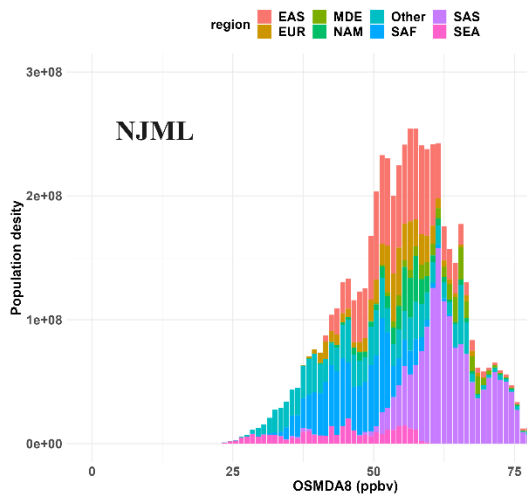
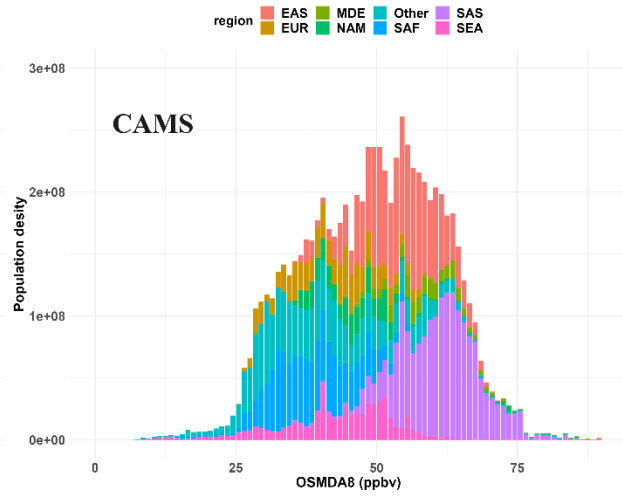
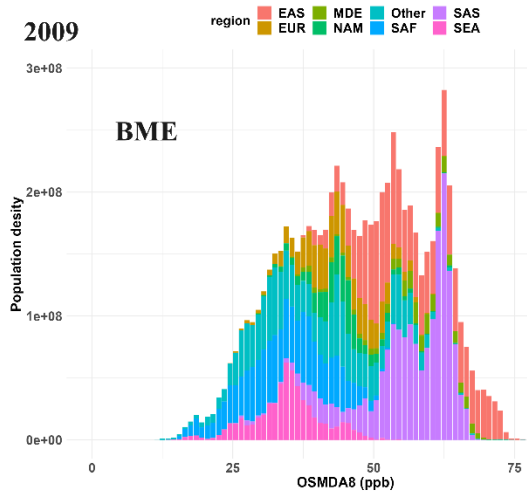
2007



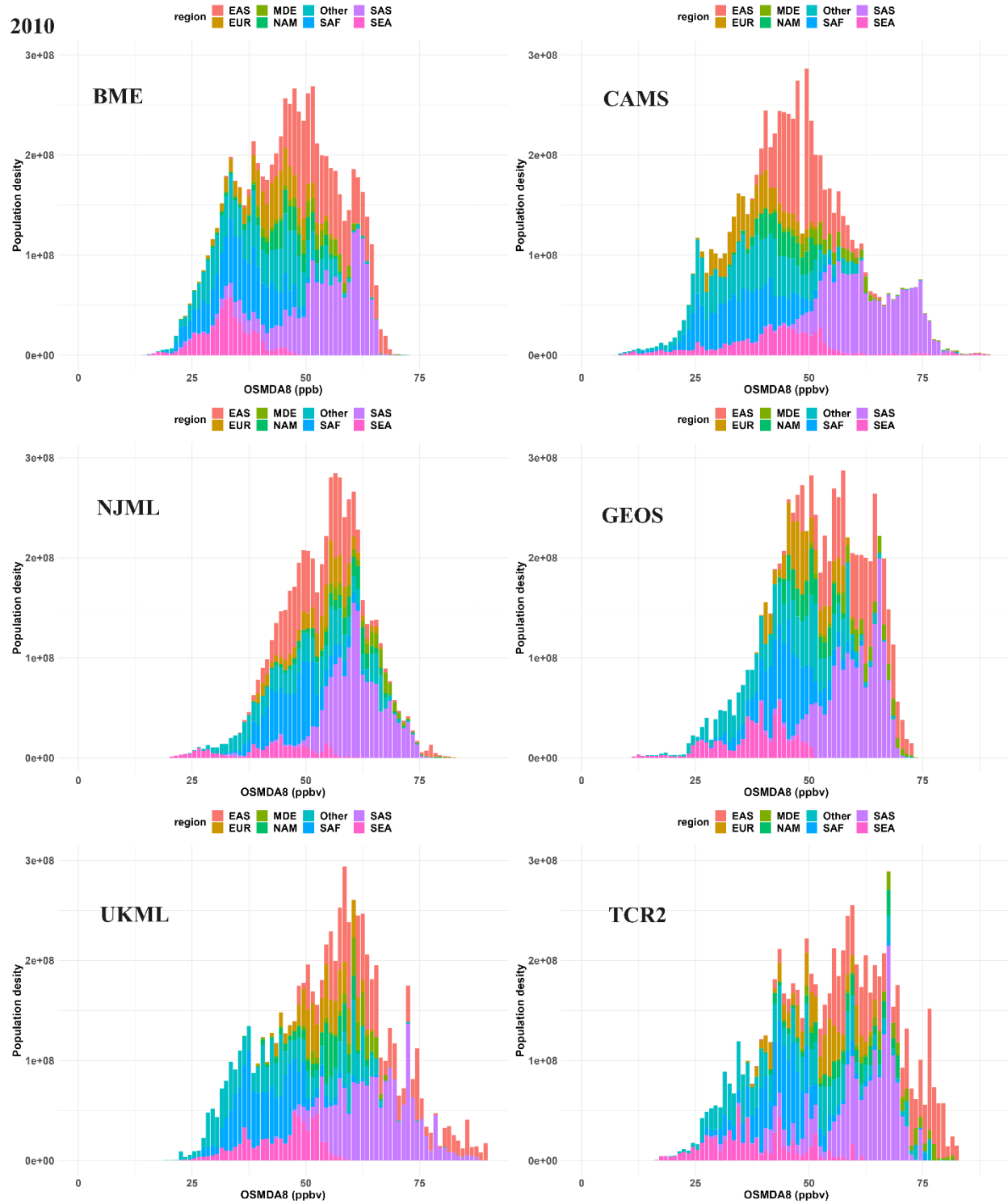
2008

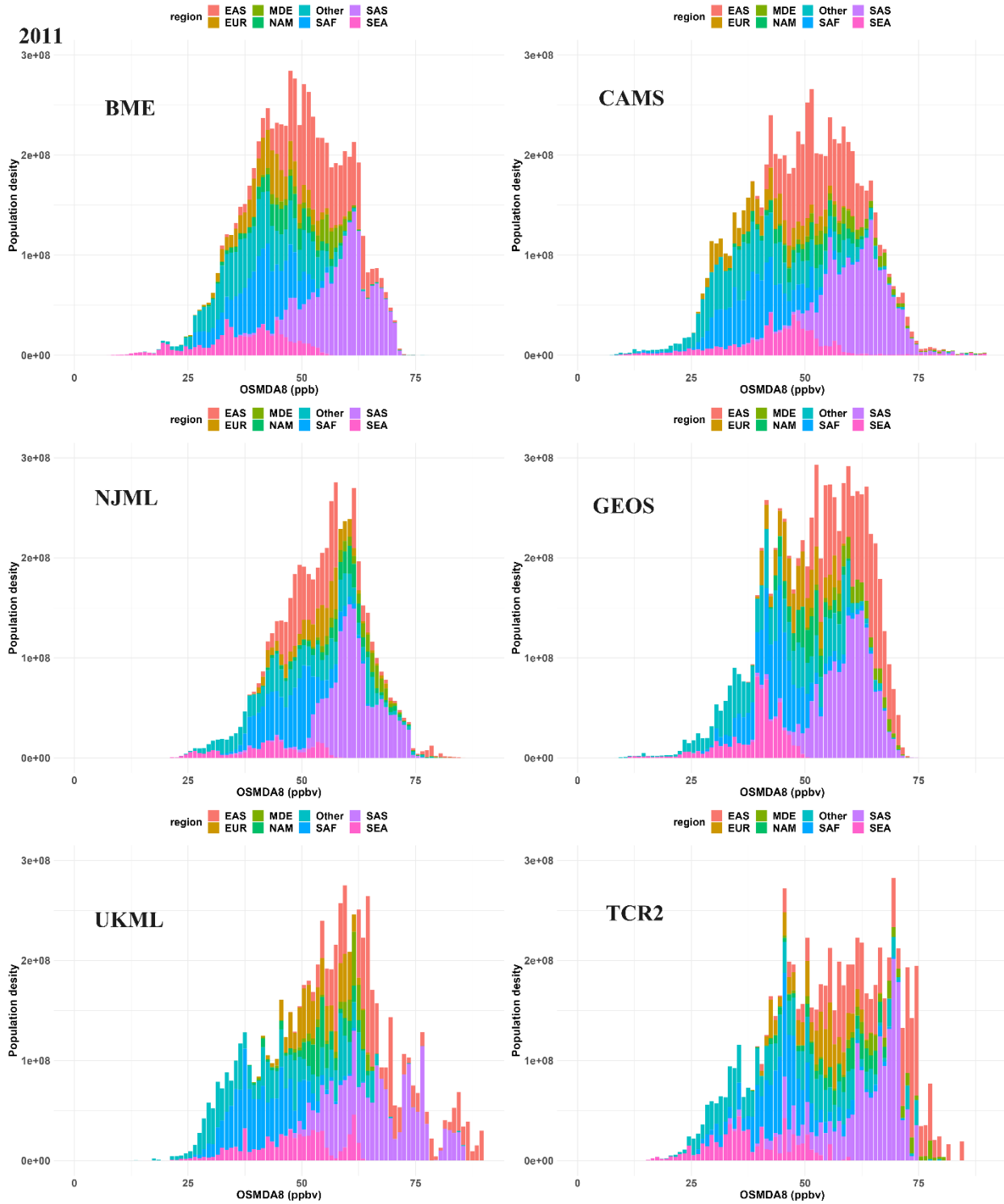


2009

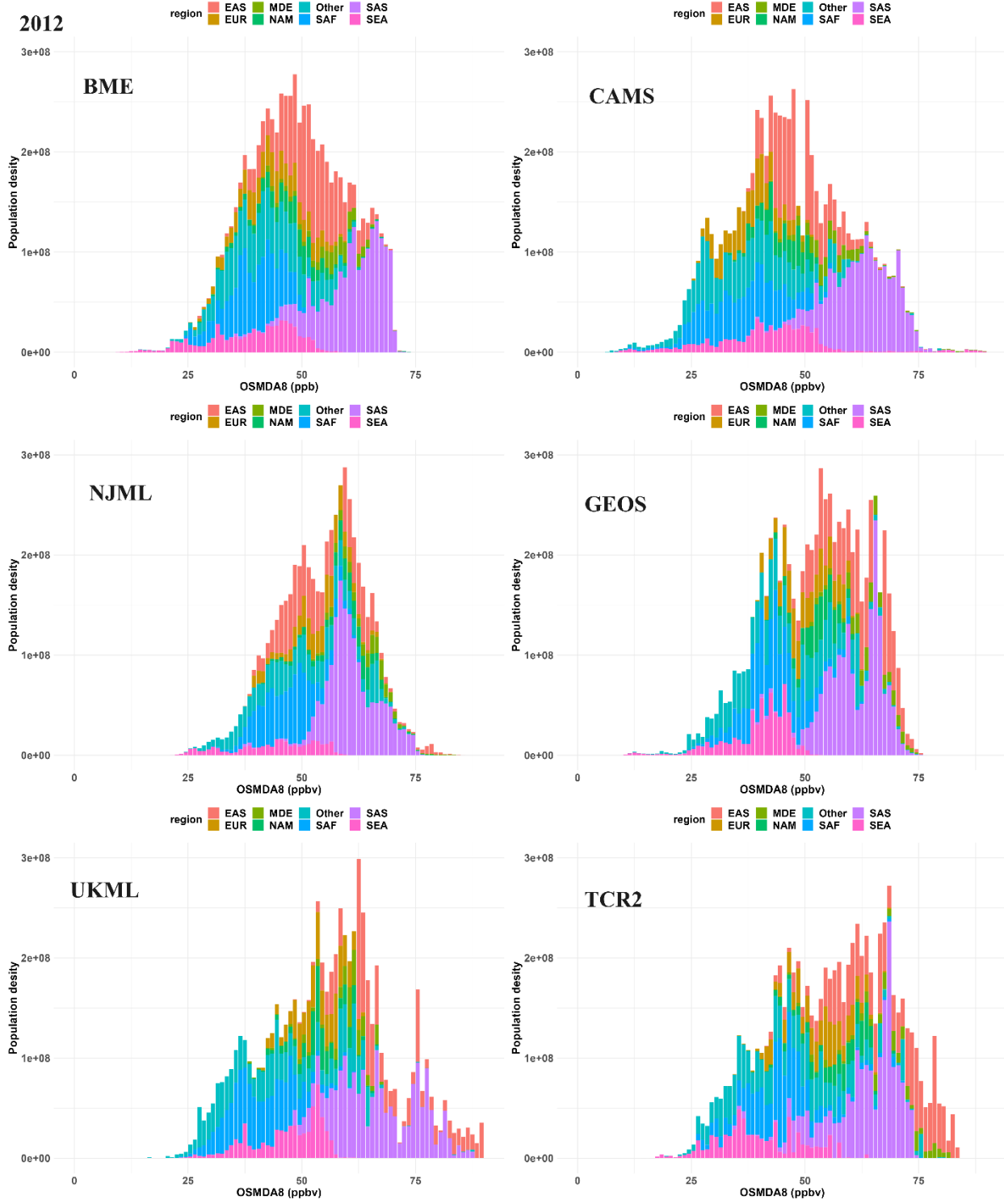


2010



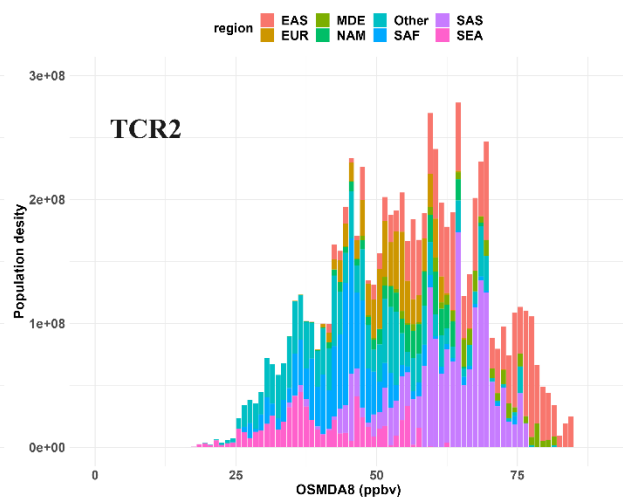
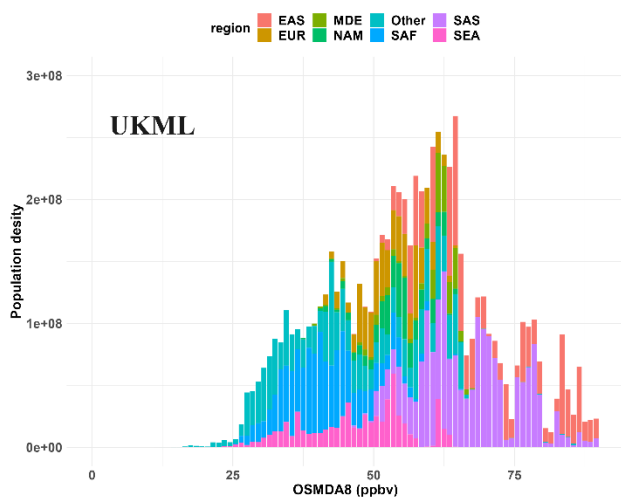
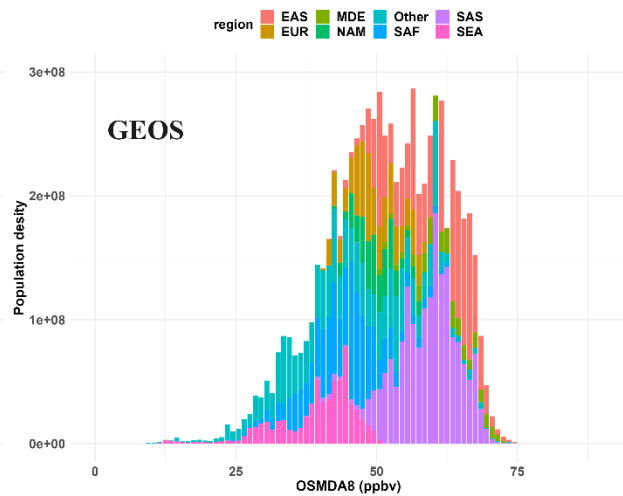
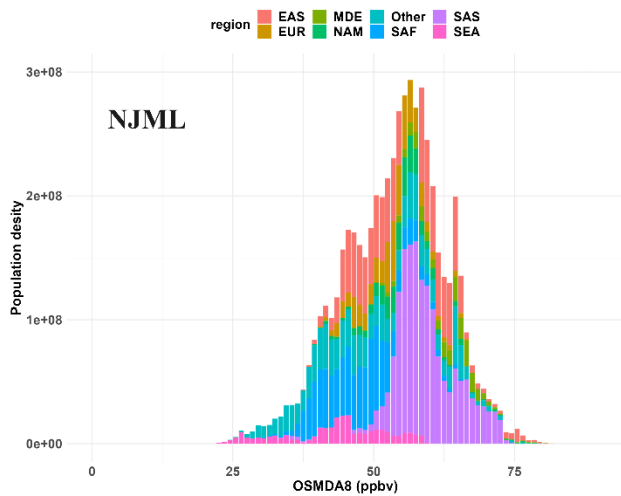
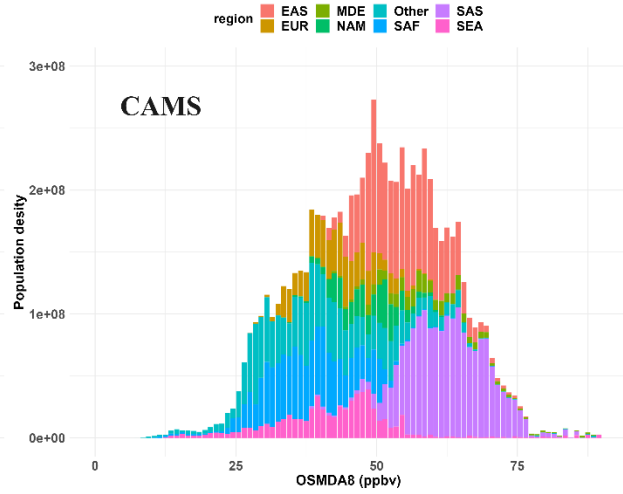
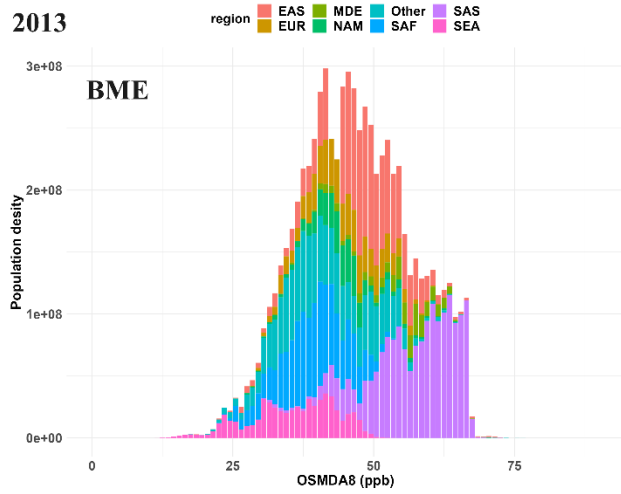


2012

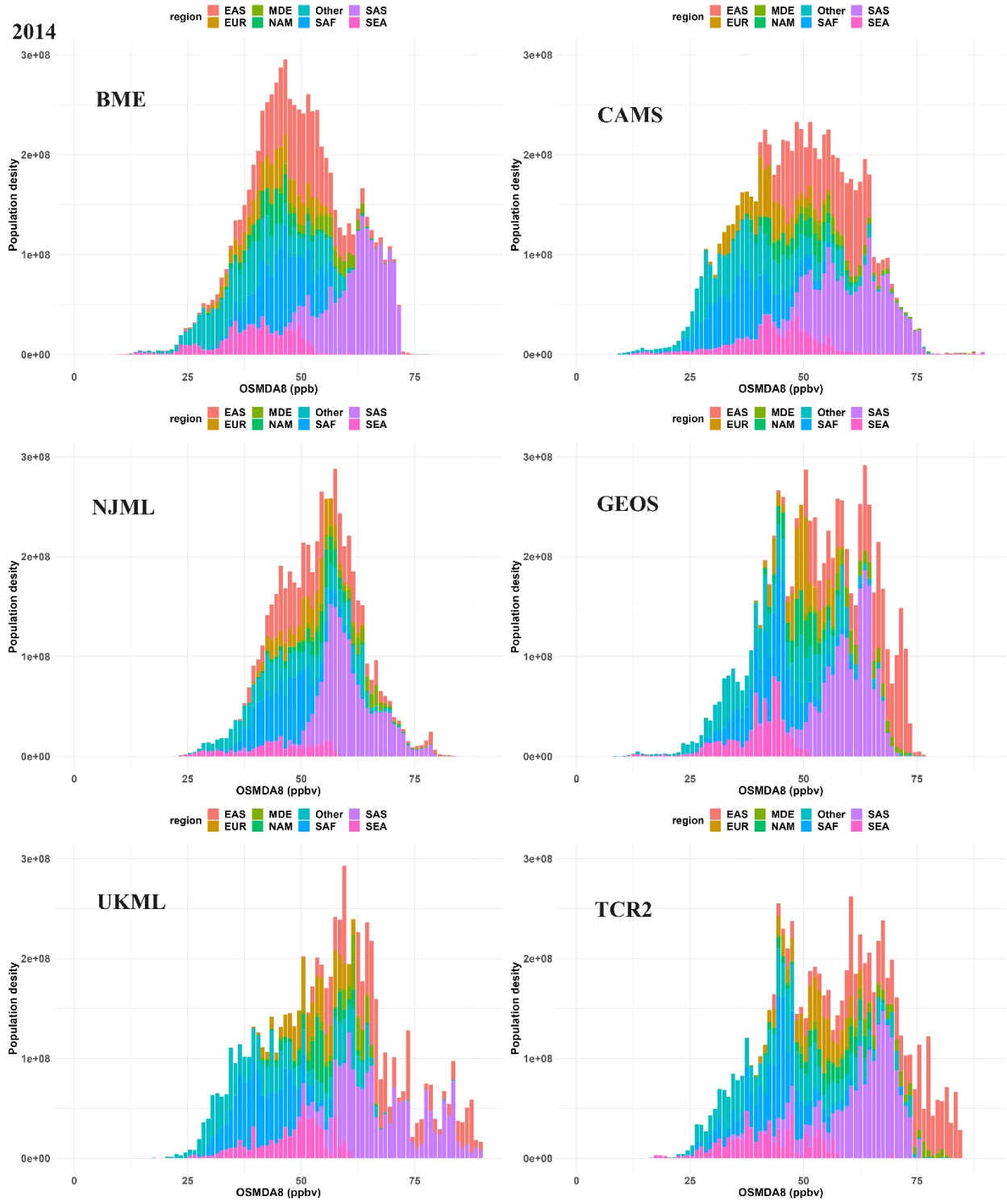


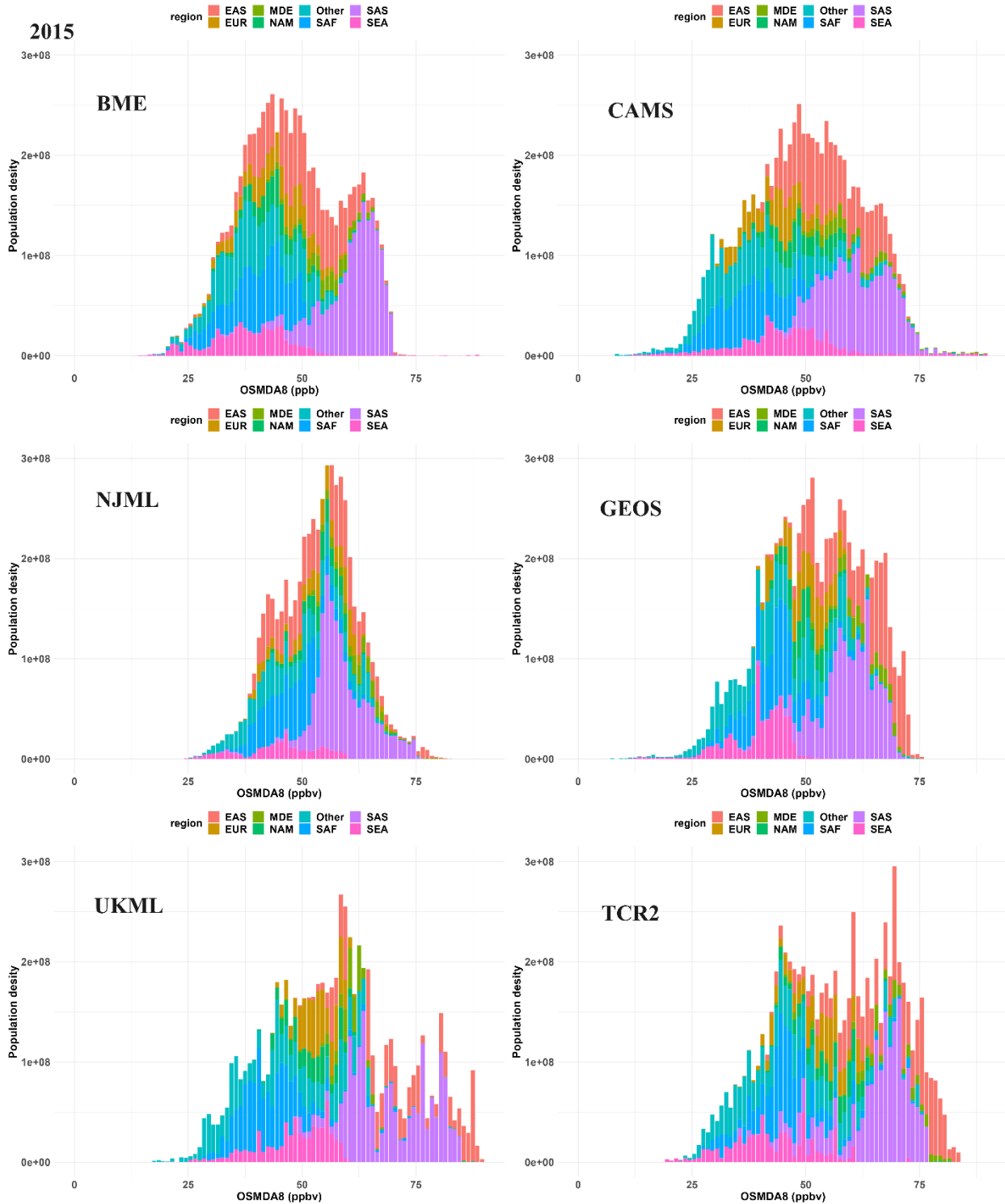


2013



2014





2016

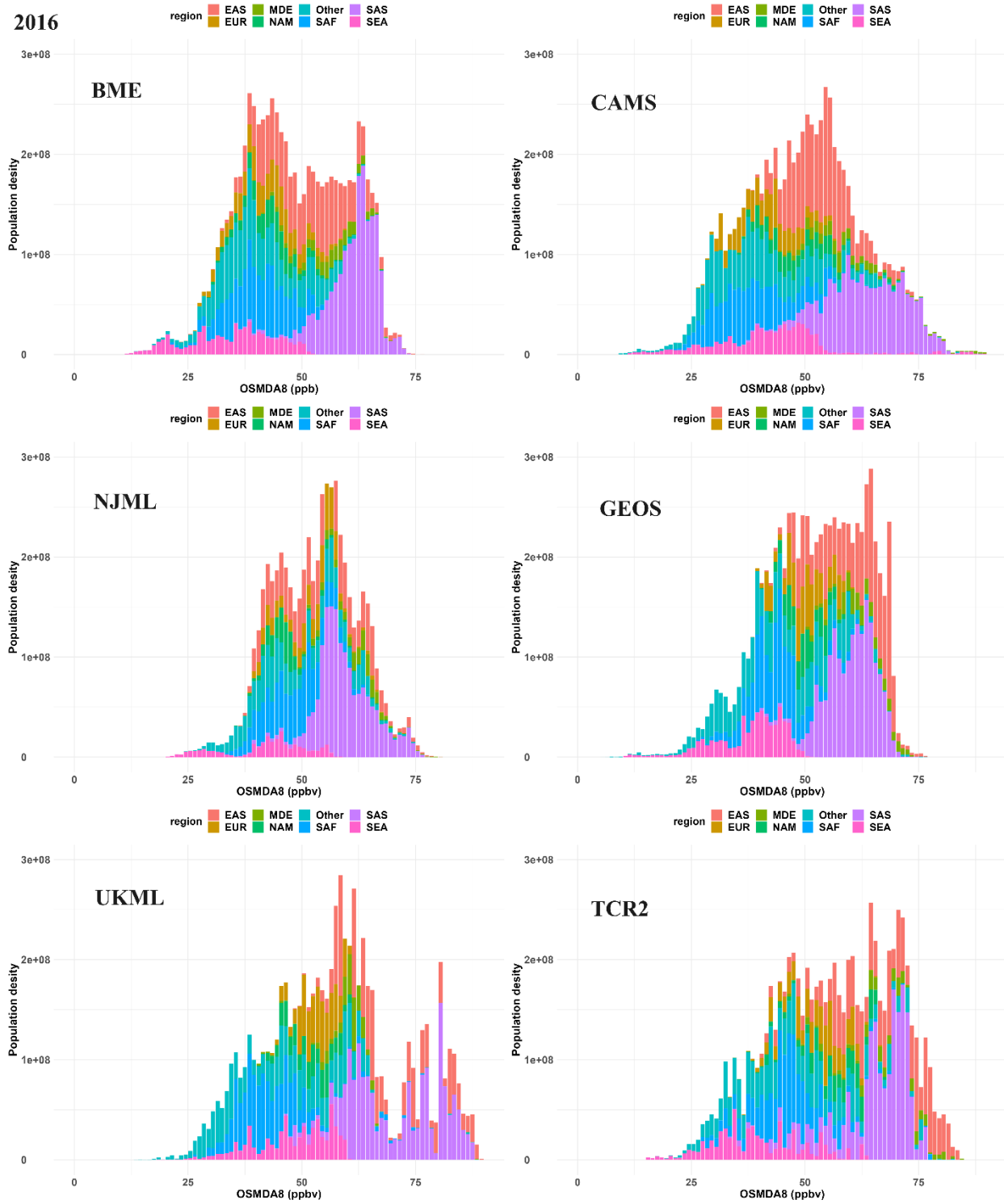
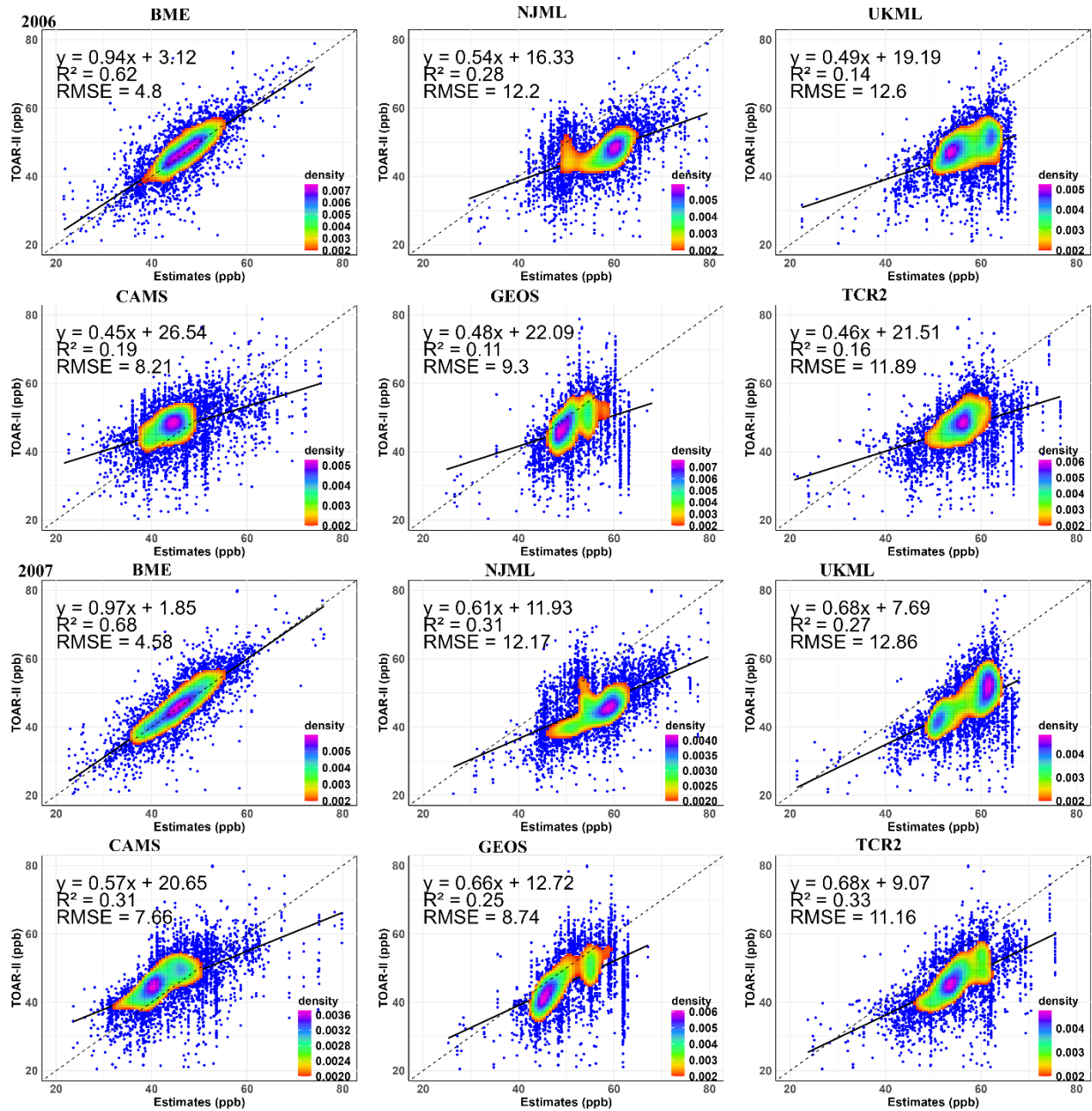
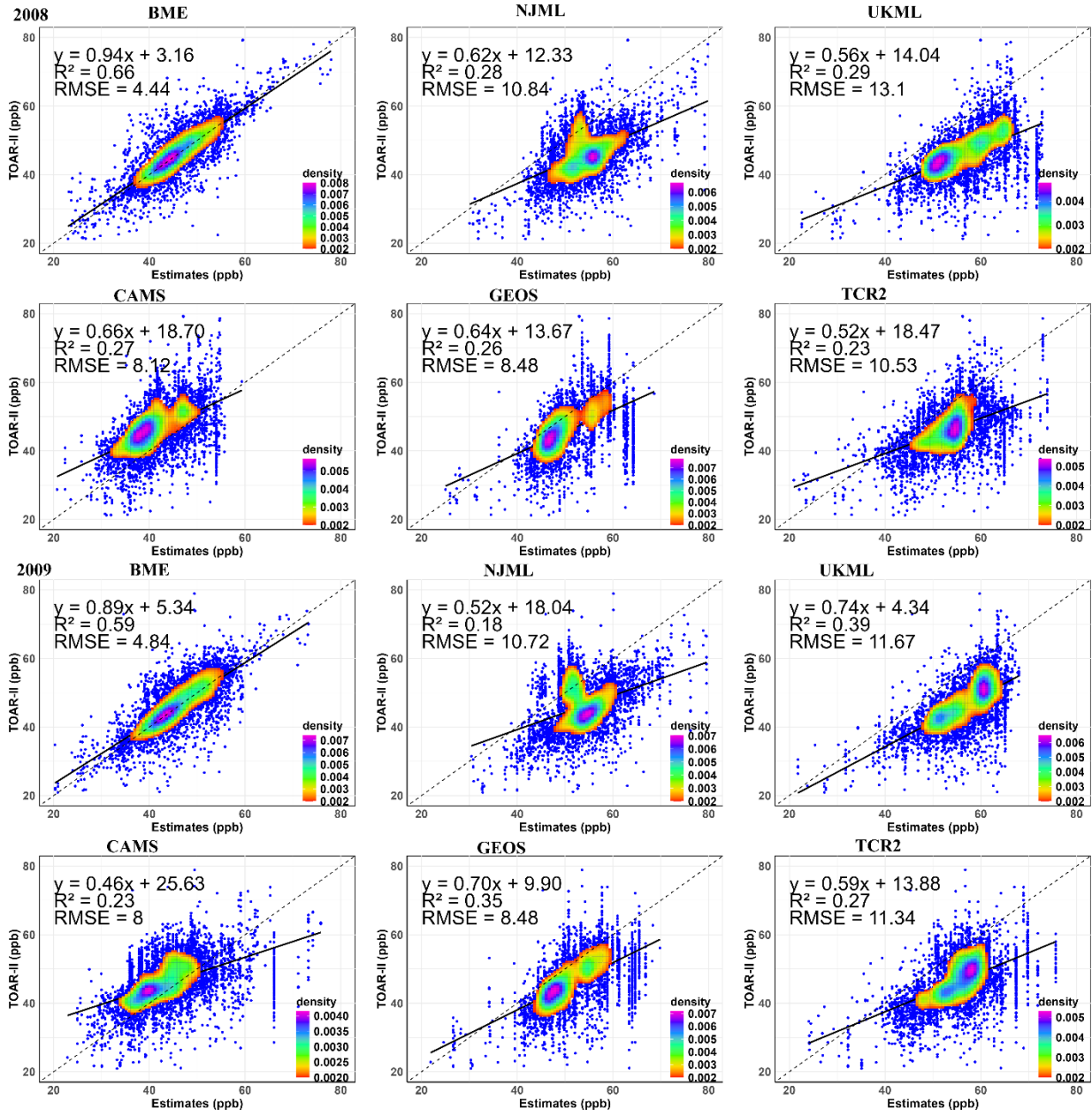
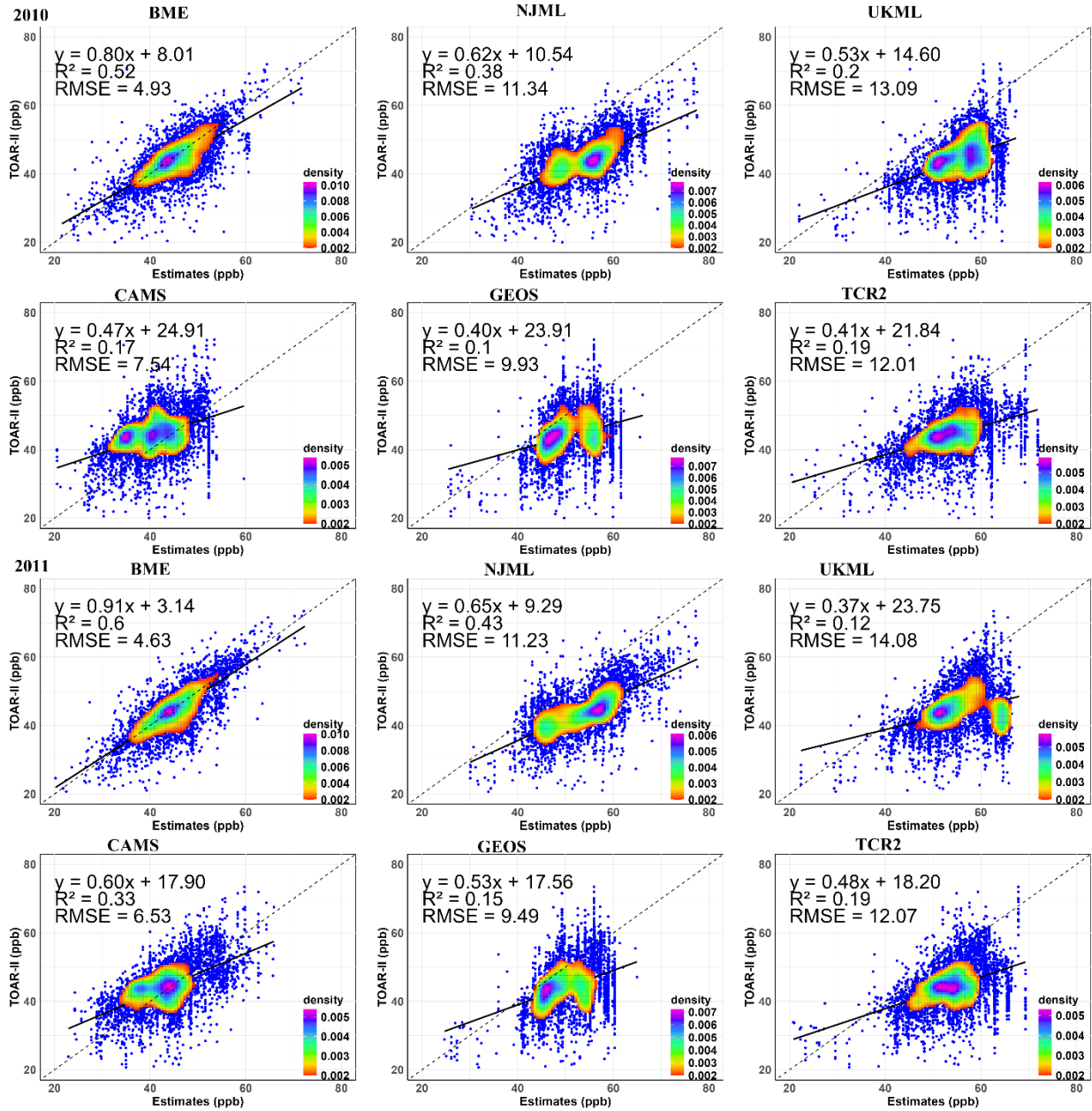
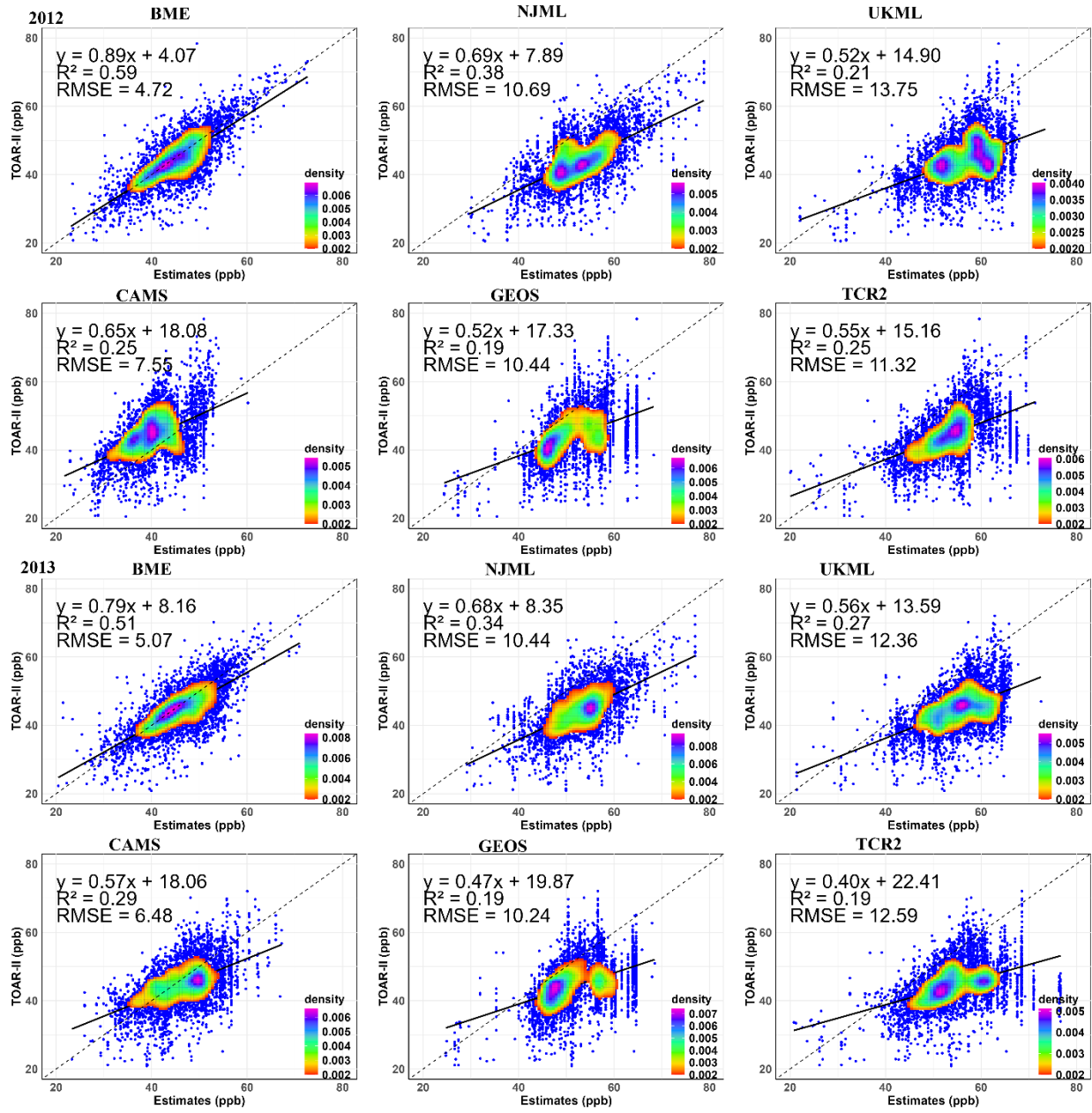


Figure S11. Performance evaluations of six datasets with TOAR-II observations for OSDMA8 for each year from 2006 to 2015. The evaluation includes all monitor stations in the TOAR-II network for each year (2006 to 2015).











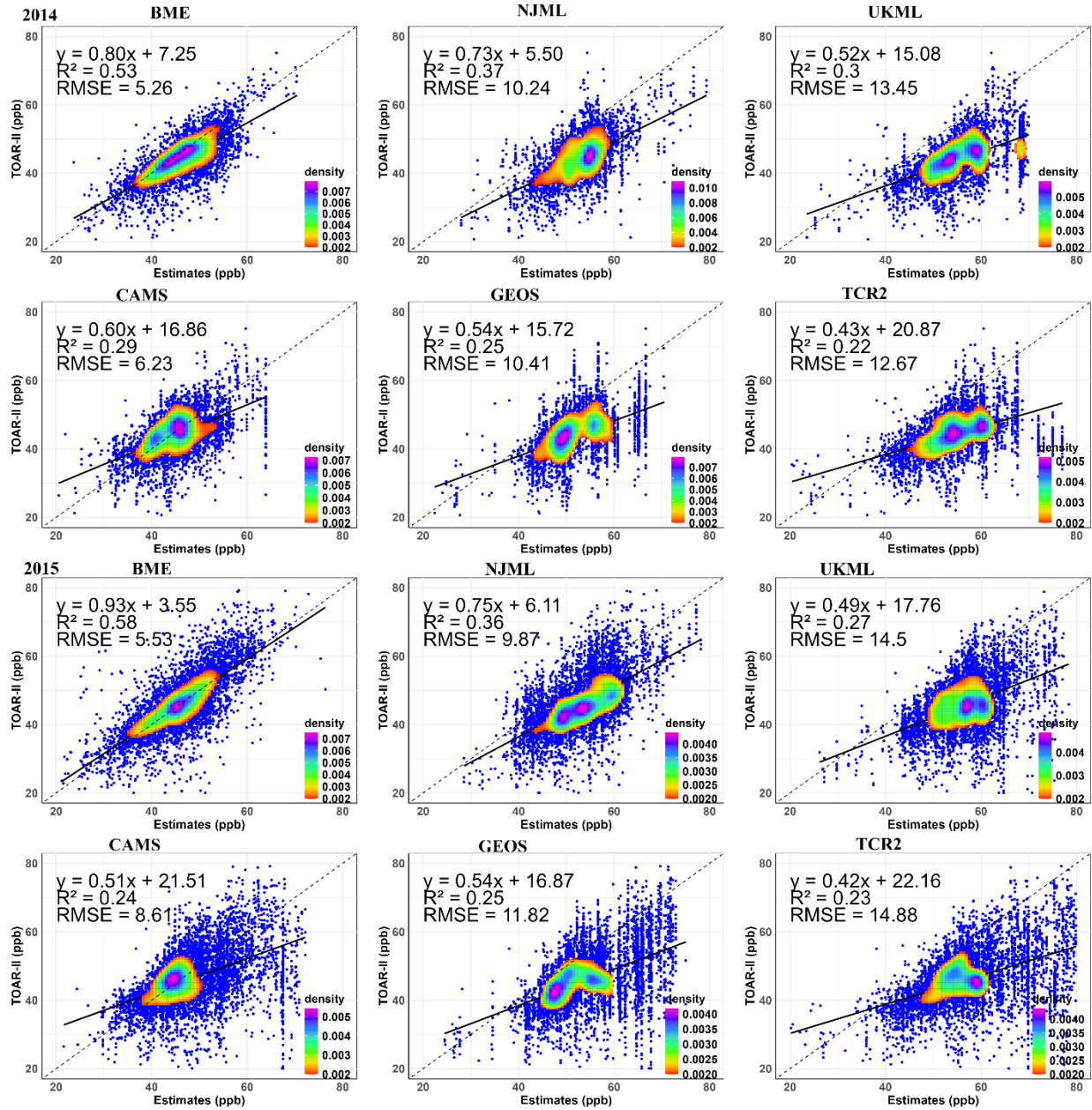
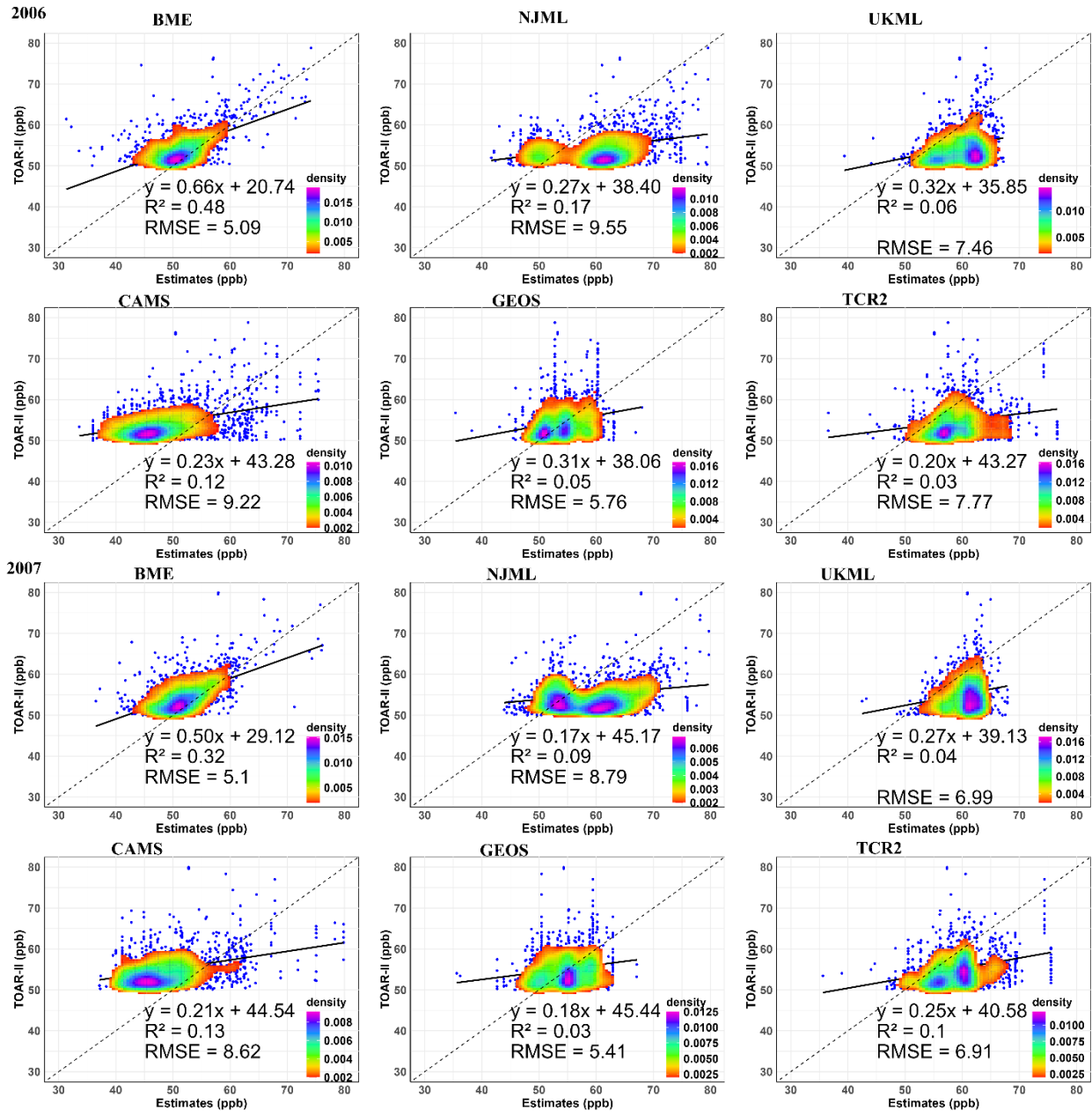
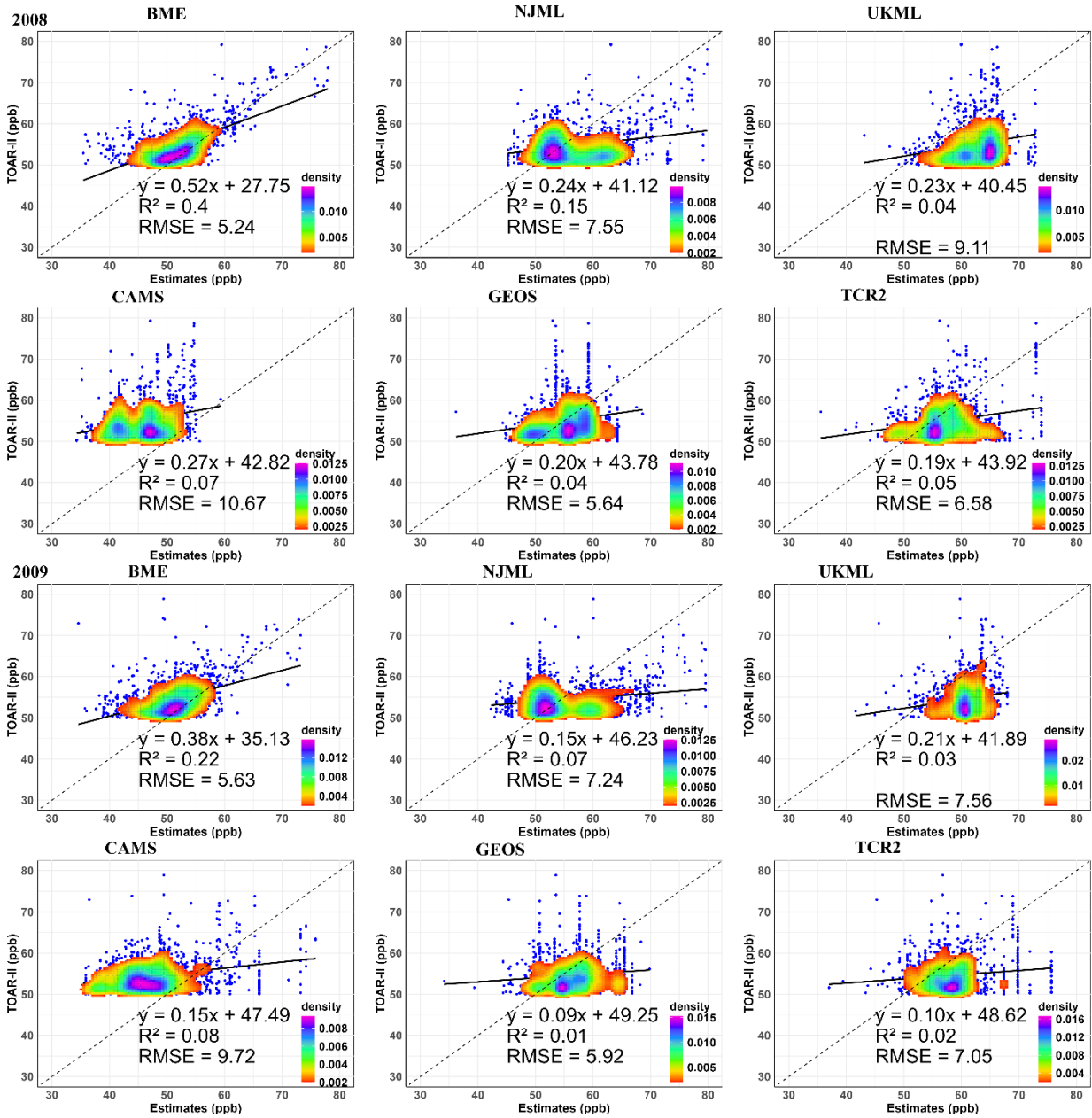
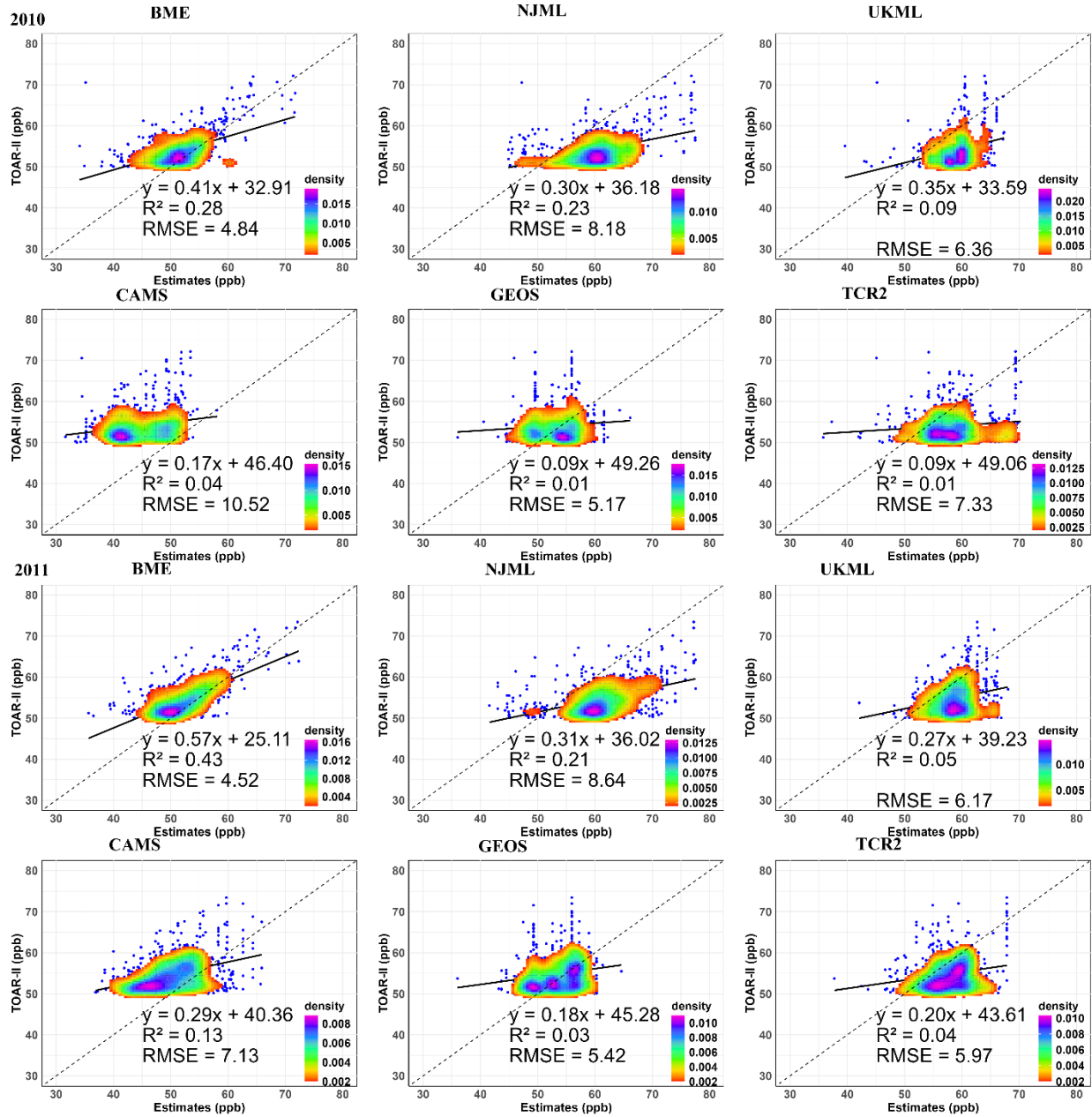
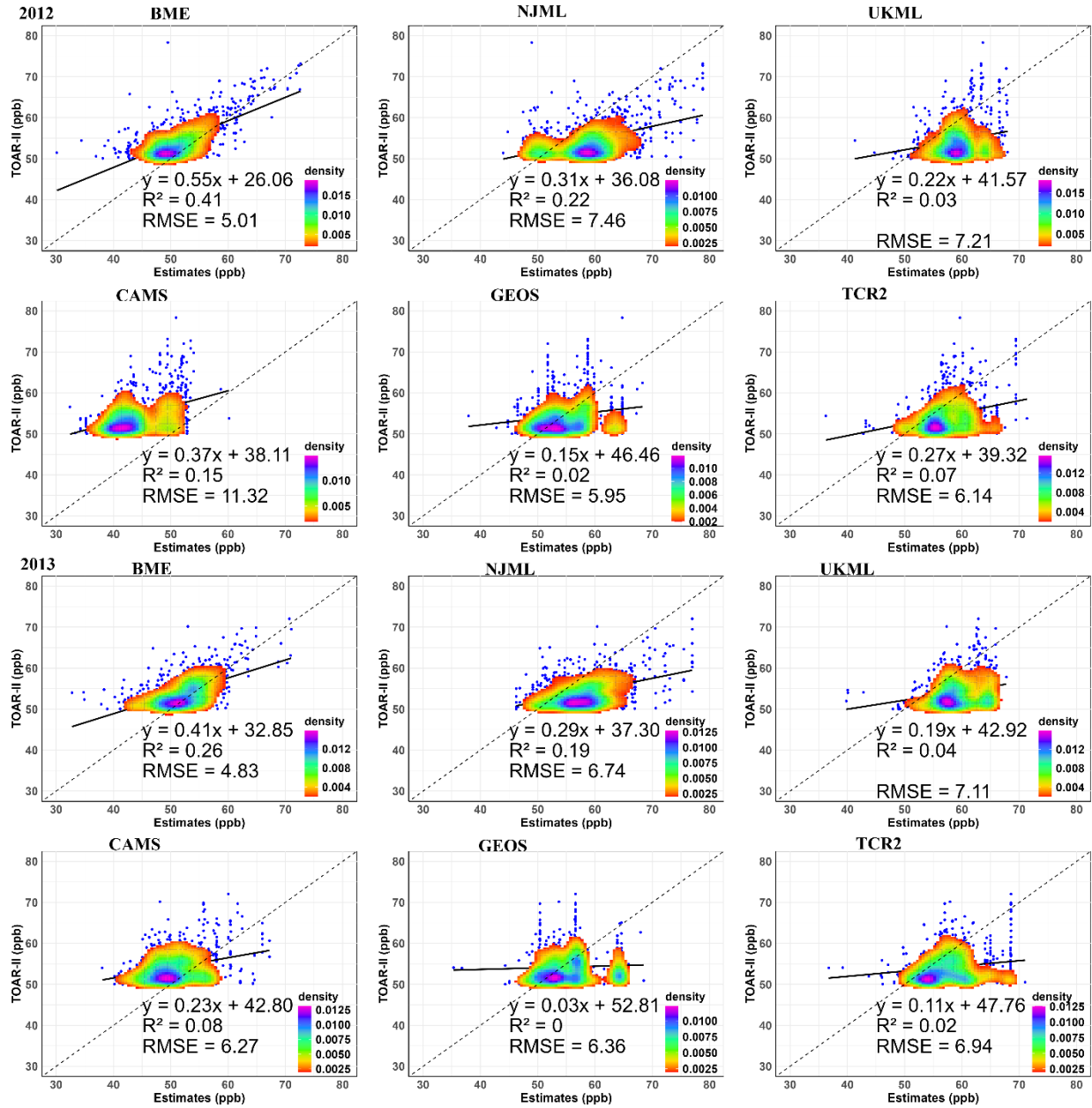


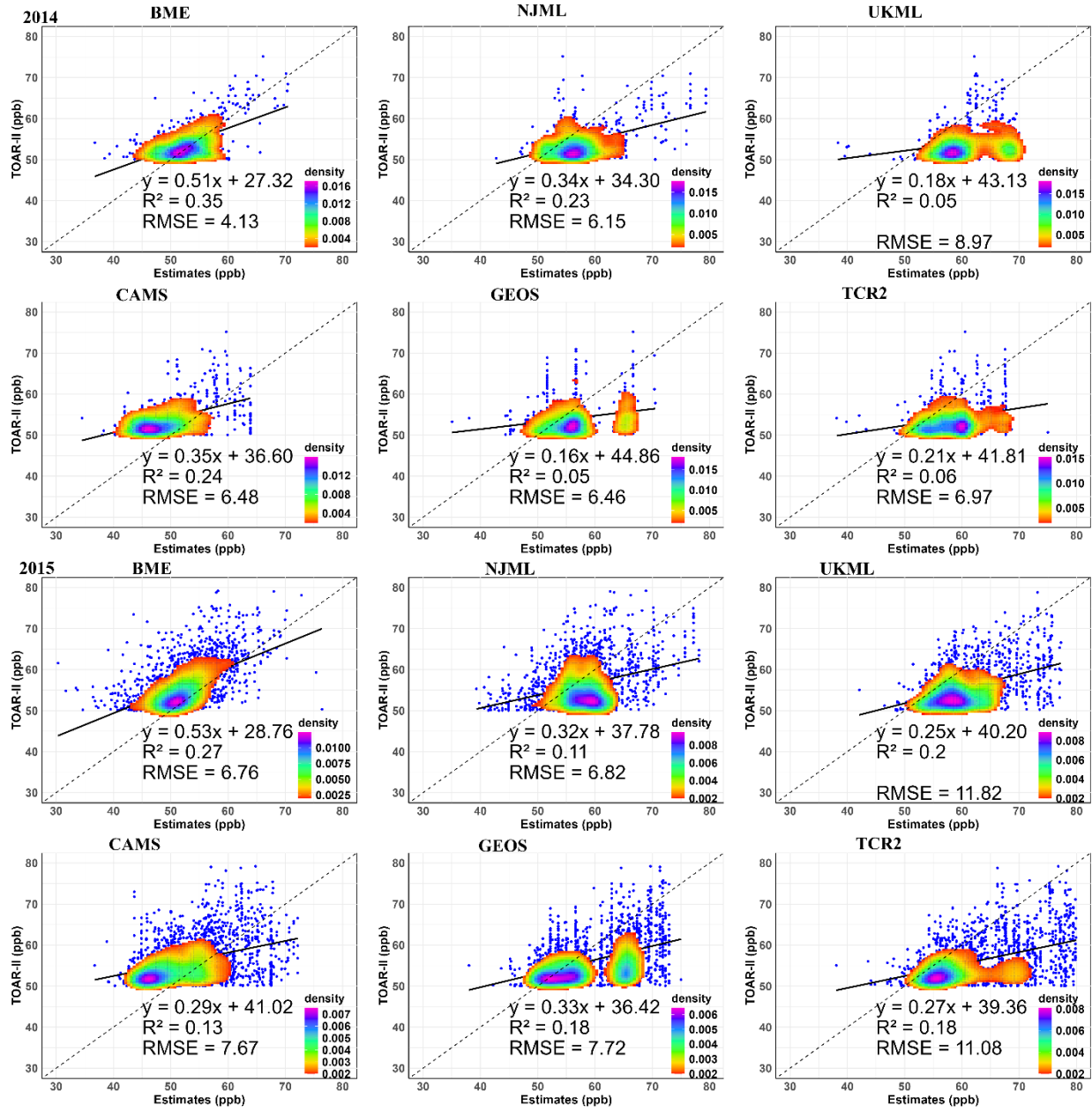
Figure S12. Performance evaluations of six datasets with TOAR-II observations for OSDMA8 for each year from 2006 to 2015. The evaluation only includes monitor stations above 50ppb in TOAR-II network for each year (2006 to 2015).

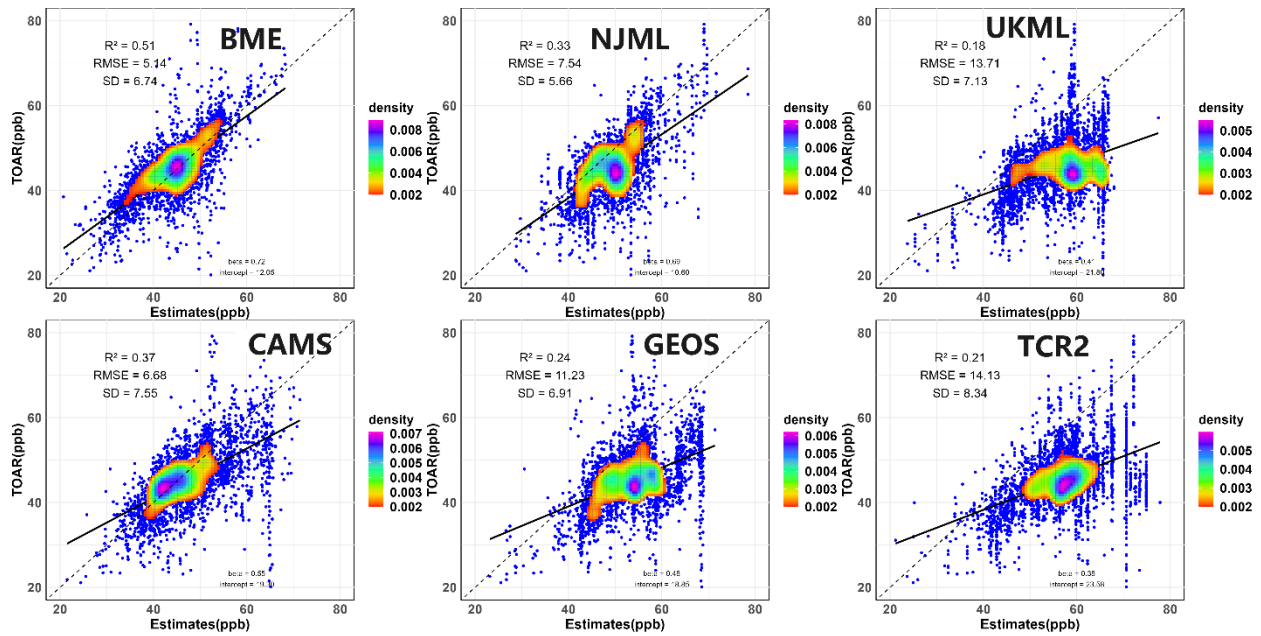












**Figure S13.** Performance evaluations of six datasets with TOAR-II observations in 2016 for OSDMA8. The evaluation excludes the monitoring stations used as input in BME data fusion in 2016. Location of monitoring stations used as input in BME data fusion in 2016 can be found in Figure S1.

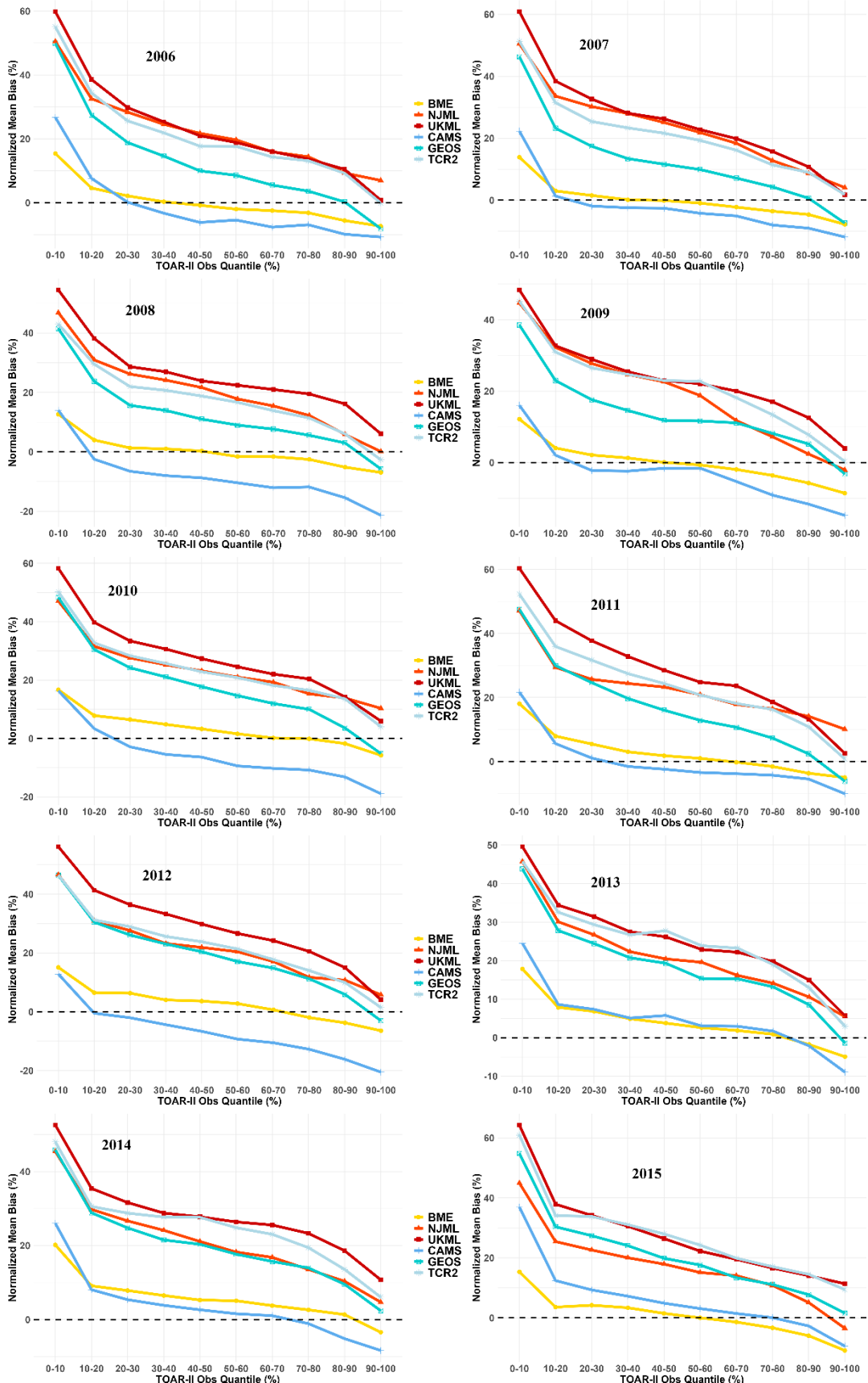




Figure S14. Normalized mean bias of six databases against TOAR-II observations (OSDMA8) at different quantiles for each year from 2006 to 2015. Different quantiles of TOAR-II observations for each year are shown in Table S9.

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