10 years of MOPITT reanalysis

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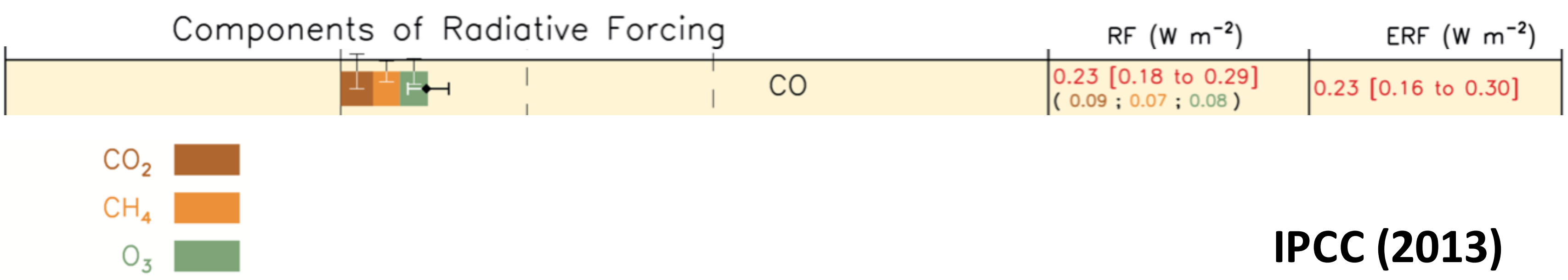
Motivations for Satellite CO assimilation and reanalysis
Atmospheric composition, Air quality & Climate

- **CO is an indirect greenhouse gas 0.23 W.m-2**
  - ✓ **Precursor of tropospheric ozone (0.08 $W.m^{-2}$)**
  - ✓ **Controlling CH_4 lifetime (0.07 $W.m^{-2}$)**
  - ✓ **CO_2 precursor (0.09 $W.m^{-2}$)**
- **Important for Air quality and Chemistry**
  - ✓ **Track pollution plumes, measuring emissions, oxidative capacity …**



IPCC (2013)

Understanding the CO budget and trends : Observations

- **Global surface network from the 90s**
- **Continuous satellite measurements since 2000**
  - ➢ Decreasing trends in concentrations, likely because decrease in Anthropogenic emissions
  - ➢ BB emissions cause a large interannual variability

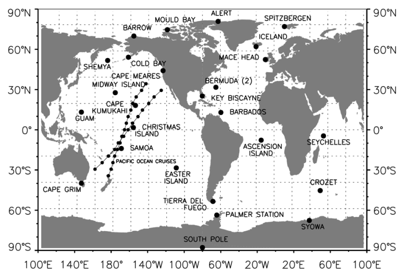


Figure 6. Map of the sites in the NOAA/CMDL cooperative air sampling network used in constructing the globally averaged CO surface.

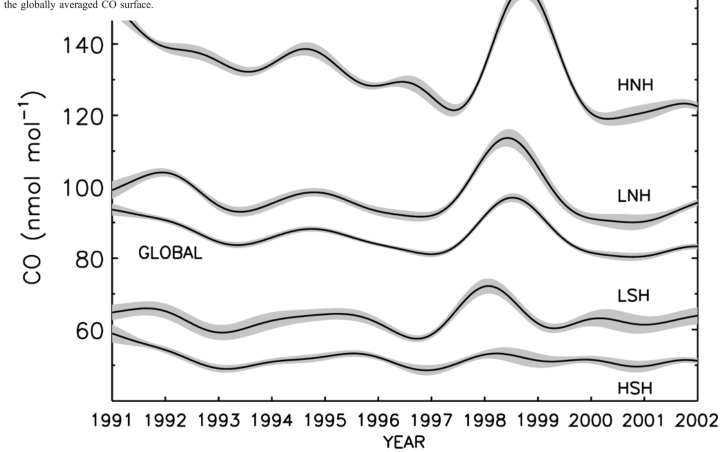


Figure 11. Global and zonal averaged trends extracted from the global surface. The trend is shown as the solid line; the shading indicates the 1 sigma uncertainty.

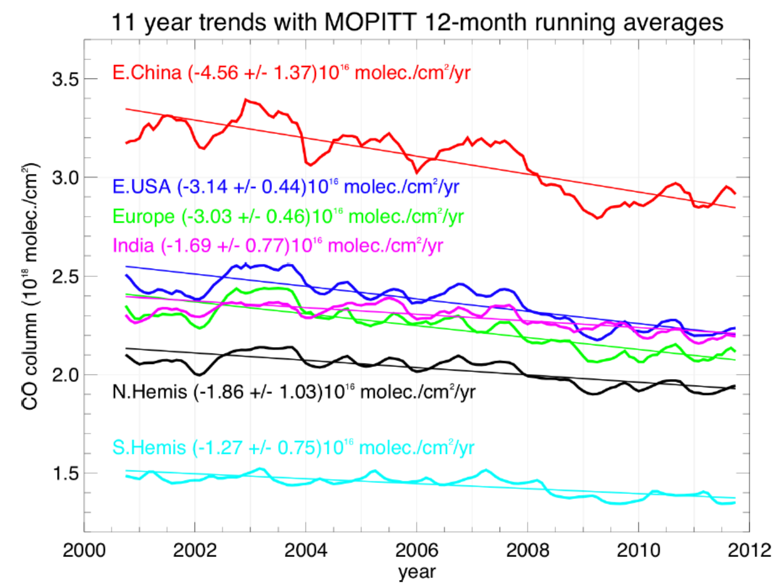
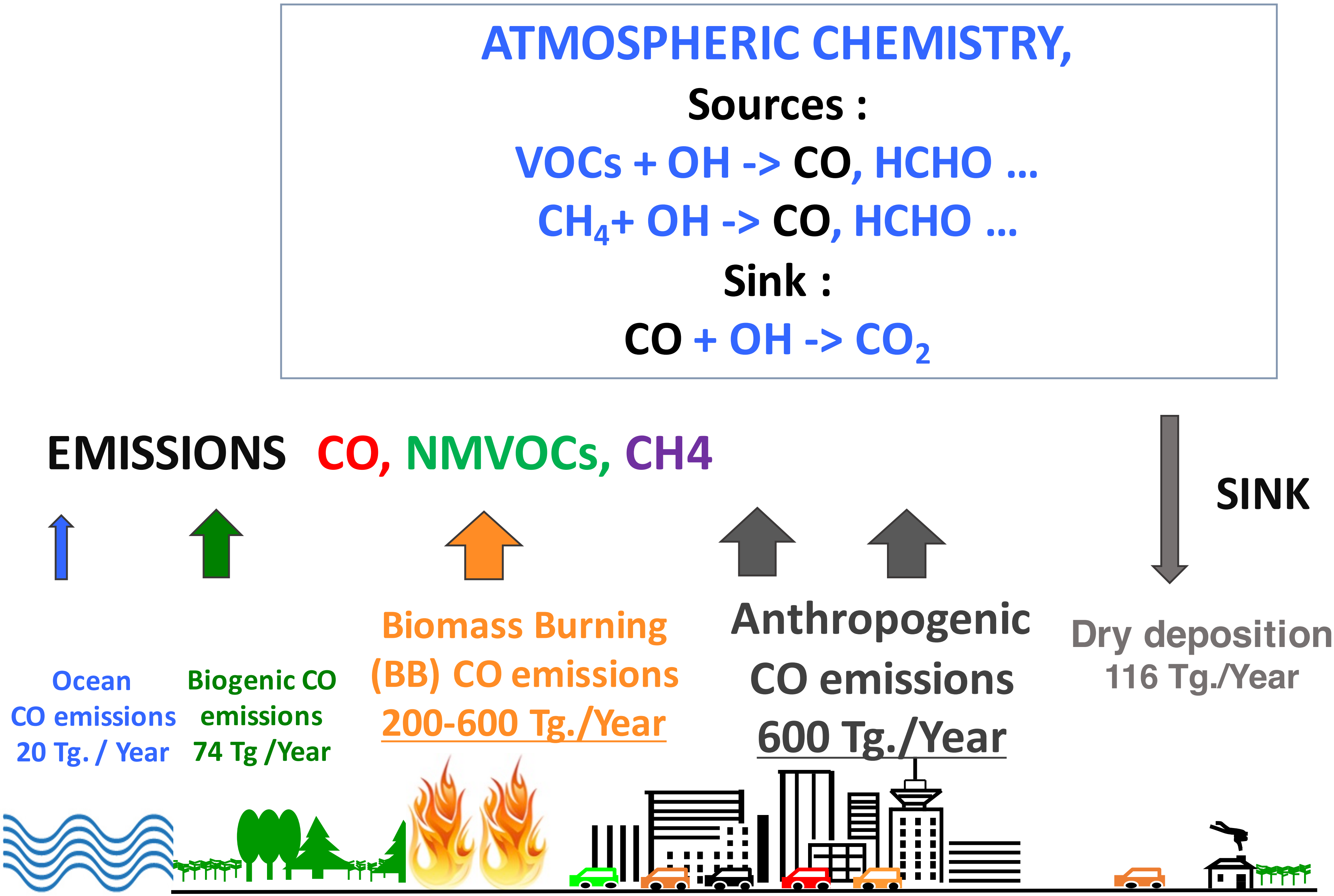


Fig. 5. MOPITT 11-yr regional trends in total CO column change per year. Regional time series with 12-month running average and corresponding linear trends are indicated by the different colors, with slope and 1σ error given in molecules/cm^2 yr^{-1}.

Novelli et al. 1998, 2003 / Worden et al. 2013

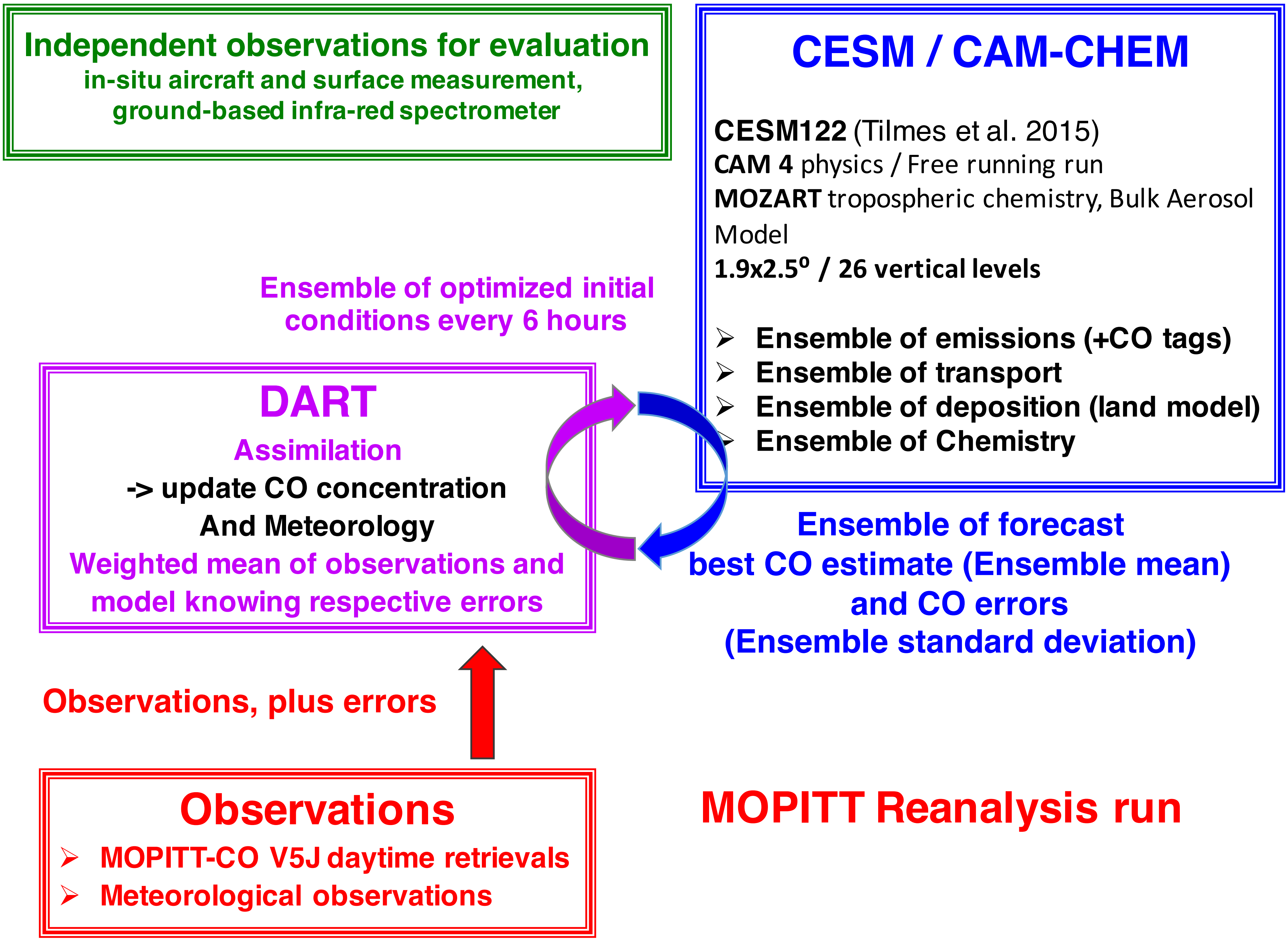
CO Budget


ATMOSPHERIC CHEMISTRY,
Sources :
VOCs + OH -> CO, HCHO …
CH_4+ OH -> CO, HCHO …
Sink :
CO + OH -> CO_2
EMISSIONS CO, NMVOCs, CH4
SINK
Ocean CO emissions 20 Tg. / Year
Biogenic CO emissions 74 Tg /Year
Biomass Burning (BB) CO emissions 200-600 Tg./Year
Anthropogenic CO emissions 600 Tg./Year
Dry deposition 116 Tg./Year

Understanding the CO budget and trends : Recent Reanalyzes

Reanalysis of Chemical composition	DA method / Optimization	CO Observations	Model, DA chain	Additional observations	Coupling
Miyazaki et al. 2015	LETKF / Total CO emissions	MOPITT V6T, only 700 hPa level	CHASER-DAS	TES O3, MLS O3, HNO3, OMI NO2	Offline
Yin et al. 2015	4D-Var / Total CO emissions & Chemical production	MOPITT V6J, total CO columns	LMDz-INCA, PYVAR-SACS	Surface CH4, Methyl-Chloroform	Offline
Inness et al. 2013	4D-Var / CO concentrations	MOPITT V4, total CO colums	IFS-MOZART	IASI-CO, MLS O3, SCIAMACHY O3 and NO2…	Coupled
Gaubert et al. 2016	EAKF / CO concentrations	MOPITT V5J, CO profiles	DART/CAM-Chem	Conventional Met Obs	Online

- Optimize emissions
- Optimize concentrations (Atmospheric burden)
- Optimize the budget, both emissions and sinks


Independent observations for evaluation
in-situ aircraft and surface measurement,
ground-based infra-red spectrometer
CESM / CAM-CHEM
CESM122 (Tilmes et al. 2015)
CAM 4 physics / Free running run
MOZART tropospheric chemistry, Bulk Aerosol Model
1.9x2.5° / 26 vertical levels
Ensemble of emissions (+CO tags)
Ensemble of transport
Ensemble of deposition (land model)
Ensemble of Chemistry
Ensemble of optimized initial conditions every 6 hours
DART
Assimilation
-> update CO concentration
And Meteorology
Weighted mean of observations and model knowing respective errors
Ensemble of forecast
best CO estimate (Ensemble mean)
and CO errors
(Ensemble standard deviation)
Observations, plus errors
Observations
MOPITT-CO V5J daytime retrievals
Meteorological observations
MOPITT Reanalysis run

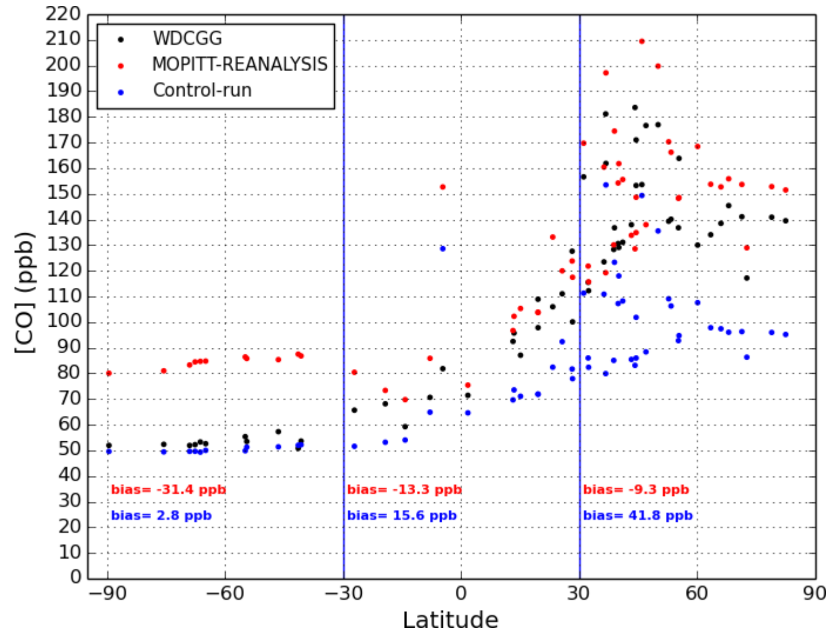
Outline

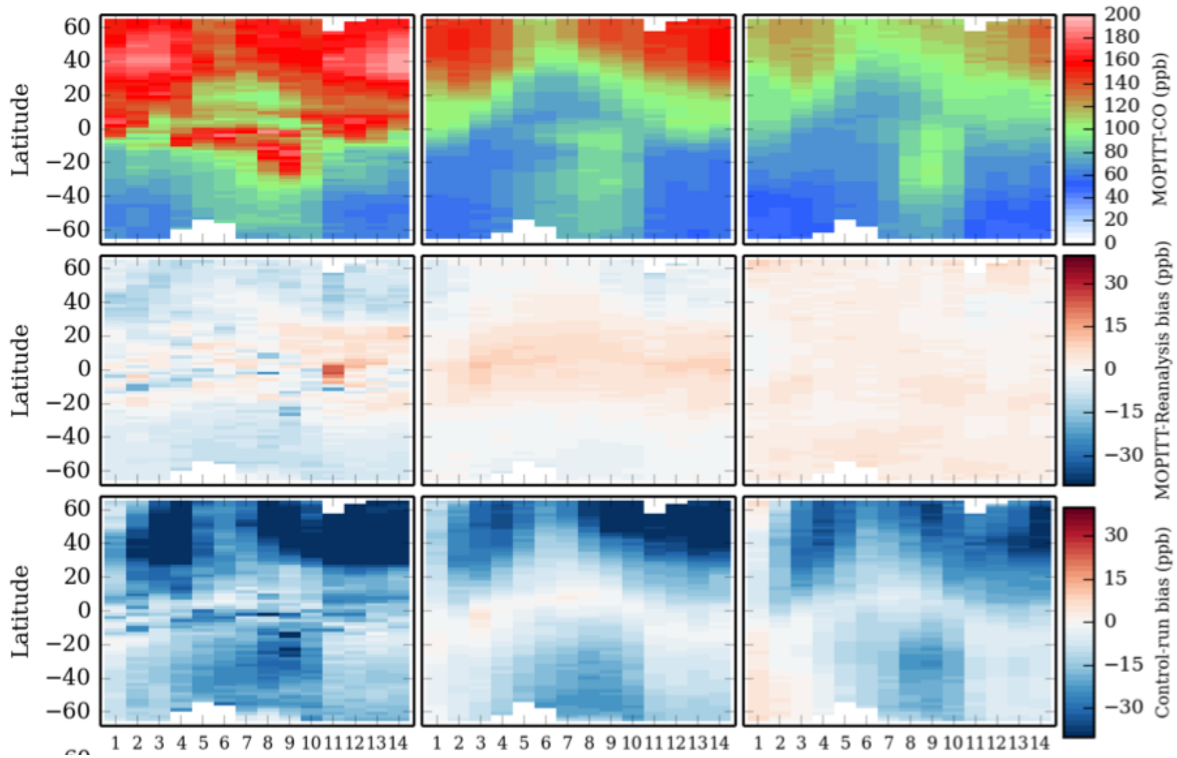
- Evaluation of the impacts of the first year of the reanalysis
  - ✓ Evaluation of the chemical response of the MOPITT assimilation
- Use of the metrics to understand the change over the decade 2002-2012
  - ✓ Verification datasets to evaluate the CO simulations

Impact of CO assimilation

1. Increase CO

- Increase of CO, correcting the NH winter bias.
- Increase of tropical CO during the fire season
- Leads to too much CO in the SH.





MOPITT

MOPITT-Reanalysis - MOPITT

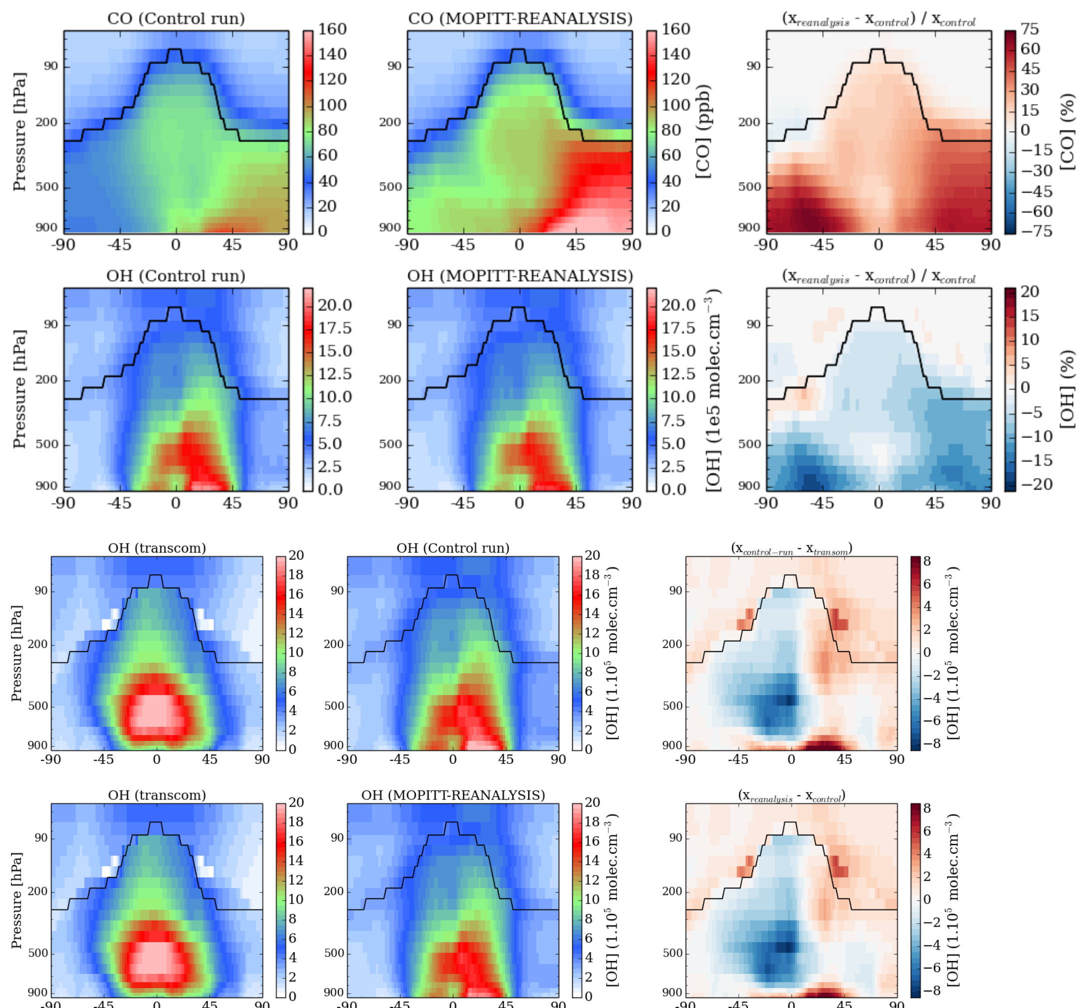
Control-run - MOPITT

Chemical response

2. Reduce OH concentration

	Model configurations	CH_4 lifetime (yr)
Tilmes et al. 2015	CAM5-Chem	8.4
	CAM5-Chem (SD)	7.83
	CAM4-Chem	8.82
	CAM4-Chem (SD)	8.4
This study	CAM4-Chem (DART)	8.7
	MOPITT Reanalysis	9.3

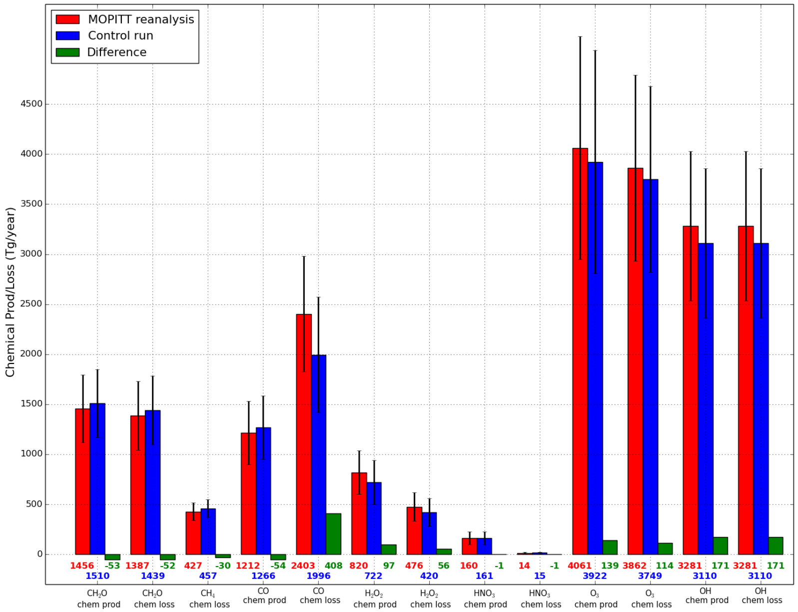
- Increase of the CH_4 lifetime
- In both cases OH seems to be too high at high latitudes.
- According to Transcom-OH is overestimated at high latitudes and underestimated at the tropics.

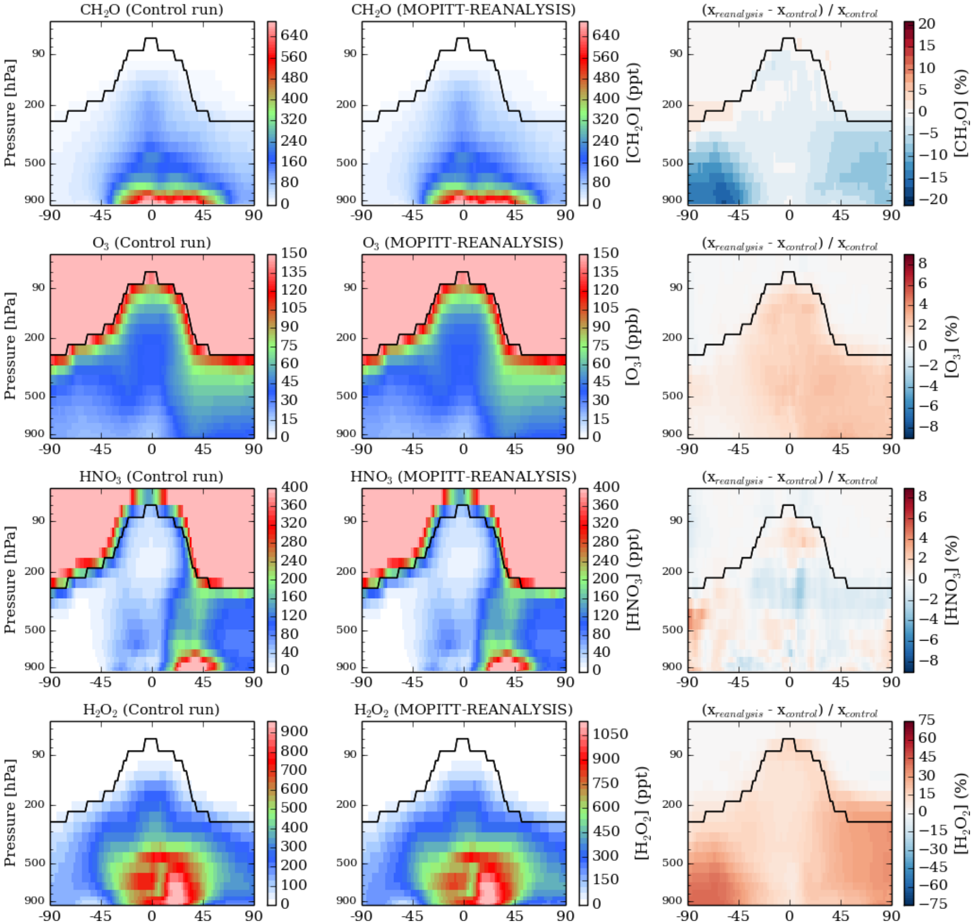


Chemical response

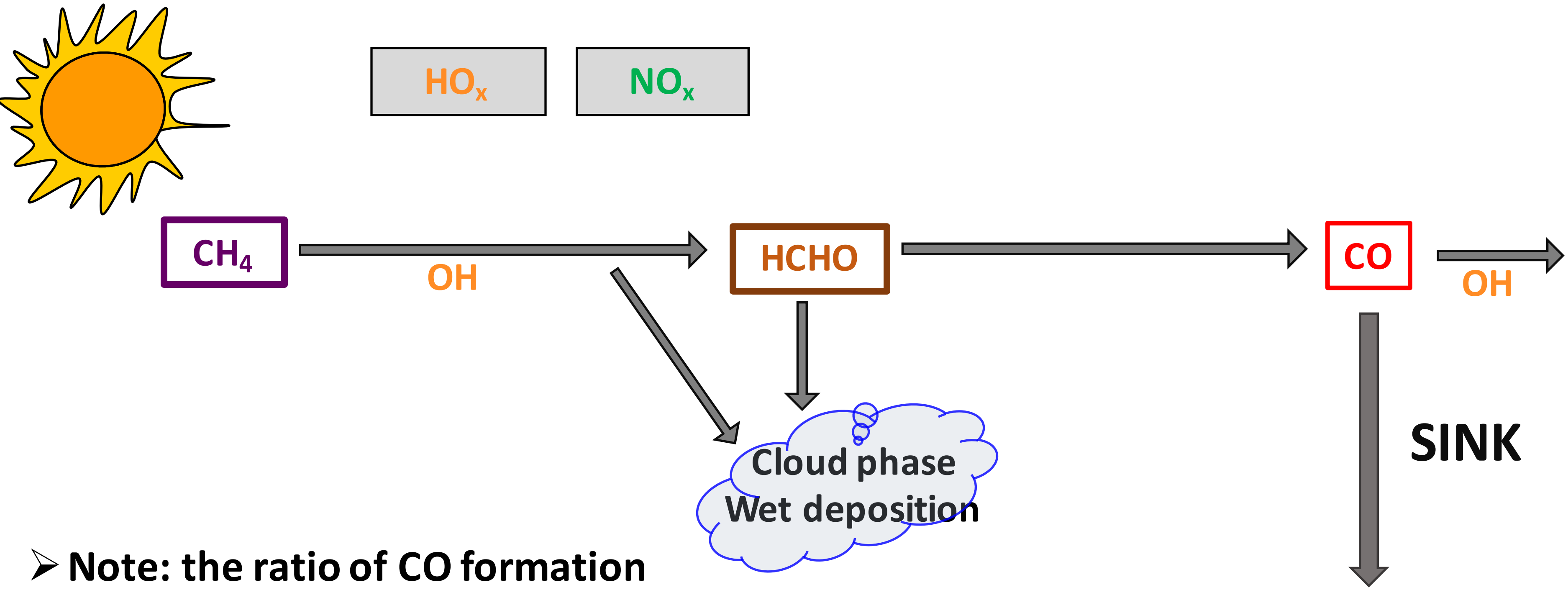
3. Increase O_3 and OH chemical production

- In presence of NOx, increase of the O_3 concentration in the NH
- The increase in OH production is mainly use to destroy CO
- Because the lack of OH leads to a reduced HCHO chemical production and concentration

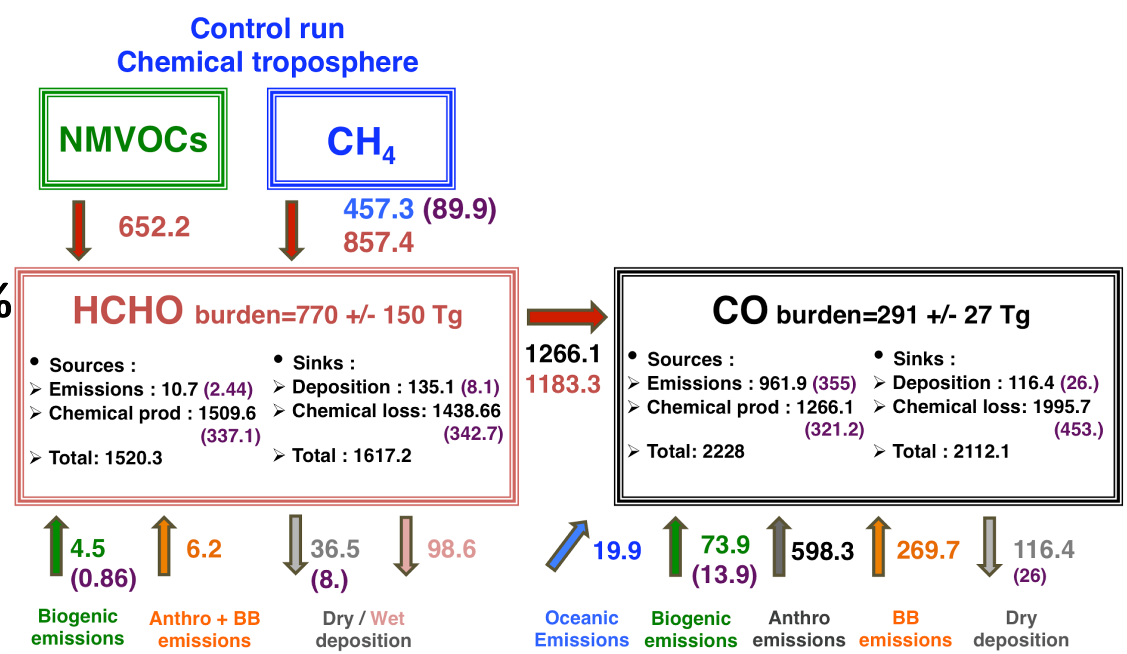




Increasing CO reduces CO secondary production !



- Note: the ratio of CO formation per CH_4 destroyed is 0.75 (supposed to be one since Duncan et al. 2007)
- The high OH and shorter CH_4 lifetime could have formed 15 % more CO relatively.
- This seems to be due to wet deposition of HCHO and other oxygenated intermediates



Conclusion and remarks

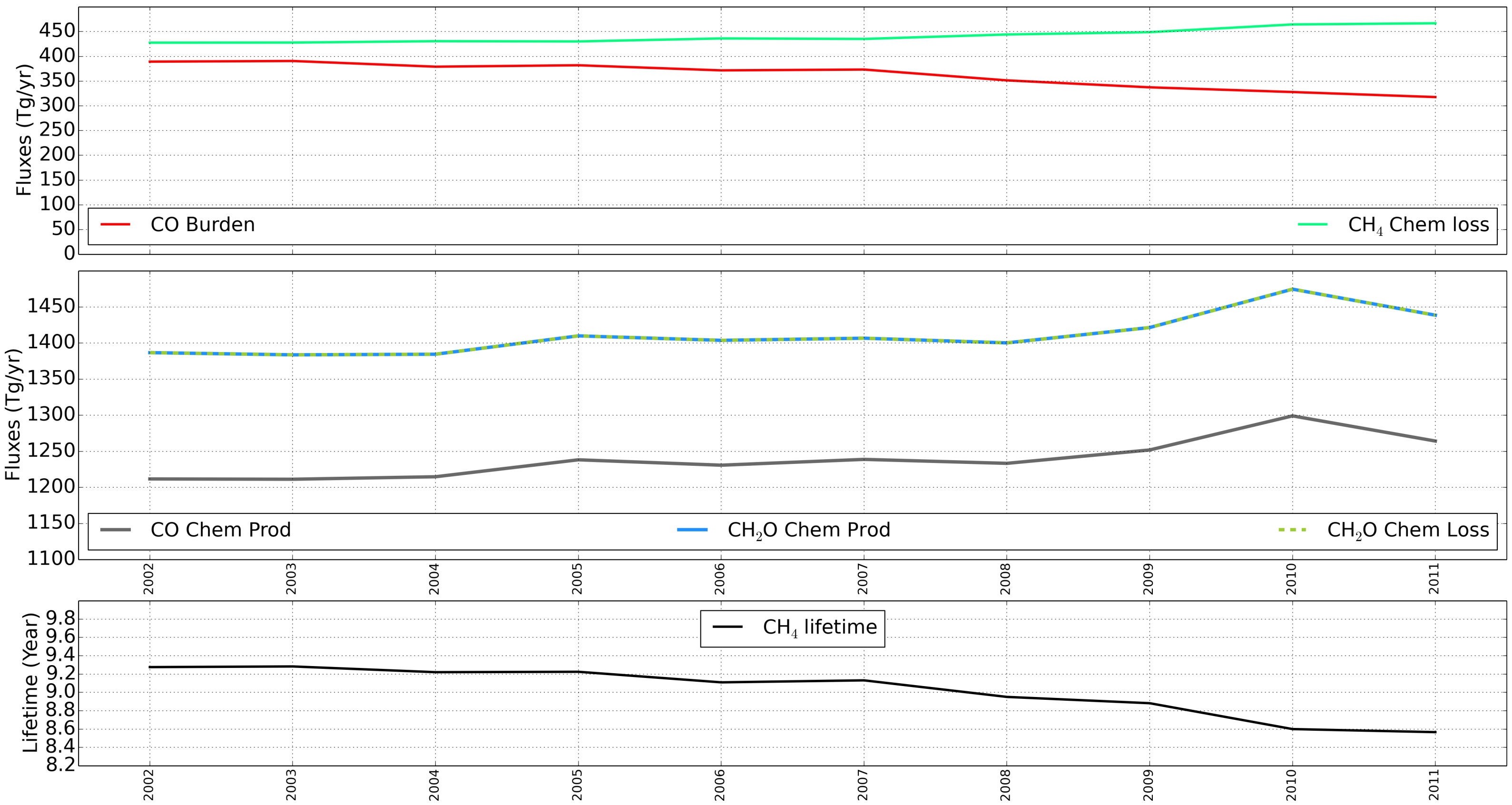
- **Lack of CO the budget, increase in CO burden for 2002/2003 from 291 Tg to 371 Tg (corrected from the SH bias)**
  - ✓ Verified by different independent datasets
  - ✓ Underestimation of primary CO emissions
  - ✓ Hard to distinguish with an OH overestimation

- **Non linearities from the full (and ensemble) response of the chemical system due to the chemical competition**
  - ✓ Increase in CH_4 lifetime
  - ✓ An Increase in CO by assimilation reduces the CO chemical production,
  - ✓ Loss through wet deposition
  - ✓ Use higher spatial resolution would transition to lower Nox regime and would reduce Ozone and subsequent OH recycling (e.g. Yan et al. 2014, 2016)
  - ✓ Budget and tags suggest a lack of secondary sources from VOC's oxidation, could possibly be done by correcting VOC thanks to the ensemble correlations

Gaubert et al., JGR 2016

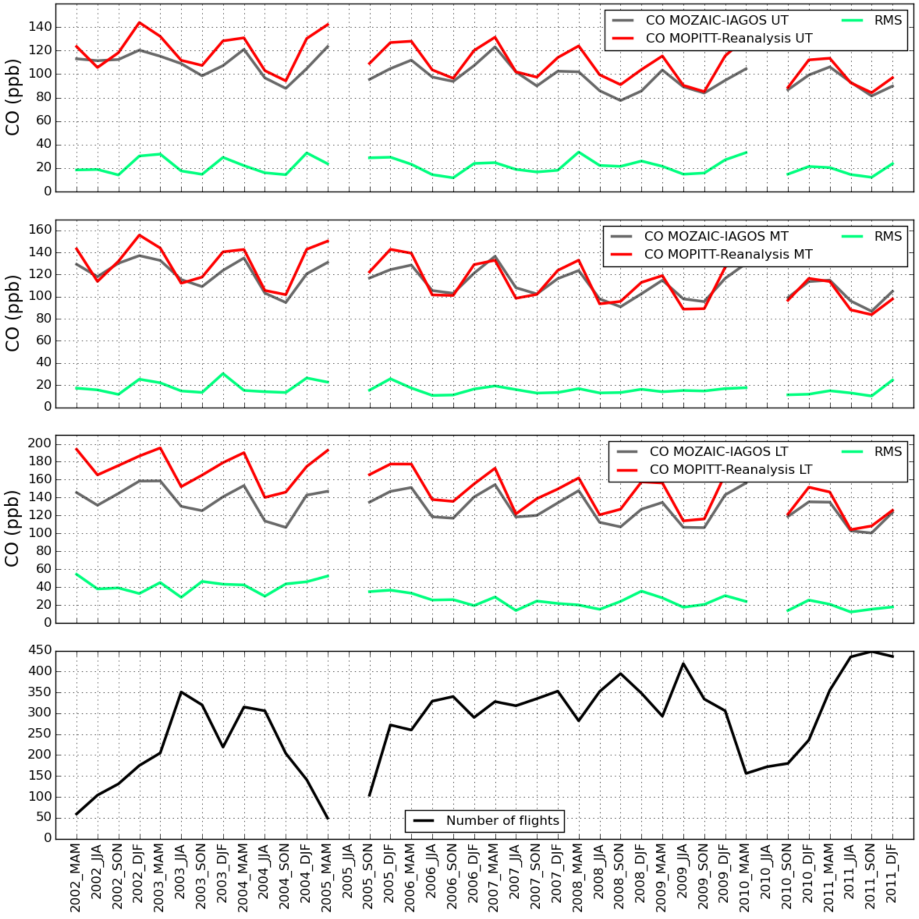
10 years of MOPITT-Reanalysis

- Decrease in CO by less than 100 Tg
- Increase in CH_4 enhances chemical CO production
- It is possible because the CO atmospheric burden decrease
- OH is well buffered and NOx increase in East Asia (Duncan et al. 2016)

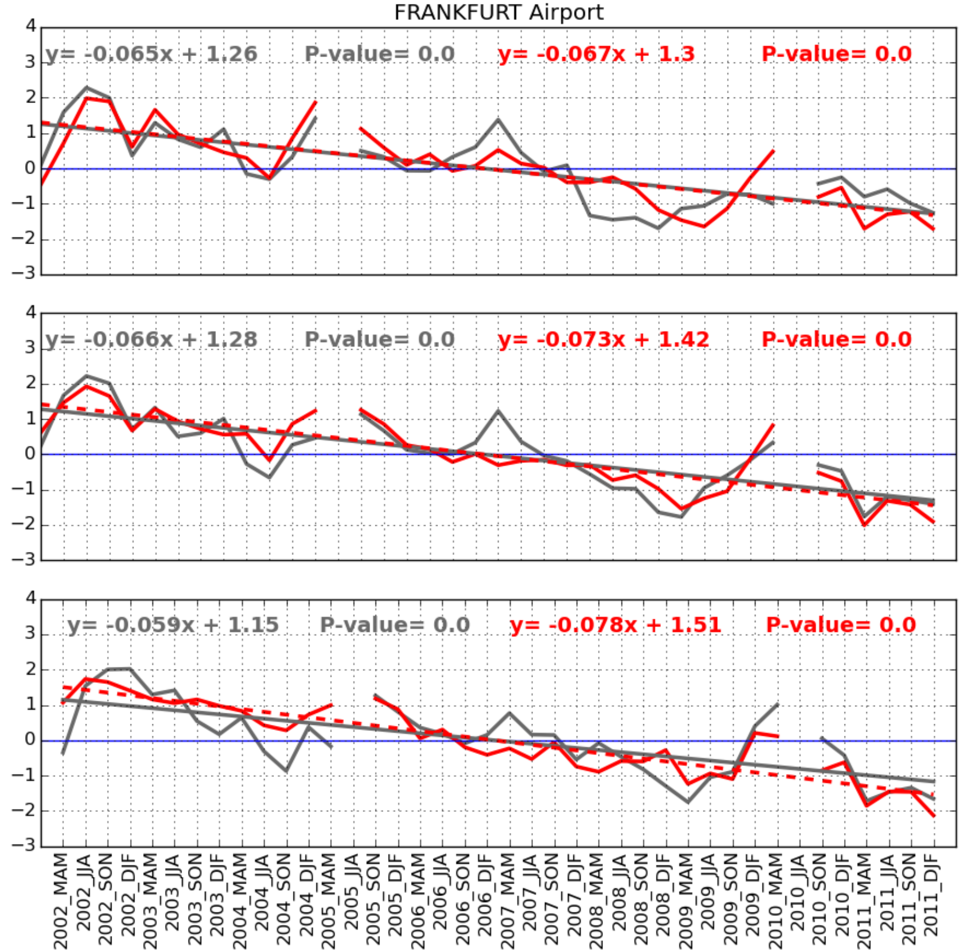


Trends analysis: MOZAIC-IAGOS data

- **Frankfurt largest database datasets (Petetin et al. 2015)**
- **LT between 1 and 4 km, MT: 4 to 7, HT: 7 to 11 km height, Seasonal average**
- **Similar slopes and P-values**



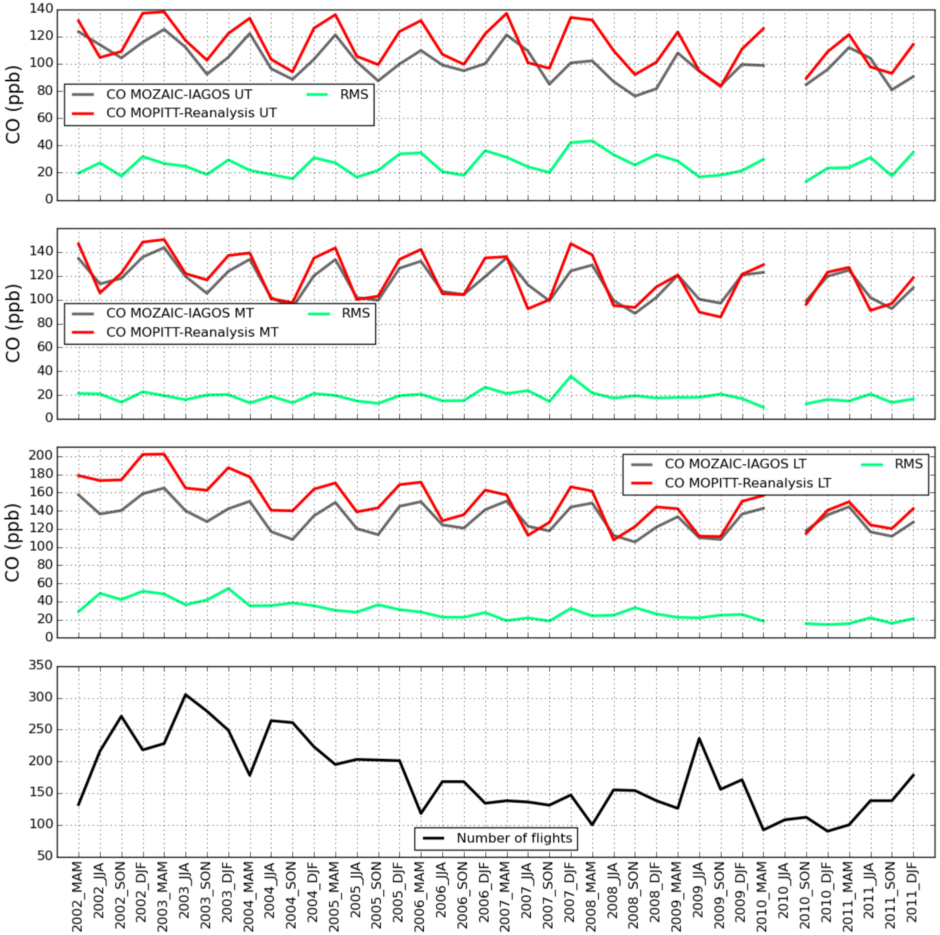
Anomalies



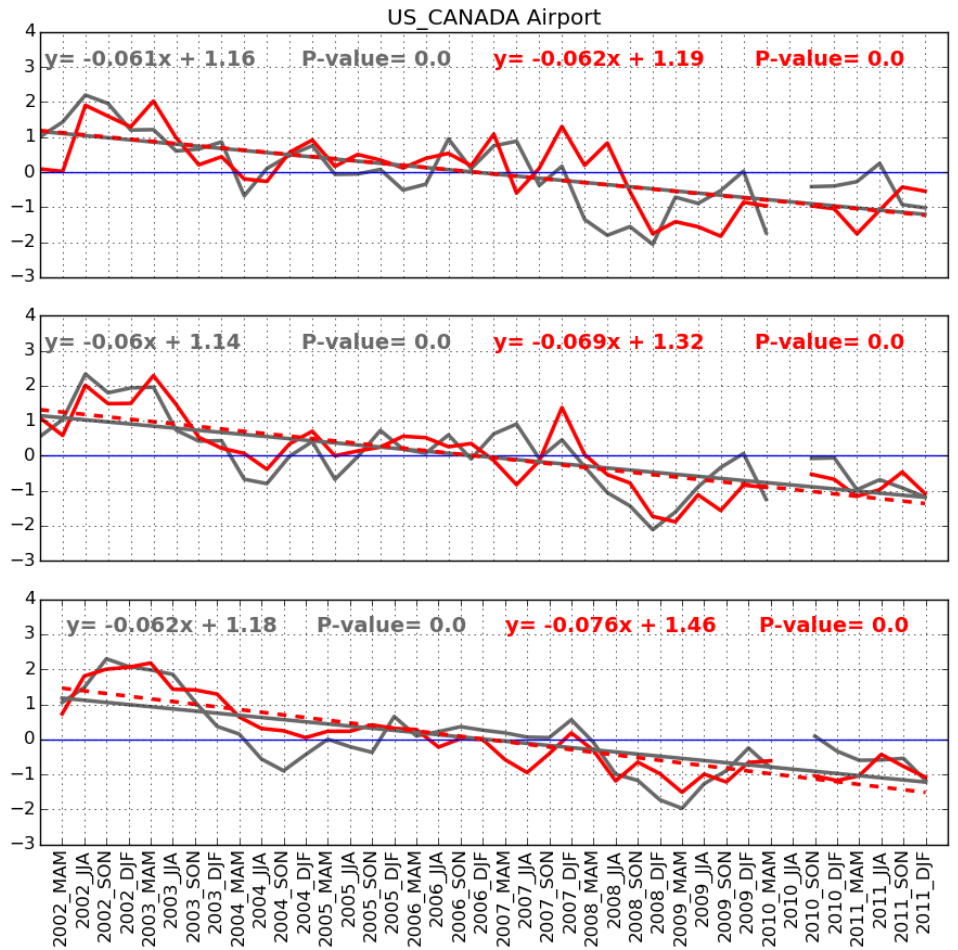
Trends analysis: MOZAIC-IAGOS data

- All airports US CONUS and Canada
- Similar slopes and P-values

CO values



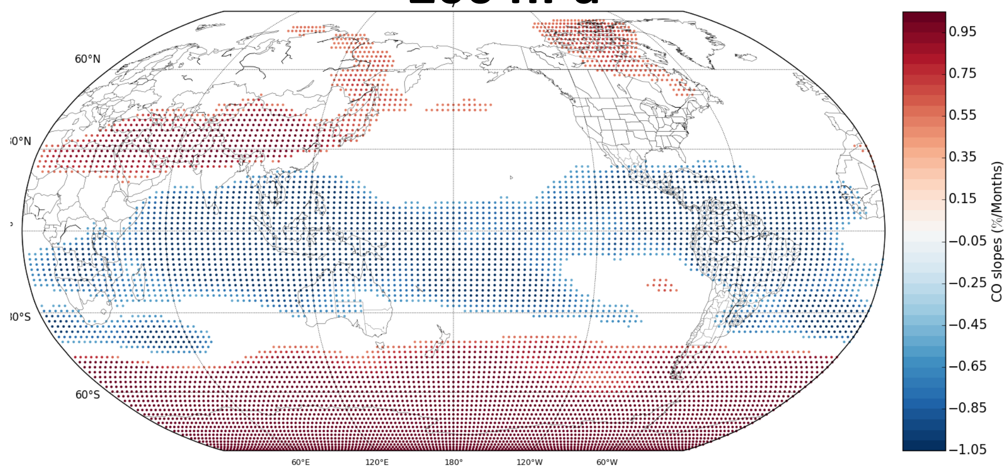
Anomalies

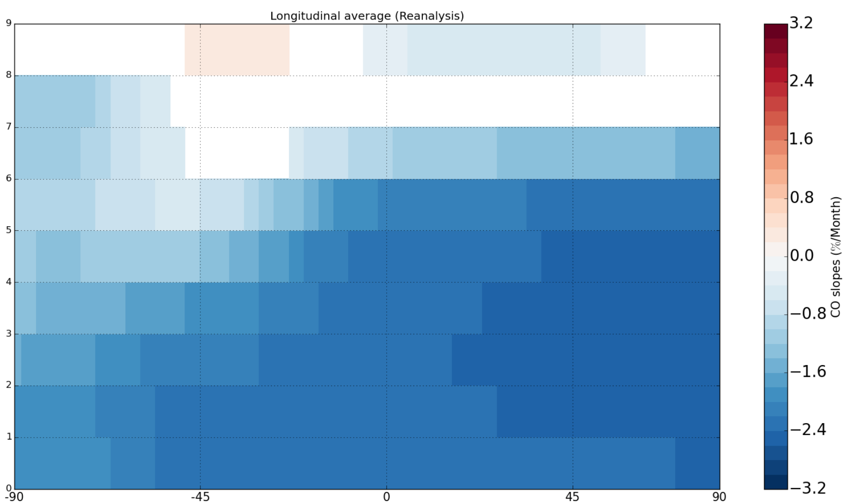


Trends analysis: Globe

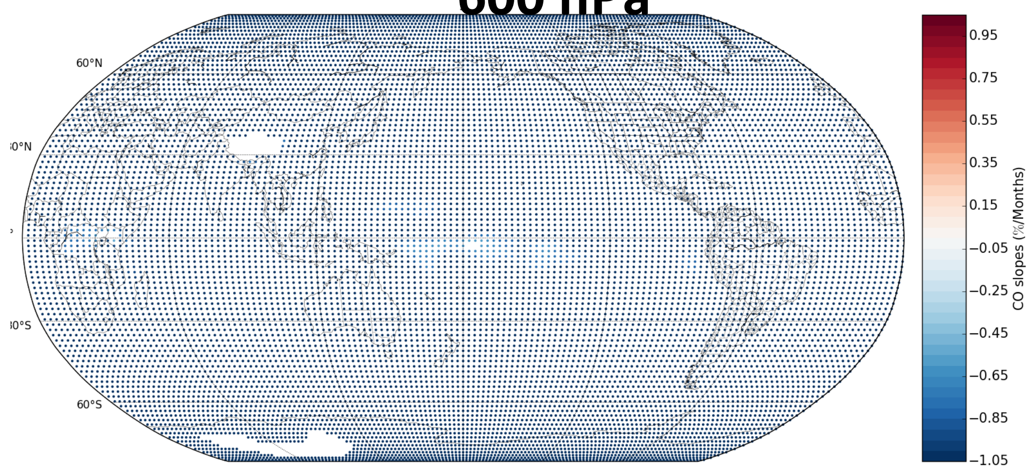
- Negative trends, everywhere but SH tropical upper troposphere
- Plots show slope when P-Value < 0.05
- Needs more uncertainty analysis
- Ensemble and MERRA run

200 hPa





600 hPa



Conclusions

- ➢ **DART-CAM-Chem system efficiently assimilate a decade of MOPITT observations**
  - ✓ **The observation operator using relocation and localization (introduced here) according to the MOPITT averaging Kernel may not be optimal, but allow extracting most of the vertical sensitivity**
- ➢ **First results showing increasing trends in the upper troposphere**
  - ✓ **Because other system were assimilating the total columns or single retrieval**
  - ✓ **Possible because we are using a full chemical mechanism**
- ➢ **Decreasing CH_4 lifetime over the decade**
  - ✓ **Shown by CH_4 modelers as well**

Barré, J., B. Gaubert, A. F. J. Arellano, H. M. Worden, D. P. Edwards, M. N. Deeter, J. L. Anderson, K. Raeder, N. Collins, S. Tilmes, et al. (2015), Assessing the impacts of assimilating IASI and MOPITT CO retrievals using CESM-CAM-chem and DART, J. Geophys. Res. Atmos., 120, 10,501–10,529, doi:10.1002/2015JD023467.
Gaubert, B., et al. (2016), Toward a chemical reanalysis in a coupled chemistry-climate model: An evaluation of MOPITT CO assimilation and its impact on tropospheric composition, J. Geophys. Res. Atmos., 121, doi:10.1002/2016JD024863.

Acknowledgments:

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- **NCAR-CISL** for computational ressources
- **DART team**
- **NASA**
- **NOAA/NDACC**
- **MOPITT data are available at:** https://www2.acd.ucar.edu/mopitt