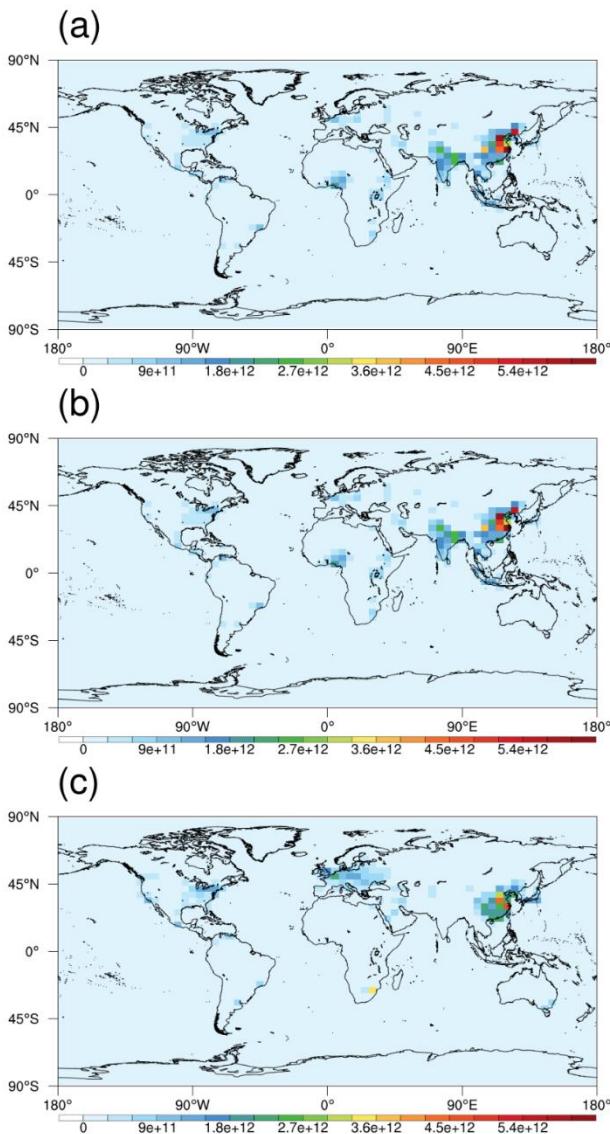


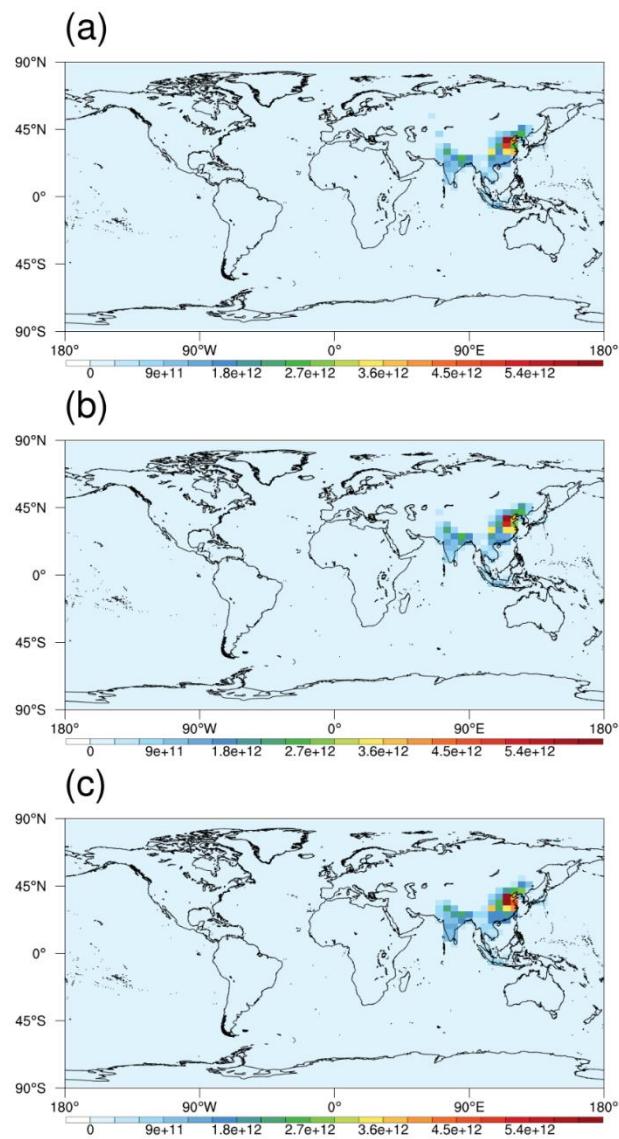
## Supplemental Information

| Inventory   | Resolution | Frequency | Period    | Sector                                                                                                                                                                                  |                                                                                                                                                                                                                                | Data source                                                                                                                                                                                     | Scaling factor            | The time range of the factor |
|-------------|------------|-----------|-----------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------|------------------------------|
| CEDS        | 0.5x0.5    | monthly   | 1750-2019 | 1. Agriculture<br>2. Energy<br>3. Industry<br>4. On-Road Transportation<br>5. Non-Road/Off-Road Transportation<br>6. Residential Combustion                                             | 7. Commercial Combustion<br>8. Other Combustion<br>9. Solvent production and application<br>10. Waste<br>11. International Shipping                                                                                            | <a href="#">McDuffie,et al.(2020)</a>                                                                                                                                                           | Diurnal scale factors     | 24 hours                     |
| MIX         | 0.25x0.25  | monthly   | 2008-2010 | 1.Power plants<br>2.Industry                                                                                                                                                            | 3.Residential<br>4.Transportation                                                                                                                                                                                              | <a href="#">Li, M.,et al(2017)</a>                                                                                                                                                              | AnnualScalar.geos.1x1.nc  | 1985-2010                    |
|             |            |           |           |                                                                                                                                                                                         |                                                                                                                                                                                                                                |                                                                                                                                                                                                 | Diurnal scale factors     | 24 hours                     |
| NEI2011     | 0.1x0.1    | monthly   | 2011      | 1. Surface inventory<br>2. other point sources<br>3. non-EGU industrial stacks<br>4. ships                                                                                              | 5. electric generating units (EGUs)<br>6. peaking electric generating units (EGUs)<br>7. oil and gas sector (new to NEI2011)                                                                                                   | <a href="https://www.epa.gov/air-emissions-inventories/2011-national-emissions-inventory-nei-data">https://www.epa.gov/air-emissions-inventories/2011-national-emissions-inventory-nei-data</a> | NEI11_CO_YRSCALE          | 2006-2013                    |
|             |            |           |           |                                                                                                                                                                                         |                                                                                                                                                                                                                                |                                                                                                                                                                                                 | NEI99.dow.geos.1x1.nc     | 1999/1~12                    |
| DICE_AFRICA | 0.1x0.1    | yearly    | 2016      | 1.Household fuelwood use<br>2.Commercial fuelwood use<br>3.Crop residue for energy<br>4.Charcoal use<br>5.Charcoal production<br>6.Kerosene use                                         | 7.Cars (gasoline and diesel use)<br>8.Motorcycles (gasoline and diesel use)<br>9.Household fuelwood use<br>10.Household generator use<br>11.Natural gas flaring<br>12.Ad hoc oil refining                                      | <a href="#">Marais,et al.</a>                                                                                                                                                                   | Diurnal scale factors     | 24 hours                     |
| EDGARV43    | 0.1x0.1    | yearly    | 1970-2011 | 1.Power generation<br>2.Energy industry<br>3.Manufacturing industry<br>4.Road transport<br>5.Railways, pipelines, off-road transport<br>6.Energy for buildings (residential combustion) | 7.Process emissions during production and application<br>8.Agriculture (excluding soil and agricultural burning)<br>9.Agricultural waste burning<br>10.Soil emissions<br>11.Waste solid and wastewater<br>12.Fossil Fuel Fires | <a href="#">Crippa, M.,et al</a>                                                                                                                                                                | EDGAR_v43.Seasonal.1x1.nc | 2010/1~12                    |
|             |            |           |           |                                                                                                                                                                                         |                                                                                                                                                                                                                                |                                                                                                                                                                                                 | COPROD_FOSSIL=1.19        | \                            |
| APEI        | 0.1x0.1    | yearly    | 1989-2016 | \                                                                                                                                                                                       |                                                                                                                                                                                                                                | <a href="http://edgar.ire.ec.europa.eu/overview.php?v=431">http://edgar.ire.ec.europa.eu/overview.php?v=431</a>                                                                                 | COPROD_FOSSIL=1.19        | \                            |
| GFED4       | 0.25x0.25  | monthly   | 1997-2019 | 1.Total dry matter<br>2.Dry matter from temperate forests<br>3.Dry matter from peat<br>4.Dry matter from savanna                                                                        | 5.Dry matter from deforestation<br>6.Dry matter from boreal forest<br>7.Dry matter from agricultural waste                                                                                                                     | <a href="#">Randerson,et al.(2015)</a>                                                                                                                                                          | GFED_emission_factors.txt | \                            |
|             |            |           |           |                                                                                                                                                                                         |                                                                                                                                                                                                                                |                                                                                                                                                                                                 | Scaling_CO=1.05           | \                            |

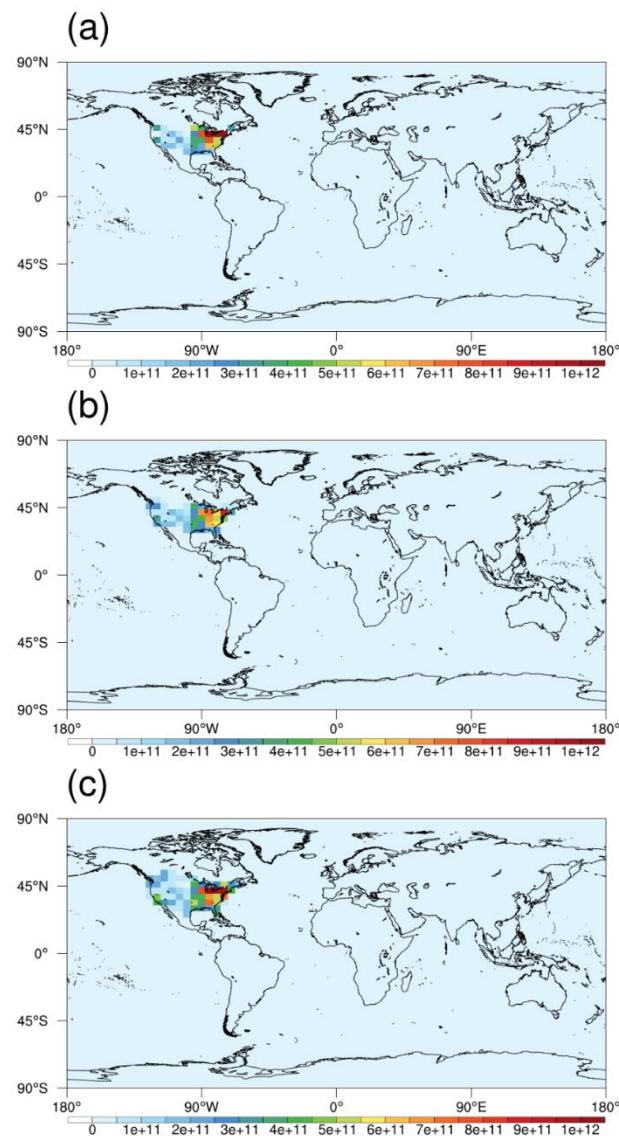
**Table S1.** Updated emission inventories adopted in GC-Adjoint-HEMCO.



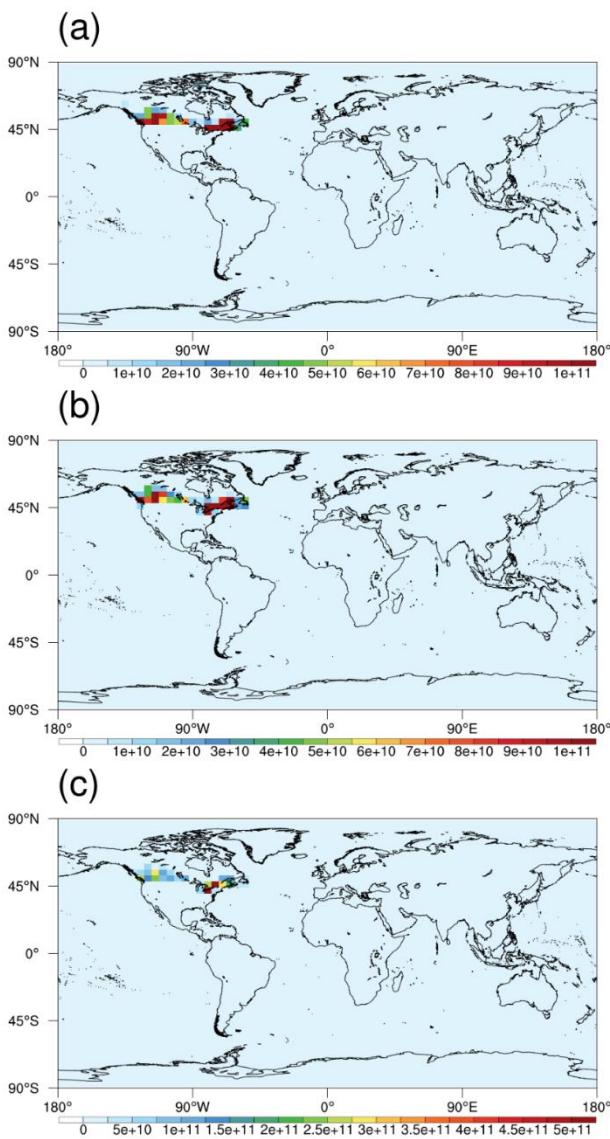
**Fig. S1.** CEDS CO emissions in 2015 from (a) GC-v12; (b) GC-Adjoint-HEMCO; and GEIA CO emissions in (c) GC-Adjoint-STD. The unit is molec/cm/s.



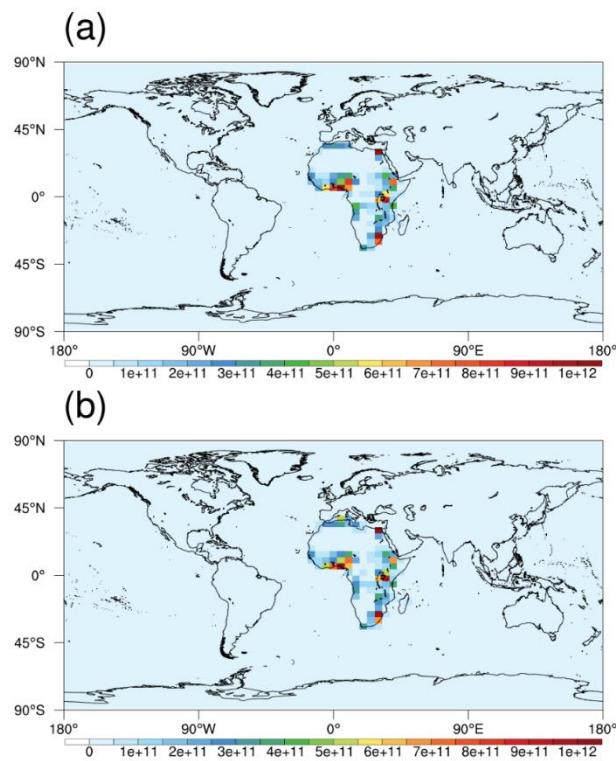
**Fig. S2.** MIX CO emissions in 2015 from (a) GC-v12; (b) GC-Adjoint-HEMCO; and INTEX-B CO emissions in (c) GC-Adjoint-STD. The unit is molec/cm/s.



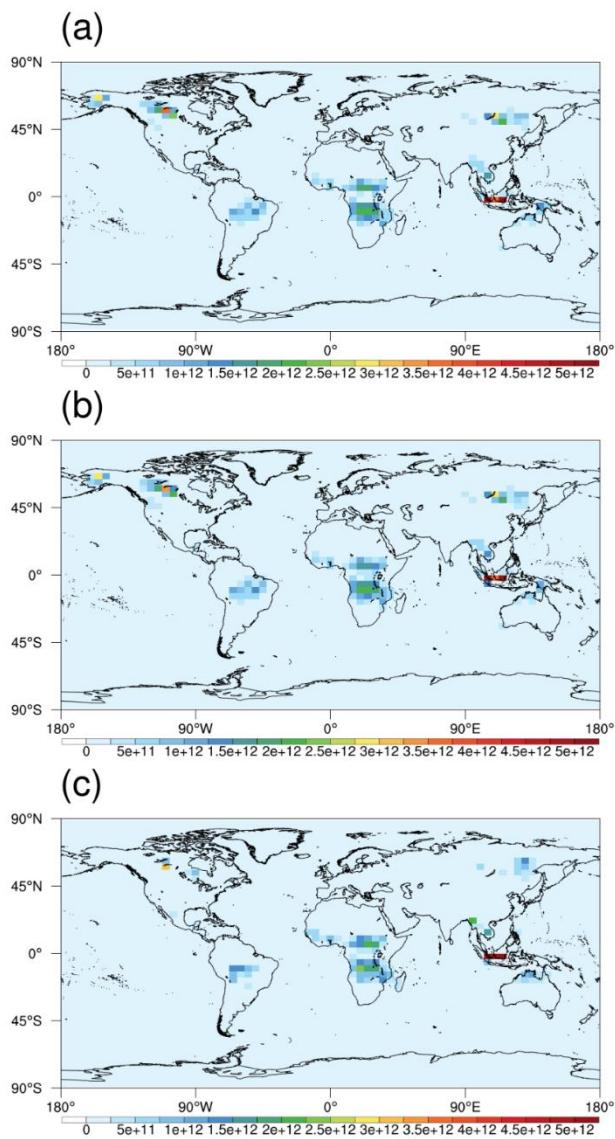
**Fig. S3.** NEI2011 CO emissions in 2015 from (a) GC-v12; (b) GC-Adjoint-HEMCO; and NEI2008 CO emissions in (c) GC-Adjoint-STD. The unit is molec/cm/s.



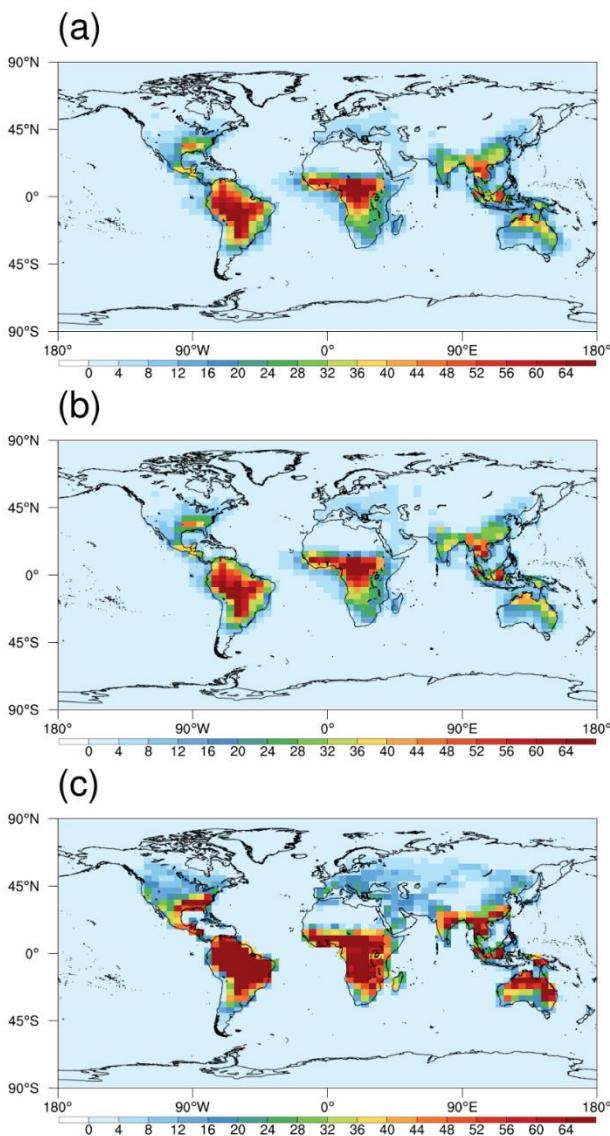
**Fig. S4.** APEI CO emissions in 2015 from (a) GC-v12; (b) GC-Adjoint-HEMCO; and CAC CO emissions in (c) GC-Adjoint-STD. The unit is molec/cm/s.



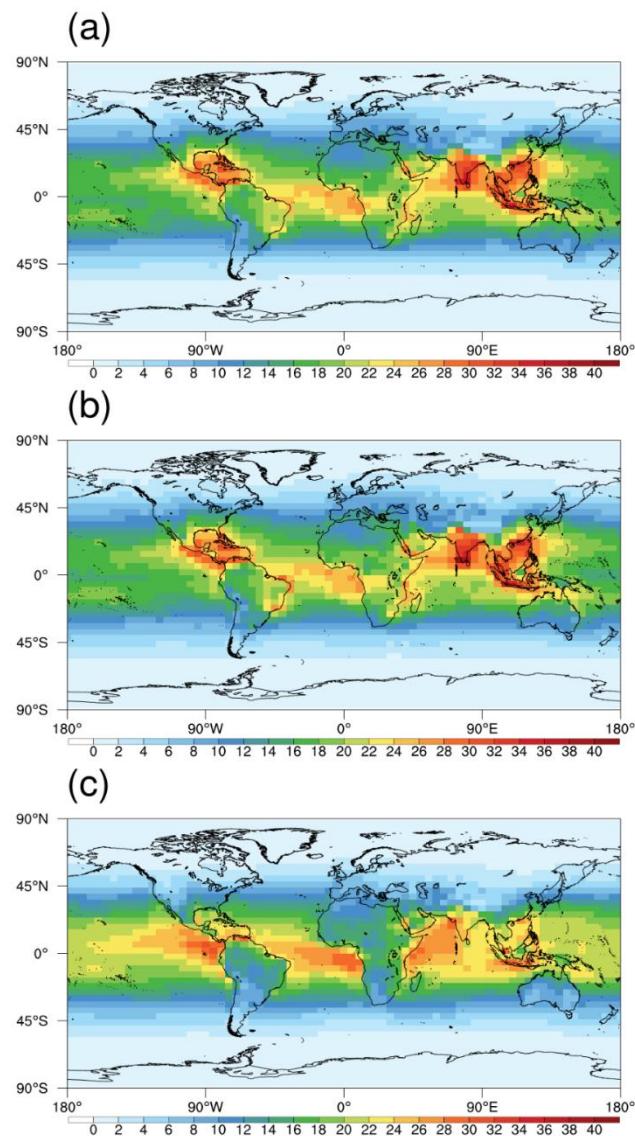
**Fig. S5.** DICE\_AFRICA+AF\_EDGAR CO emissions in 2015 from (a) GC-v12; (b) GC-Adjoint-HEMCO. The unit is molec/cm/s.



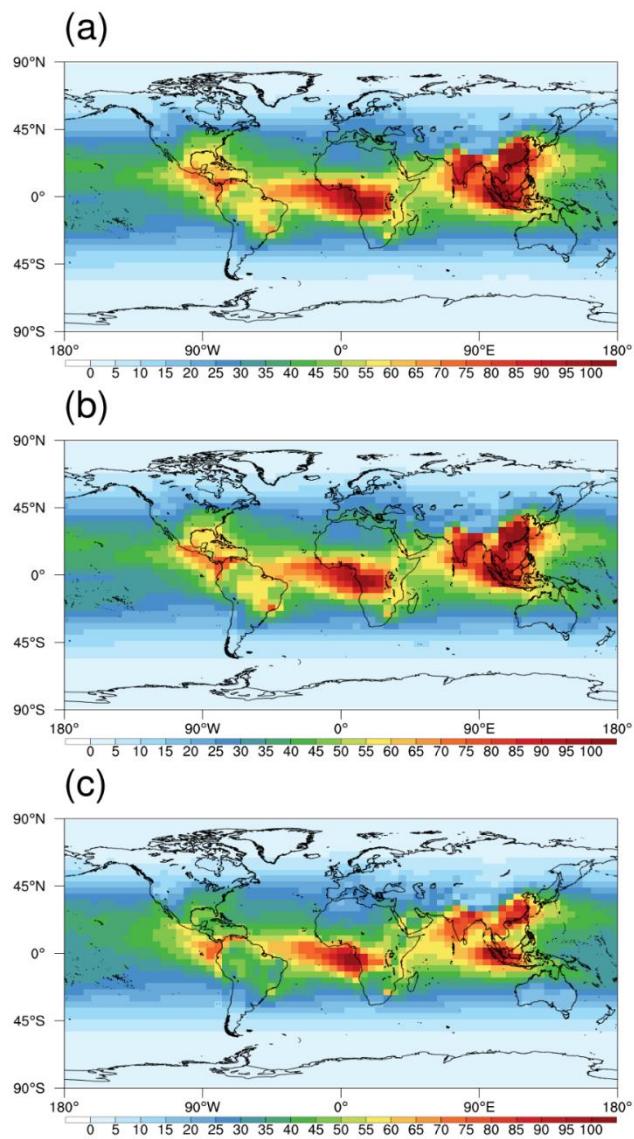
**Fig. S6.** GFED4 CO emissions in 2015 from (a) GC-v12; (b) GC-Adjoint-HEMCO; and GFED3 CO emissions in (c) GC-Adjoint-STD. The unit is molec/cm/s.



**Fig. S7.** PCO\_NMVOc columns from (a) GC-v12; (b) GC-Adjoint-HEMCO; (c) GC-Adjoint-STD in 2015. The unit is kg/s.



**Fig. S8.** PCO<sub>2</sub> columns from (a) GC-v12; (b) GC-Adjoint-HEMCO; (c) GC-Adjoint-STD in 2015. The unit is kg/s.



**Fig. S9.** CO\_OH columns from (a) GC-v12; (b) GC-Adjoint-HEMCO; (c) GC-Adjoint-STD in 2015. The unit is kg/s.