# Investigating the Utility of $CO_2$ and CO Analysis in Tracking Fossil Fuel $CO_2$

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# Manuscript in Review

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## Project Collaborators

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NASA ACMAP: Substantiating Key Synergies Between Air Quality (AQ) and Greenhouse Gas (GHG) Monitoring from Space: A Case for Anthropogenic $CO_2$ and $CH_4$ Constraints from CO and $NO_2$

funding from NASA

## Related IGAC Activity

AMIGO

Analysis of eMIssions usinG Observations

**A synthesis of research using observations-based analysis techniques that aim to better quantify emissions**

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# CO is mainly a product of incomplete combustion

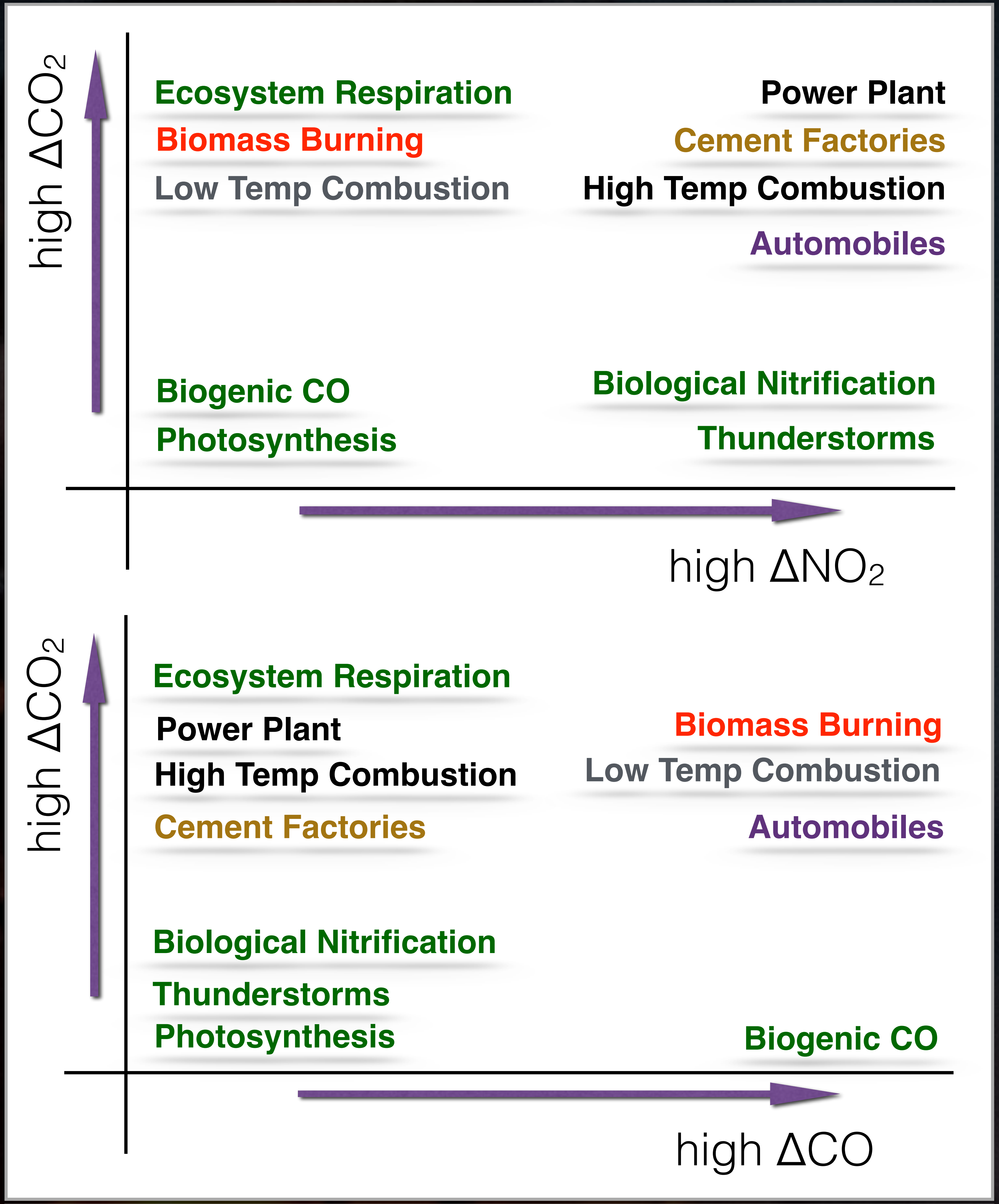
Fuel | Combustion Efficiency

$$C_{x_1}H_{x_2}O_{x_3}N_{x_4}S_{x_5} + n_1(1+e)(O_2 + 3.76N_2) \rightarrow$$
$$n_2CO_2 + n_3H_2O + n_4O_2 + n_5N_2 +$$
$$n_6CO + n_7NO + n_8NO_2 + n_9SO_2 + n_{10}C + \cdots$$

Combustion Products

Low Temperature Combustion: High CO, Low $NO_2$

High Temperature Combustion: Low CO, High $NO_2$, High BC




**Observations from AQ can provide information on efficiency and sectoral emission characteristics (at city scale and chemical weather scale).**

# CO plays a role in global AQ chemistry

$$\frac{d[CH_4]}{dt} = E_{CH_4} - (k_{CH_4+OH}[OH][CH_4])$$

$$P_{CH_4 \to CO} / \gamma_{CH_4 \to CO}$$

A loss in $CH_4$ is a gain in CO…

$$\frac{d[CO]}{dt} = (E_{CO} + P_{CH_4 \to CO} + P_{NMVOC \to CO}) - (k_{CO+OH}[OH][CO])$$

$$\frac{d[OH]}{dt} = P_{OH} - (k_{CO+OH}[OH][CO]) - (k_{CH_4+OH}[OH][CH_4]) - (k_{X+OH}[OH][X])$$

Main OH loss

…a loss in CO is a gain in $CO_2$

**Since CO (τ ~month) is right in the middle between OH (τ ~seconds) and $CH_4$ & $CO_2$ (τ ~years), one can suppose that AQ chemistry should matter at the scales of carbon weather**

# On AQ/GHG Synergies: Chemical Tracer Info

## Constraints from CO:

1) Help assess/monitor combustion efficiency (and MCE for fires)

2) Help identify errors in spatial and temporal distribution of emissions in the absence of $CO_2$ (and/or $CH_4$) data

3) Help track pollution plume transport at carbon weather scale (incl. convection)

4) Identify sectoral shifts/changes in fuel use/type (e.g., coal to natural gas)

5) Help provide more accurate representation of $CH_4$ loss

## Constraints from $NO_2$:

1) Help distinguish high temperature combustion activities (in the absence of lightning)

2) Help identify errors in spatial and temporal distribution of emissions in the absence of $CO_2$ (and/or $CH_4$) data

3) Help track pollution plumes (and transport) at city scale

4) Identify sectoral shifts/changes in fuel use/type (e.g., coal to natural gas)

5) Help provide more accurate representation of $CH_4$ loss (via OH source) and $O_3$

# Evaluating CAMS CO and $CO_2$ using KORUS-AQ

| | | Seoul | Taehwa | West Sea | Seoul–Jeju jetway | Seoul–Busan jetway | All |
|---|---|---|---|---|---|---|---|
| $dCO/dCO_2$ ($ppbv\ ppmv^{-1}$) | DC-8 measurement | $9.09 \pm 0.48$ | $15.3 \pm 0.56$ | $28.17 \pm 0.75$ | $10.37 \pm 0.31$ | $15.86 \pm 0.73$ | $13.29 \pm 0.21$ |
| | FC16s | $9.84 \pm 0.29$ | $14.31 \pm 0.40$ | $30.86 \pm 1.64$ | $13.00 \pm 0.27$ | $13.39 \pm 0.51$ | $12.28 \pm 0.15$ |
| | ANs | $8.21 \pm 0.45$ | $13.71 \pm 0.48$ | $30.60 \pm 1.73$ | $14.98 \pm 0.45$ | $12.68 \pm 0.47$ | $12.60 \pm 0.2$ |
| | FC9s | $11.56 \pm 0.62$ | $16.06 \pm 0.57$ | $32.44 \pm 1.77$ | $11.68 \pm 0.35$ | $13.87 \pm 0.54$ | $12.52 \pm 0.2$ |
| Correlation of CO and $CO_2$ | DC-8 measurement | 0.78 | 0.68 | 0.89 | 0.62 | 0.60 | 0.66 |
| | FC16s | 0.94 | 0.83 | 0.42 | 0.83 | 0.74 | 0.82 |
| | ANs | 0.77 | 0.71 | 0.25 | 0.61 | 0.76 | 0.63 |
| | FC9s | 0.78 | 0.70 | 0.36 | 0.60 | 0.73 | 0.65 |
| Correlation of $Bias_{CO}$ and $Bias_{CO_2}$ | FC16s | 0.90 | 0.61 | 0.80 | 0.46 | 0.55 | 0.61 |
| | ANs | 0.66 | 0.59 | 0.82 | 0.36 | 0.63 | 0.51 |
| | FC9s | 0.64 | 0.52 | 0.82 | 0.33 | 0.54 | 0.49 |

While we find overestimation in $CO_2$ (and underestimation in CO), CAMS shows a remarkable agreement in observed ratios (i.e., being able to capture the observed combustion contrast between Korea and China) . We also found that CAMS have issues at local-to-urban scale (e.g., weak PBL mixing). More importantly, we find that initial condition AND resolution matter in forecast skill of these species (Tang et al., ACP, 2018).

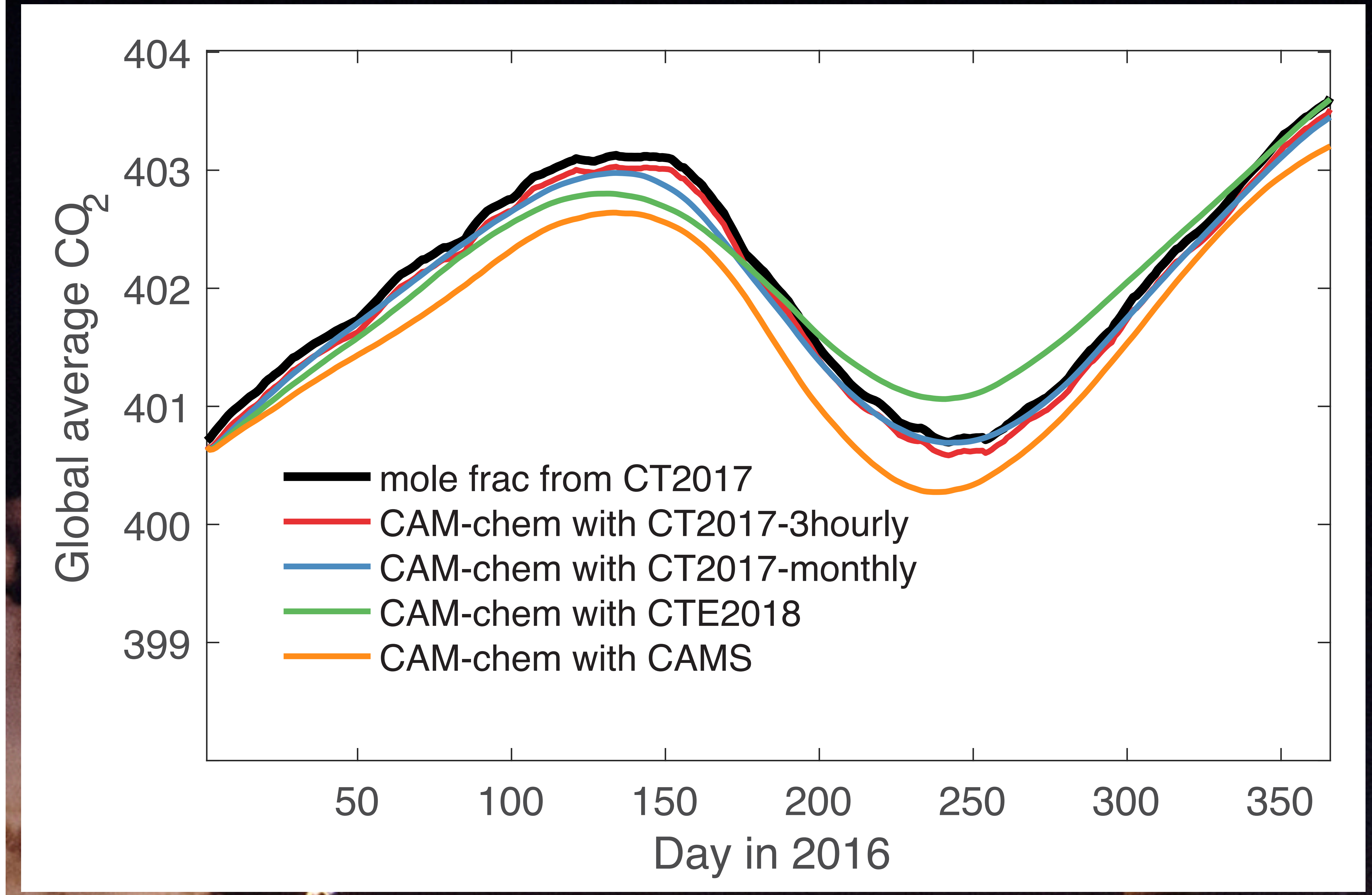
# Tractable Approach to Joint $CO_2$ and CO Simulation

| $CO_2$ fluxes | Spatial Res. | Temporal Res. | Period | Transport Model | Fossil Fuel Priors | Biosphere and Fires Priors | Ocean Priors | Main Reference |
|---|---|---|---|---|---|---|---|---|
| CAMS (v17r1) | 3.75° lon 1.875 ° lat | 3-hourly | 1979-2017 | LMDz[1] | EDGAR scaled to CDIAC | ORCHIDEE (climatology) + GFEDv4 | Landschüster et al. (2014) | Chevallier (2018)[2] |
| CT2017 | 1° lon 1° lat | 3-hourly monthly | 2000-2017 | TM5 | “Miller" (EDGAR scaled to CDIAC) & "ODIAC" | CASA w/ GFED 4.1s GFED_CMS | Jacobson et al. (2007) Takahashi et al. (2009) | Peters et al. (2007)[3] |
| CTE2018 | 1° lon 1° lat | monthly | 2000-2016 | TM5 | EDGAR+ IER scaled to CDIAC | SiBCASA-GFED4 | Jacobson et al. (2007) | van der Laan-Luijkx et al. (2017)[4] |

Posterior GHG fluxes are incorporated in CESM/CAM-Chem ‘CTM’. We use CO emissions based on ‘fine tuning’ HTAP prior emissions to match observed CO (Tang et al., JGR, 2019). However, ensemble of CO emissions (and $NO_2$) are planned. We also plan to incorporate other sources of posterior $CO_2$ (and $CH_4$) fluxes in our succeeding work (e.g., CMS flux, OCO-2 Flux MIP)

# Sanity Check: $CO_2$ and CO Budget during KORUS-AQ

| | | | $CO_2$ (PgC) | | | CO (TgC) |
|---|---|---|---|---|---|---|
| | | Region | CT2017 | CTE2018 | CAMS | |
| Sources | fossil fuel or anthropogenic | Korea | 0.01 | 0.01 | / | 0.11 |
| | | Japan | 0.02 | 0.03 | / | 0.13 |
| | | EA-S | 0.07 | 0.07 | / | 1.68 |
| | | EA-M | 0.11 | 0.11 | / | 2.71 |
| | | EA-N | 0.05 | 0.04 | / | 1.05 |
| | | the rest | 0.53 | 0.53 | / | 18.44 |
| | fire | | 0.11 | 0.11 | / | 9.69 |
| | biosphere | | / | / | / | 3.25 |
| | ocean | | / | / | / | 0.61 |
| | chemical production | | / | / | / | 58.40 |
| | source total | | 0.90 | 0.89 | / | 96.07 |
| Sinks | biosphere | | 0.63 | 0.90 | / | / |
| | ocean | | 0.26 | 0.18 | / | / |
| | chemical loss* | | / | / | / | 102.76 |
| | sink total | | 0.88 | 1.08 | / | 102.76 |
| Net (Sources-Sinks) | | | 0.01 | -0.19 | -0.04 | -6.69 |
| Initial Burden | | | 854.83 | 854.37 | 853.98 | 156.82 |
| Final Burden | | | 854.93 | 854.19 | 853.93 | 145.10 |
| Initial-Final | | | -0.10 | 0.18 | 0.05 | 11.72 |
| Budget delta | | | -0.08 | -0.01 | 0.01 | 5.03 |




CAM-Chem with CT2017 fluxes match CT2017 $CO_2$, providing some confidence in CAM-Chem (i.e., no significant bias in conserving mass and synoptic-scale transport). Incorporating GHG in AQ CTMs, enables us to probe AQ constraints on $FFCO_2$ (and $CH_4$).

# Tagging $FFCO_2$ and CO in CAM-Chem

$$\frac{\partial [X]^{itag}}{\partial t} = \left.\frac{\partial [X]^{itag}}{\partial t}\right|_{transport} + \left.\frac{\partial [X]^{itag}}{\partial t}\right|_{sources} - \left.\frac{\partial [X]^{itag}}{\partial t}\right|_{sinks} \qquad f_{CO_2}^{itag} = f_{CO_2} \cdot \left( {[CO_2]_{srf}^{itag}} \middle/ {[CO_2]_{srf}} \right)$$

&


This enables us to examine regional/sectoral emission contributions. While $CO_2$ appears to be broadly consistent in spatial pattern with CO, $FFCO_2$ is even more correlated to CO, suggesting (qualitatively) that CO might be a good tracer of $FFCO_2$ (esp. useful in the absence (or lack) of $CO_2$ data).

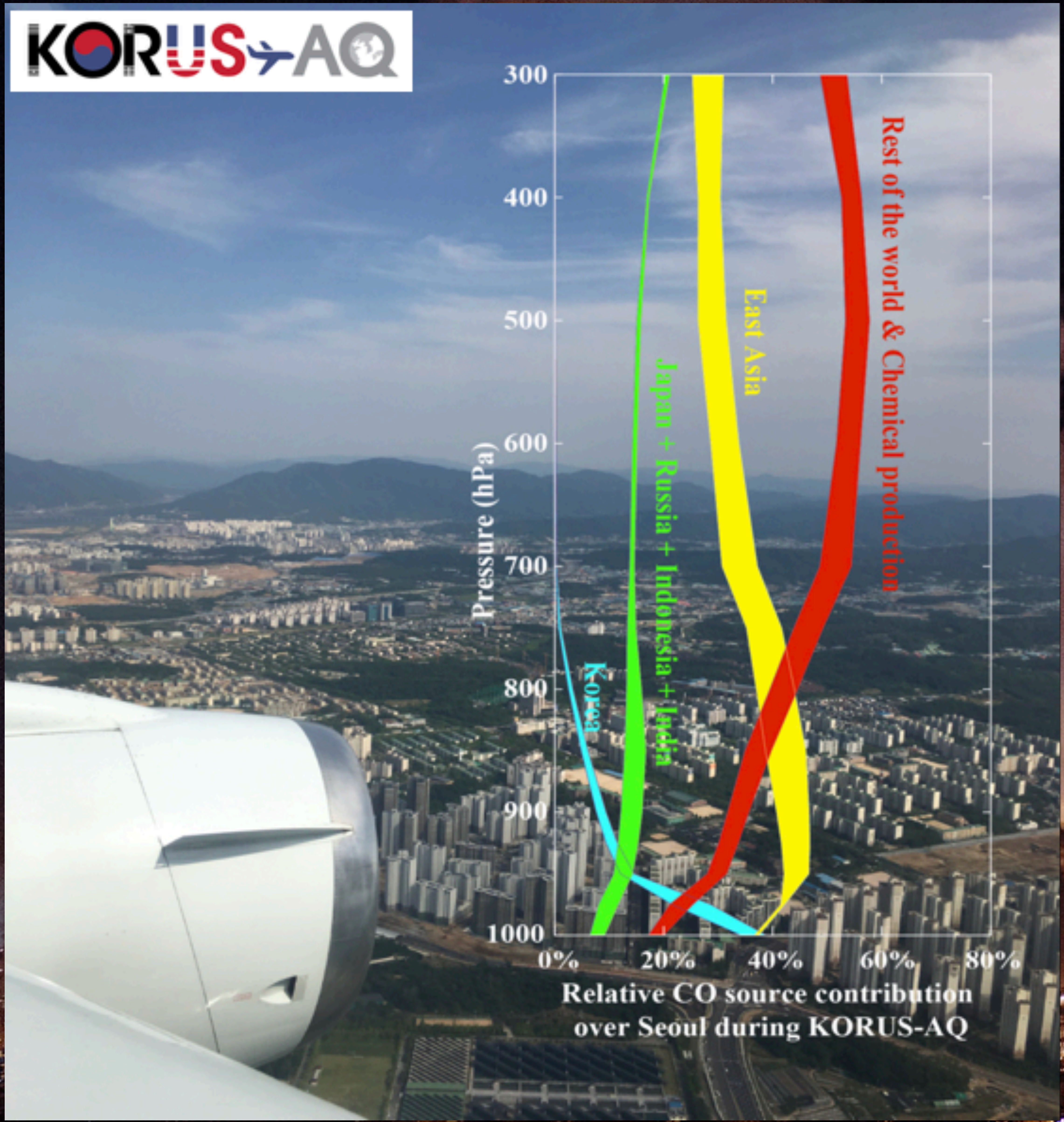
# Evaluating CAM-Chem CO and $CO_2$ using KORUS-AQ

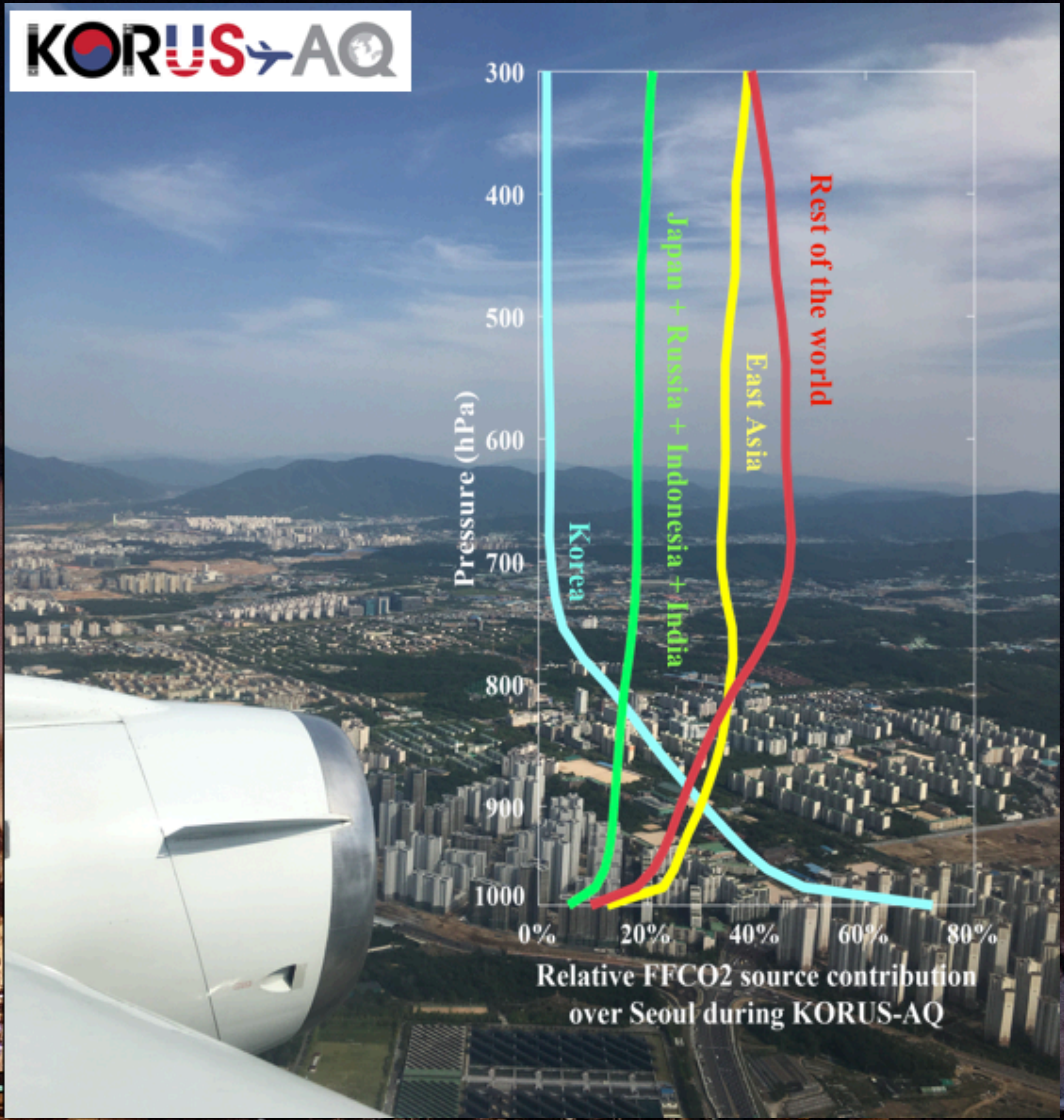
| | | NOAA/ESRL CCGG | | | | TCCON | | | | NASA DC-8 KORUS-AQ | | | | | OCO-2 MOPITT |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | AMY | LLN | UUM | WLG | Amy | Sag | Tsu | Rik | Seoul | Taehwa | West Sea | Seoul Jeju | Seoul Busan | Study Domain |
| Obs Mean | $CO_2$ | 415 | 407 | 406 | 405 | 403 | 406 | 403 | 403 | 415 | 408 | 411 | 411 | 408 | 405 |
| | CO | 217 | 124 | 142 | 130 | 109 | 108 | 103 | 99 | 266 | 163 | 234 | 223 | 183 | 111 |
| Obs Std | $CO_2$ | 12 | 3 | 6 | 3 | 3 | 2 | 2 | 3 | 13 | 5 | 5 | 10 | 4 | 2 |
| | CO | 67 | 55 | 26 | 26 | 8 | 15 | 14 | 15 | 113 | 73 | 143 | 101 | 64 | 19 |
| Obs $R_{CO2,CO}$ | | 0.32 | 0.31 | 0.48 | 0.36 | 0.59 | 0.52 | 0.37 | 0.28 | 0.79 | 0.68 | 0.89 | 0.62 | 0.60 | 0.22 |
| Obs $dCO/dCO_2$ | | 5.90 | 18.90 | 4.53 | 9.42 | 2.86 | 7.40 | 5.63 | 4.81 | 9.13 | 15.28 | 28.20 | 10.37 | 15.92 | 11.90 |
| Model Mean | $CO_2$ | 414 | 405 | 405 | 406 | 403 | 405 | 404 | 403 | 411 | 407 | 408 | 411 | 408 | 405 |
| | CO | 239 | 142 | 129 | 187 | 105 | 111 | 102 | 93 | 237 | 143 | 202 | 213 | 155 | 118 |
| Model Std | $CO_2$ | 6-8 | 2 | 6-8 | 5-7 | 2-3 | ~2 | ~2 | 3-4 | 6-11 | 2-4 | 2-4 | 7-10 | 2-6 | 1-2 |
| | CO | 124 | 103 | 52 | 173 | 12 | 19 | 17 | 20 | 133 | 70 | 119 | 117 | 62 | 27 |
| Model $R_{CO2,CO}$ (min/max) | | -0.12 | 0.46 | 0.16 | 0.40 | -0.2 | 0.51 | 0.33 | 0.05 | 0.56 | 0.21 | -0.10 | 0.65 | 0.25 | 0.25 |
| | | 0.18 | 0.70 | 0.27 | 0.71 | -0.1 | 0.54 | 0.44 | 0.29 | 0.73 | 0.60 | 0.76 | 0.81 | 0.66 | 0.41 |
| Model $dCO/dCO_2$ (min/max) | | 21.01 | 48.85 | 6.64 | 33.88 | NaN | 9.53 | 7.43 | 5.57 | 12.61 | 16.56 | 33.66 | 11.54 | 10.68 | 16.96 |
| | | 26.17 | 59.80 | 8.68 | 44.47 | | 11.24 | 8.67 | 7.53 | 20.91 | 30.91 | 48.28 | 16.08 | 26.79 | 27.05 |

# Simulated and Observed $FFCO_2$ during KORUS-AQ

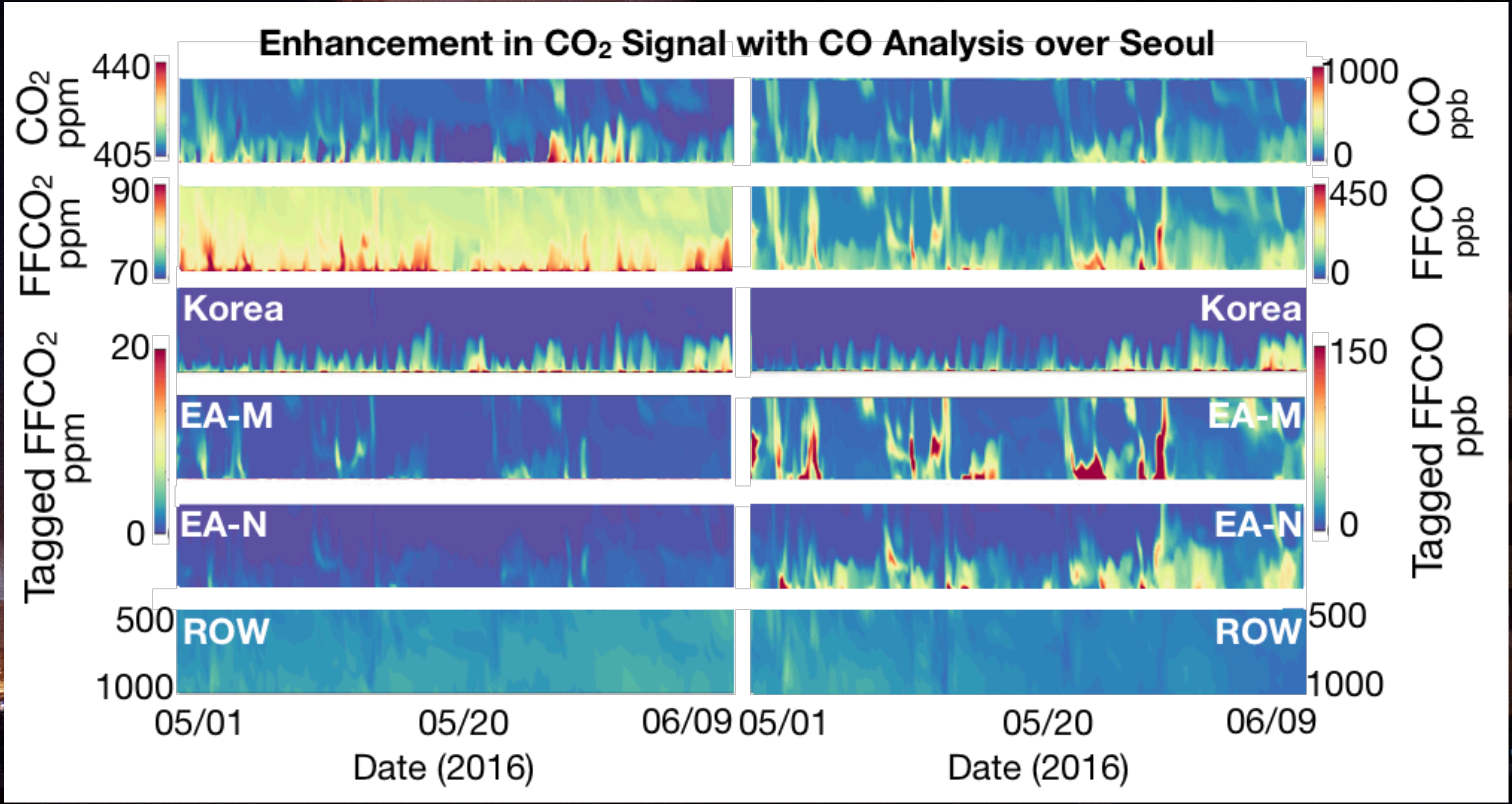

CAM-Chem tagged $FFCO_2$ shows good overall agreement with derived $FFCO_2$ from radiocarbon samples. Again, we find that CO is more correlated with $FFCO_2$ than $CO_2$, suggesting that CO data can be useful to track $FFCO_2$. Our ability to estimate regional contributions may be useful for complementary $FFCO_2$ inversions (e.g. chemical downscaling).

# Profiles of Tagged CO and $FFCO_2$ over Seoul



# CO as a Chemical Tracer of $FFCO_2$ Transport



# Info on Combustion Efficiency from Tagged Tracers

# CHIME

**CHemical Inverse Modeling system Experiments**

**An intercomparison activity on current chemical data assimilation and inverse modeling systems (a.k.a. TransCom-X)**

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## Research Objectives

1. Assess the state of CO as inferred from these reanalyses
2. Characterize associated uncertainties incl. estimates of emissions and other drivers of CO trends

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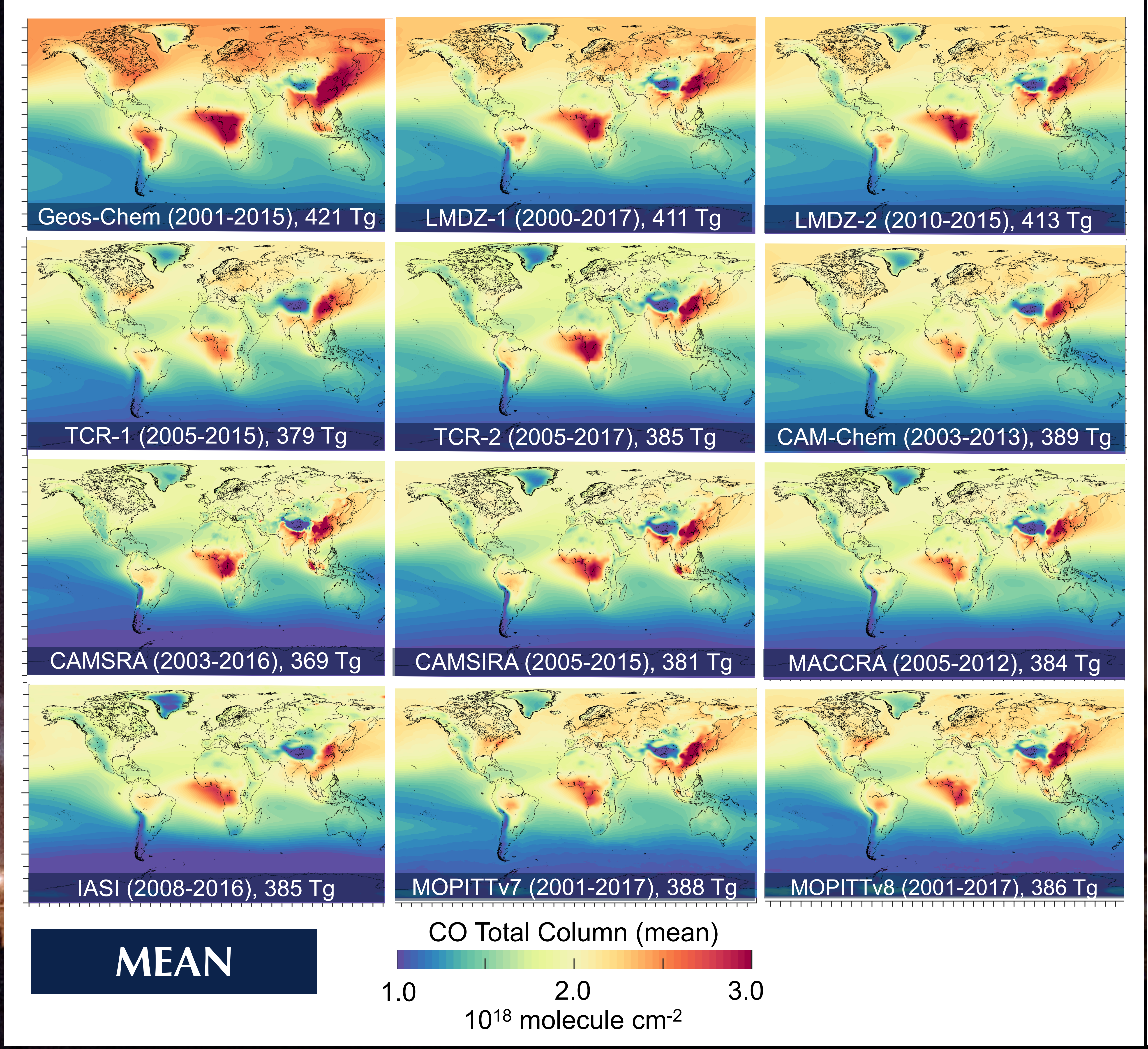
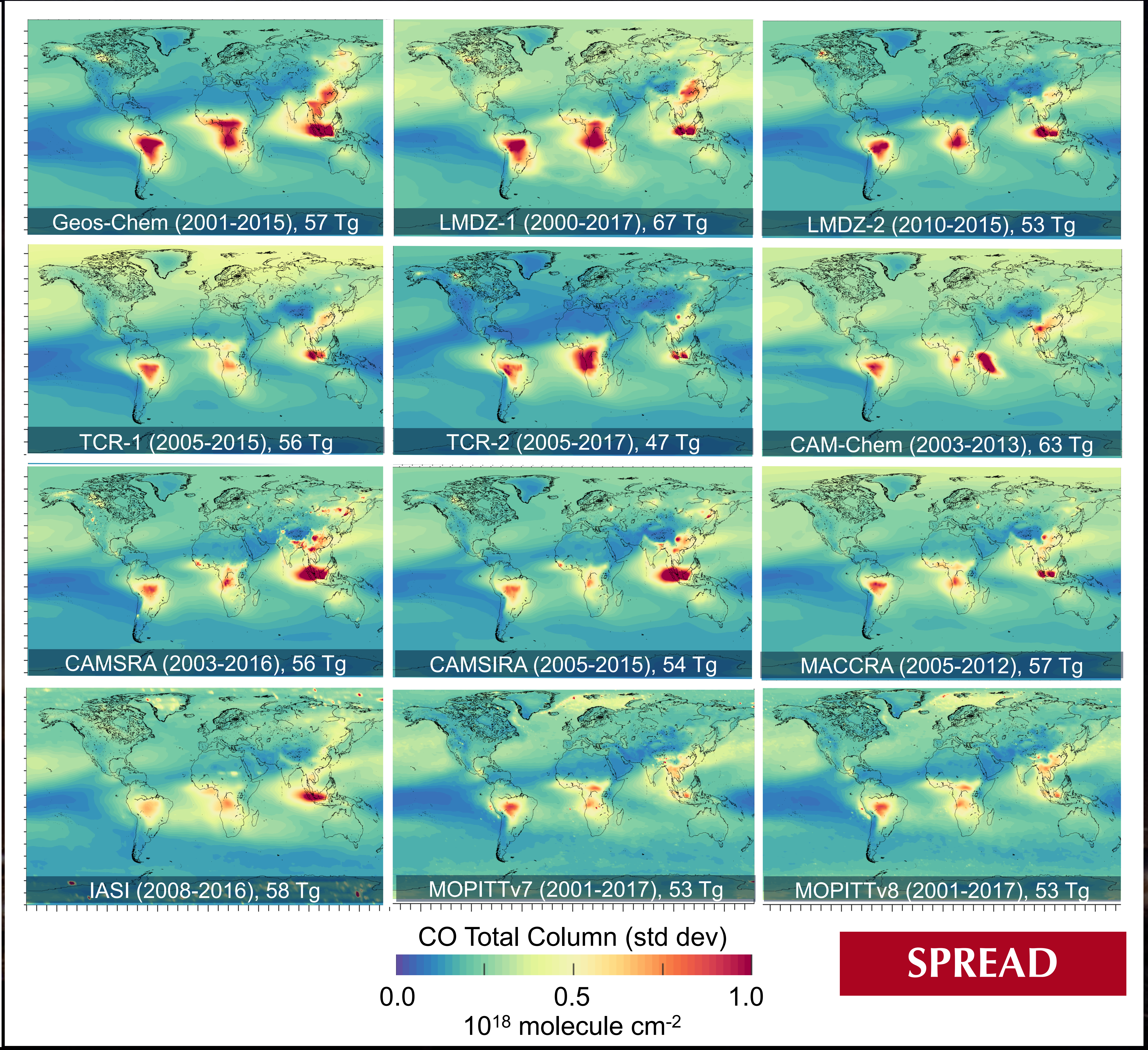
Potential Application: Provide an ensemble of OH fields (for $CH_4$), constraints on fire $CO_2$

| Product | Model ***** | Resolution* lonº x latº lev | Period ** | Emissions *** | Update **** | CO Obs ***** | Algorithm ****** | Main Reference |
|---|---|---|---|---|---|---|---|---|
| **CAMSiRA** | IFS/CY40R2 C-IFS CB05 | ~1.1 x ~1.1 (T159) 60 lev | 2003-2015 | MACCity, GFASv1.2 MEGAN2.1 | State | MOPITTv5 | 4D-Var IFS/CY40r2 | Flemming et al., ACP, (2017) |
| **CAMSRA** | IFS/CY42R1 MOZART-3 | ~1.-0 x ~1.0 (T255) 60 lev | 2003-2016 | MACCity GFED&GFASv0 MEGAN2.1 | State | MOPITTv6 | 4D-Var IFS/CY42r1 | Inness et al., ACP, (2019) |
| **MACCRA** | IFS/CY36R2 MOZART-3 | ~1.1 x ~1.1 (T255) 60 lev | 2003-2012 | MACCity GFED&GFASv0 MEGAN2.1 | State | MOPITTv4 & IASI | 4D-Var IFSCY36r2 | Inness et al., ACP, (2013) |
| **CAM-Chem** | CESM1.2/ CAM-Chem | 2.5 x ~1.9 26 lev | 2002-2013 | MACCity FINN1.5 MEGAN2.1 | State | MOPITTv5 | EAKF DART | Gaubert et al., JGR, (2016) |
| **Geos-Chem** | GEOS-Chem | 5 x 4 30/47 lev | 2001-2015 | EDGAR3.2 FT2K GFED3 MEGAN2.0 | Source | MOPITTv6 | 4D-Var | Jiang et al., ACP, (2017) |
| **TCR-1** | CHASER | ~2.8 x ~2.8 32 (26) lev | 2005-2012 | EDGAR4.2 GFED3.1 GEIA | State & Source | MOPITTv6 | LETKF | Miyazaki et al., ACP, (2015) |
| **TCR-2** | MIROC-Chem | ~1.1 x ~1.1 32 (26) lev | 2005-2017 | EDGAR GFED GEIA | State & Source | MOPITTv7 | LETKF | Miyazaki et al., ACP, in prep |
| **LMDZ-1** | LMDz SACS | ~ 3.7 x ~1.9 39 lev | 2000-2017 | MACCity GFED4s MEGAN/POET | Source | MOPITTv7 | 4D-Var PYVAR | Zheng et al., ERL, (2018) |
| **LMDZ-2** | LMDz v4 SACS | ~3.7 x ~1.9 32 lev | (2002-2011) 2010-2015 | MACCity GFED3.1 LMDz-INCA | Source | MOPITTv6 | 4D-Var PYVAR | Yin et al., ACP, (2015) |

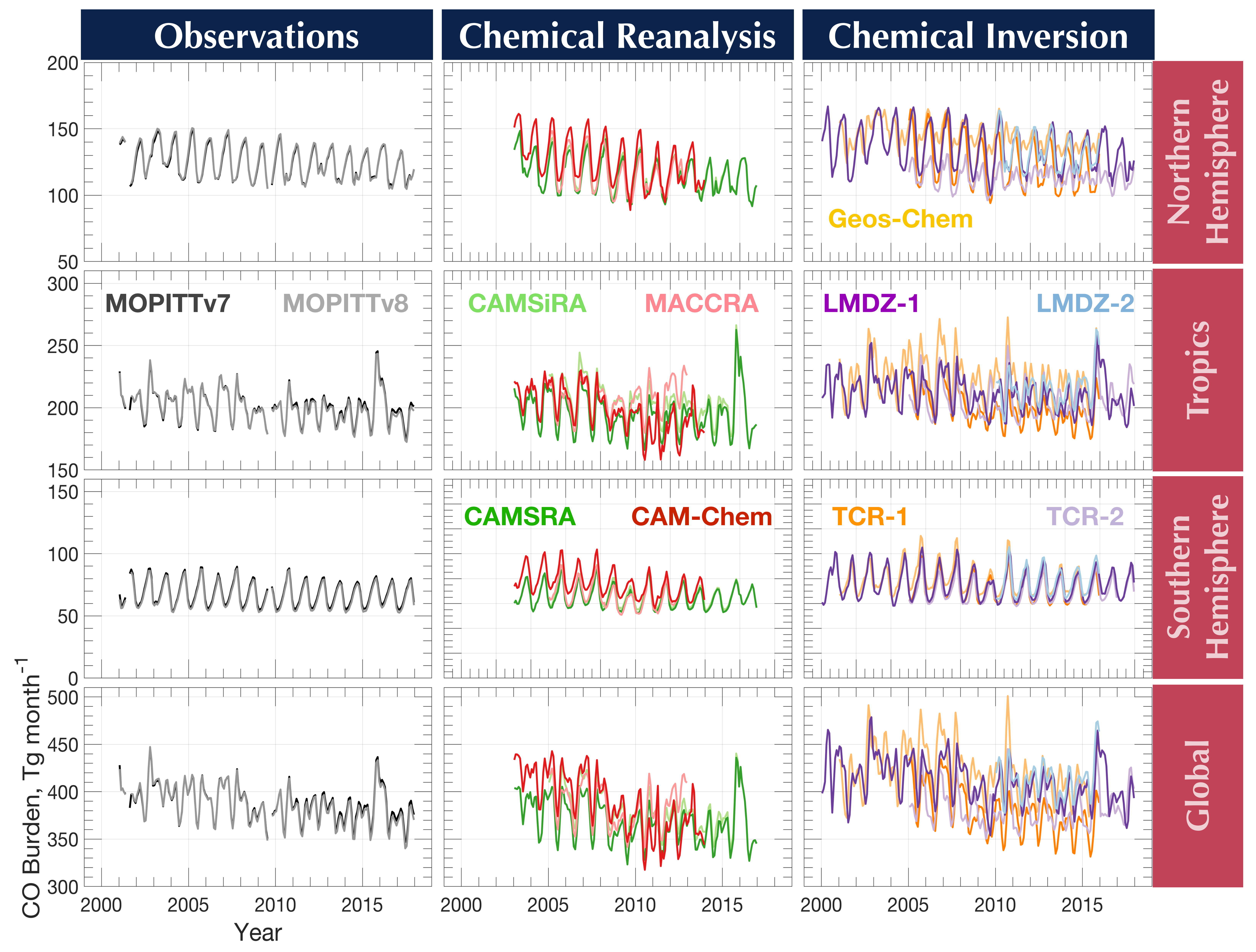
* approximate resolution based on post-processing from the product team or as requested
** published original period may be longer, period as requested, use of full year for starting year
*** base anthro/fire emission, actual may be an updated version, see details in main reference
**** can be multi-species

# Spatial Distribution of CO Burden

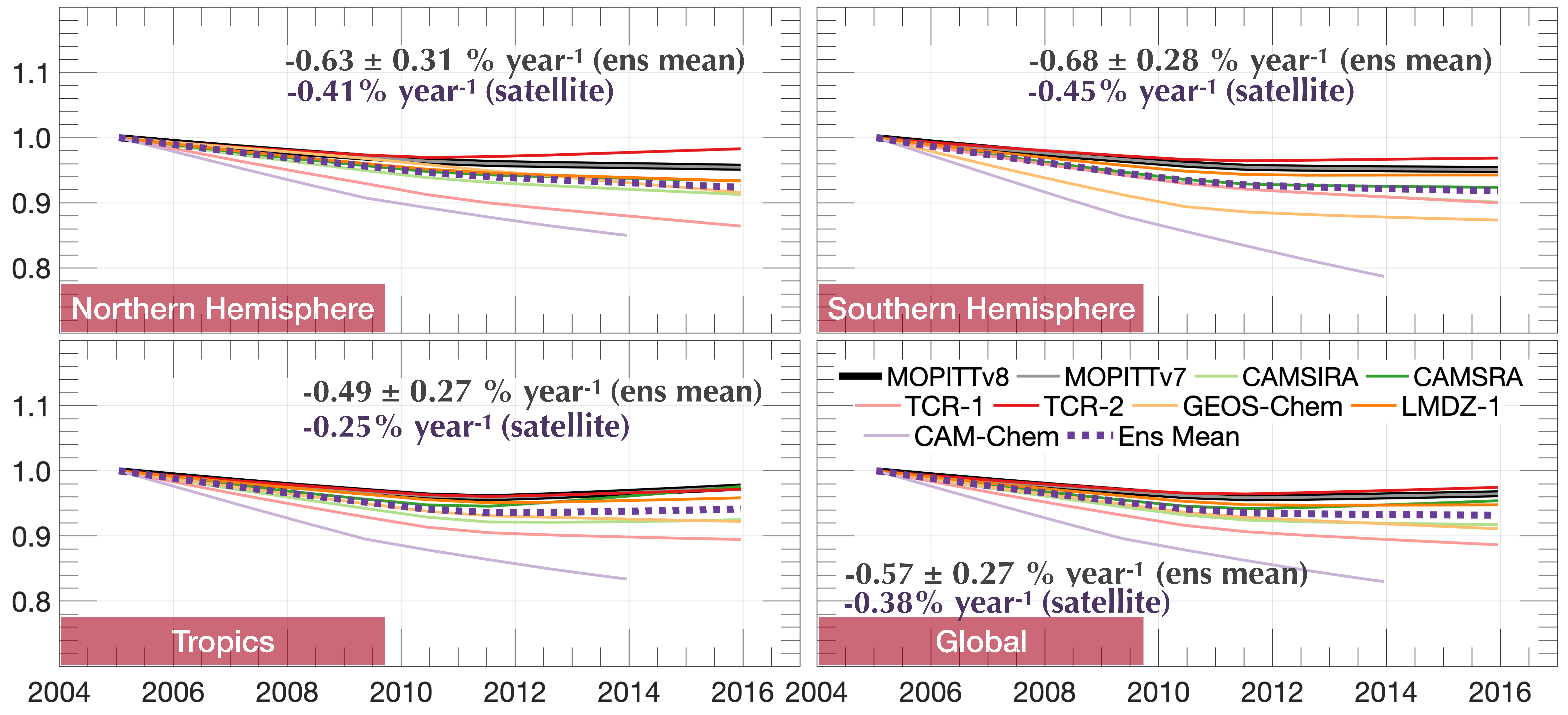


# Temporal Distribution of CO Burden



# Long-term Trend Relative to 01/2005 (satellite)



# Mean Seasonality of CO Abundance

# Interannual Variability of CO Abundance

# Summary:

- CO and $NO_2$ data can be used as chemical tracers of $FFCO_2$, as well as constraints on tropospheric $O_3$ chemistry (via OH on $CH_4$ loss).

- Careful consideration on how these observational constraints are translated to top-down emission estimates is warranted.

- Development of a tractable, ensemble-based, GHG/AQ system which uses posterior fluxes from inversions and reanalysis is on-going; Has potential application with TROPOMI, GOSAT 2/3, GeoCarb, and OCO-2/3 for example. Collocated in-situ and airborne measurements of these species are critical.

- They should be considered as auxiliary/supplementary datasets in tracking $FFCO_2$ and $CH_4$ and in reducing uncertainties on associated emission estimates (esp. w/ regards to consistency across species).

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