Analysis of the impact of biomass burning emissions on global ozone production in the upper troposphere with MOCAGE CTM and IAGOS airborne data.

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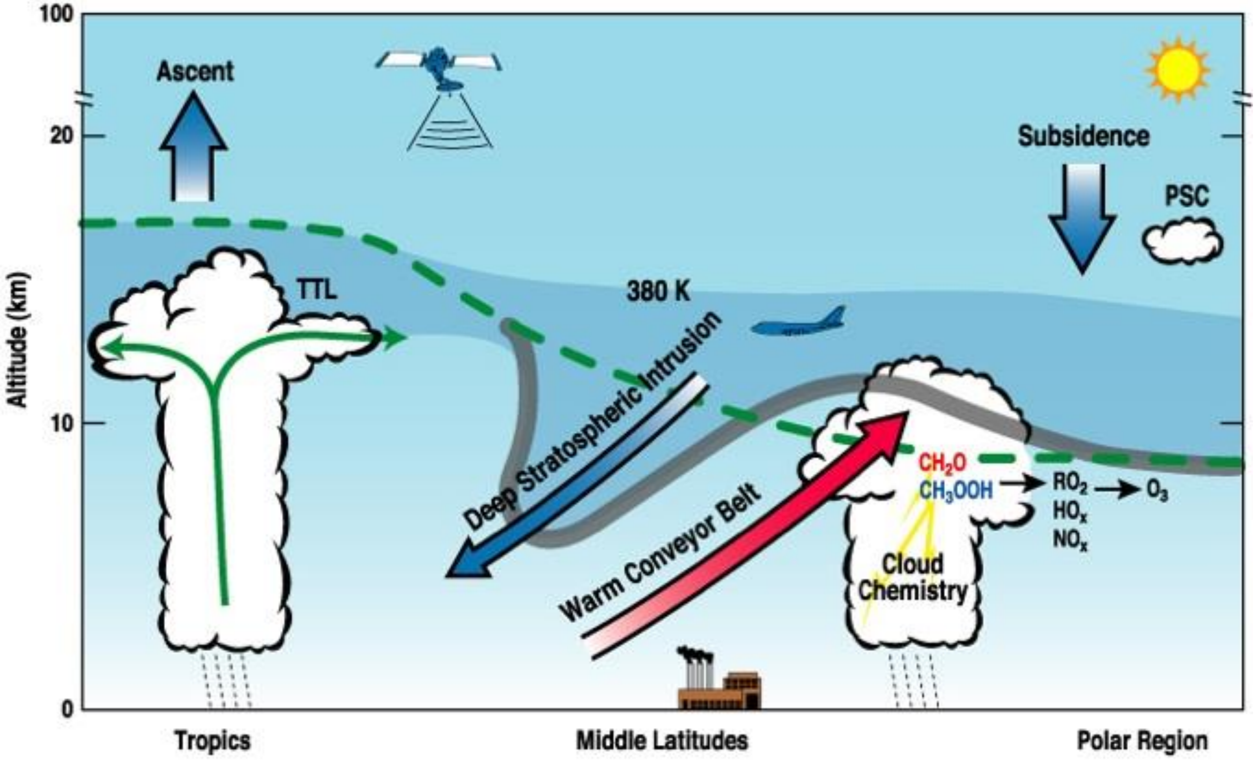
Introduction & Context



The Upper Troposphere/Lower-Stratosphere

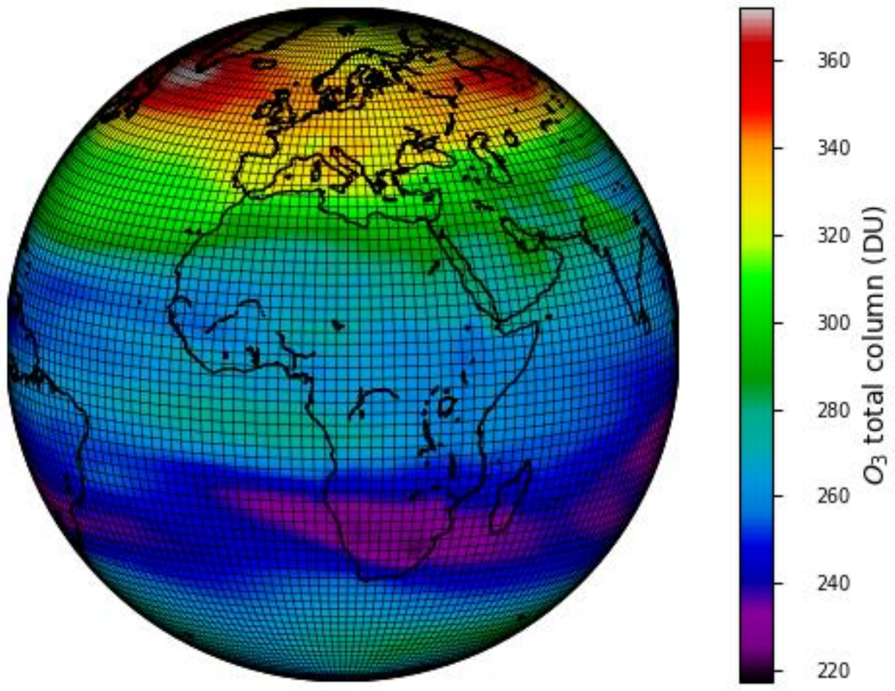
Interface between the two first layers of the atmosphere, subject to many physical and chemical processes. Biomass burning for instance is an important source of ozone precursors in the upper troposphere.

Key species :

- **O_3** : Radiative impact
- **CO, NO_x** : O_3 production

Objective : Study characteristics of the chemical composition of the Upper Troposphere. Focus on Biomass Burning emissions, associated processes and their impacts.

Modelling tool : MOCAGE Chemistry-Transport Model



- 47 σ-hybrid vertical levels from surface to 5hPa
- 8 levels in the UTLS / 800m vertical resolution
- Chemical schemes :
  →RACM for the troposphere
  →REPROBUS for the stratosphere

Reference simulation over 2013 :

- 1°.1° Global resolution
- Hourly meteorological forcing from ARPEGE
- Emission inventories :
  →Anthropogenic : MaccCity
  →Biomass Burning : daily GFAS v1.2
  (including injection height)

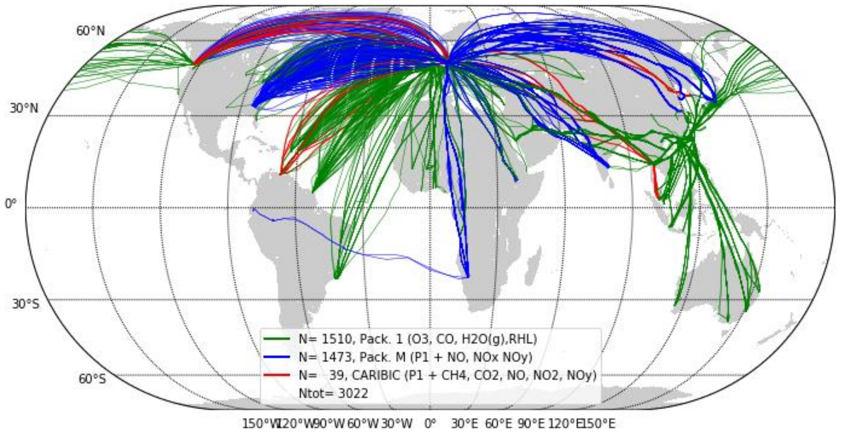
Measurements : IAGOS airborne data

In service Aircraft for a Global Observing System

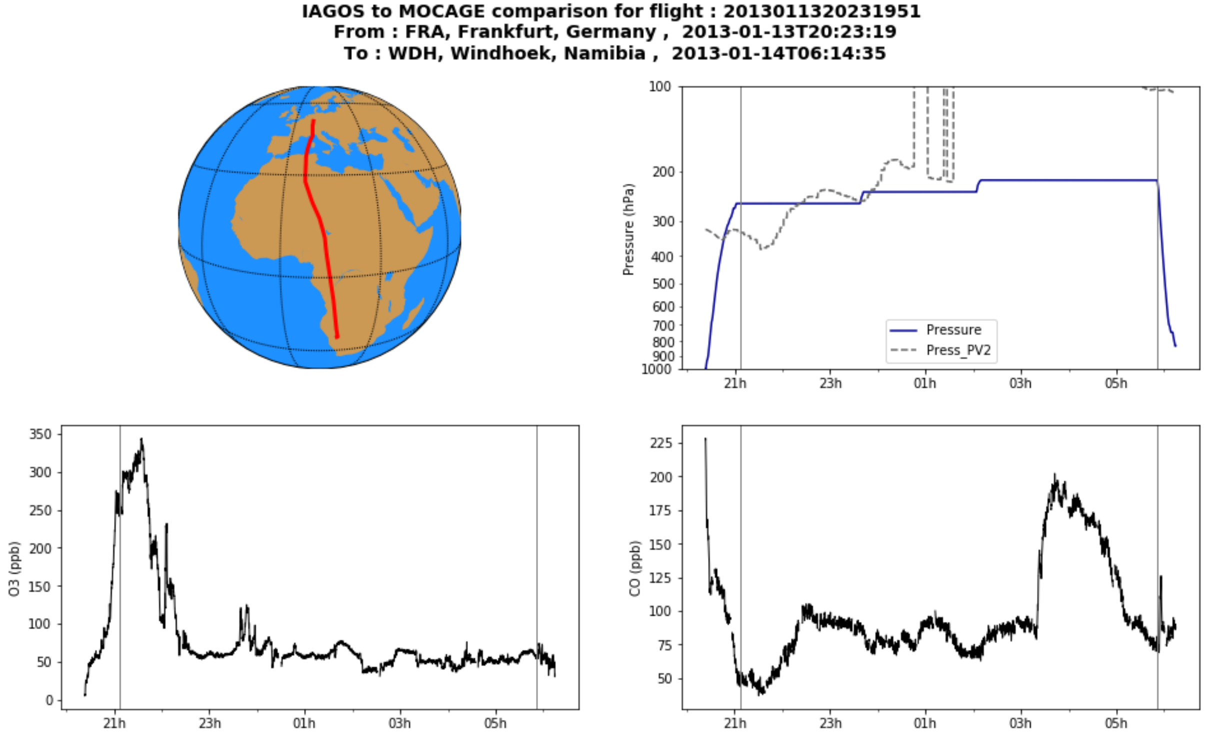
(http://www.iagos.org)

Commercial airplanes equipped to measure air state and chemical composition.

+ Good vertical resolution in UTLS
+ Quasi-global spatial coverage.
+ Good temporal coverage (6 to 10 flights/day)



2013 flight tracks



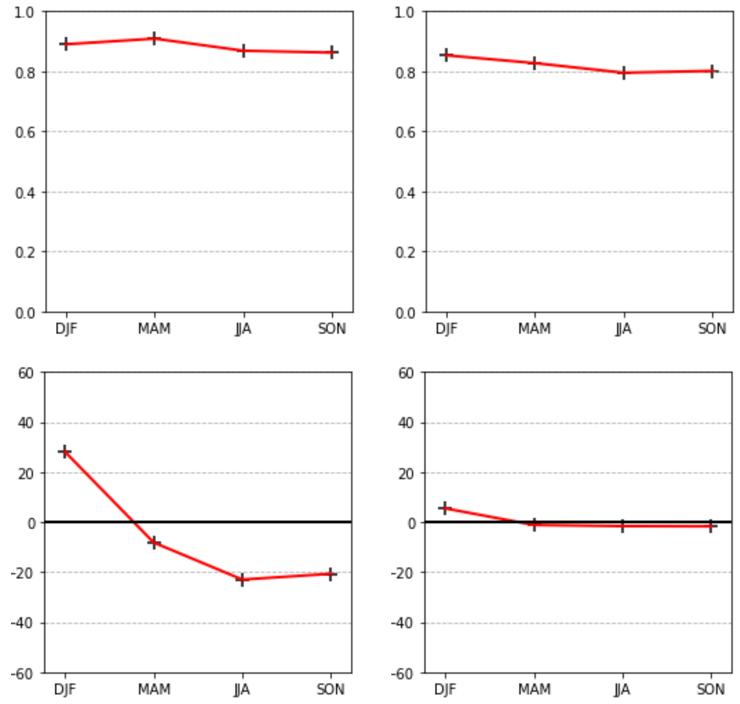
General evaluation in the UTLS

Simulation results are interpolated to match measurements time and location. Resulting dataset is analysed :
- Measurements at cruise altitude for UTLS validation
- Ascent and descent phase for vertical structure of troposphere

O_3 | **CO**

Correlation
all flights

Bias (ppb)
all flights

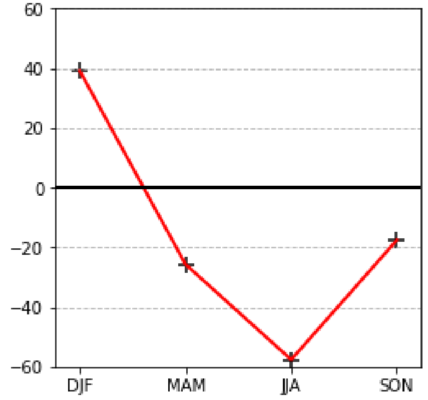


Very good correlation for both O_3 and CO

- CO bias is stable and low

- O_3 bias varies more with seasons
=> Summer O_3 underestimation in the mid-latitudes

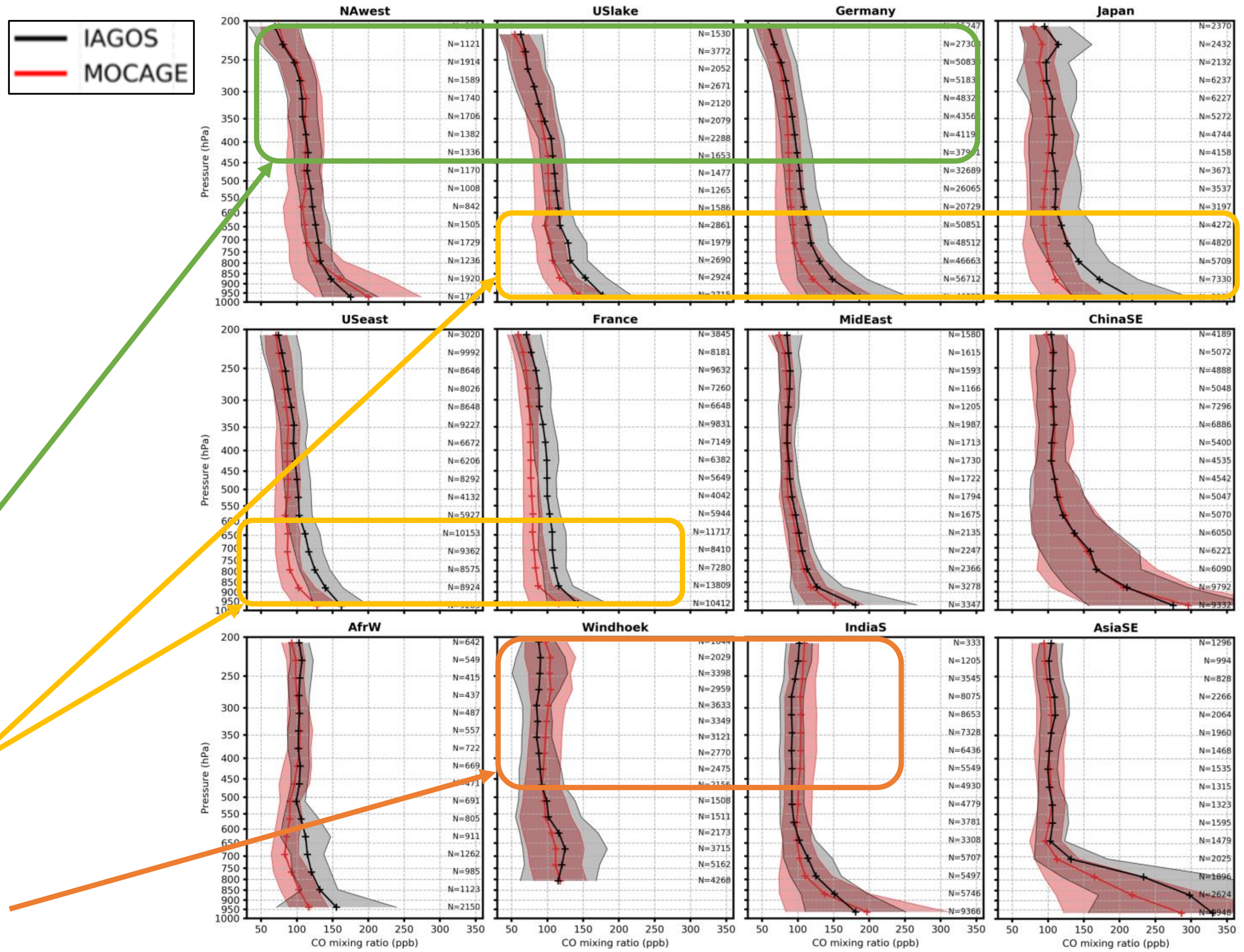
O_3 bias (mid-latitudes flights only)



Overall MOCAGE is able to well reproduce the UTLS in comparison to IAGOS.

CO vertical profiles at airport clusters

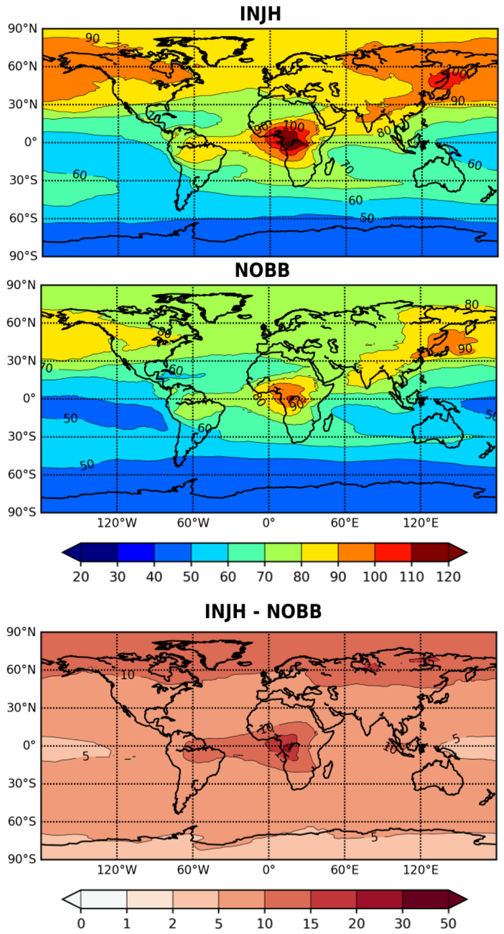
Cluster's name	Airports	N. profiles
NAwest	Seattle, Vancouver	215
USlake	Chicago, Detroit, Toronto	89
USeast	New York City, Boston, Washington D.C., Philadelphia	265
France	Paris (CDG and ORY)	318
Germany	Frankfort, Düsseldorf, Munich	1567
MidEast	Dubai, Abou Dabi, Muscat, Riyadh, Kuwait	158
Windhoek	Windhoek (Namibia)	215
ChinaSE	Taipei, Hong Kong, Xiamen	259
Japan	Narita, Nagoya, Osaka	196
AsiaSE	Bangkok, Ho Chi Minh	199
IndiaS	Chennai, Bombay, Hyderabad	210
AfrW	Niamey, Ouagadougou, Bamako	65
NSAm	Cayenne, Caracas	151



- CO vertical gradient in the UT is well represented when captured by IAGOS planes
- Overall good agreement for CO mixing ratios in the free and upper troposphere.
- Some clusters show a lack of CO in the PBL, maybe linked to underestimated anthropogenic sources.
- Slight CO excess in the free troposphere at the only two southern hemisphere airport clusters.

Global Results : CO

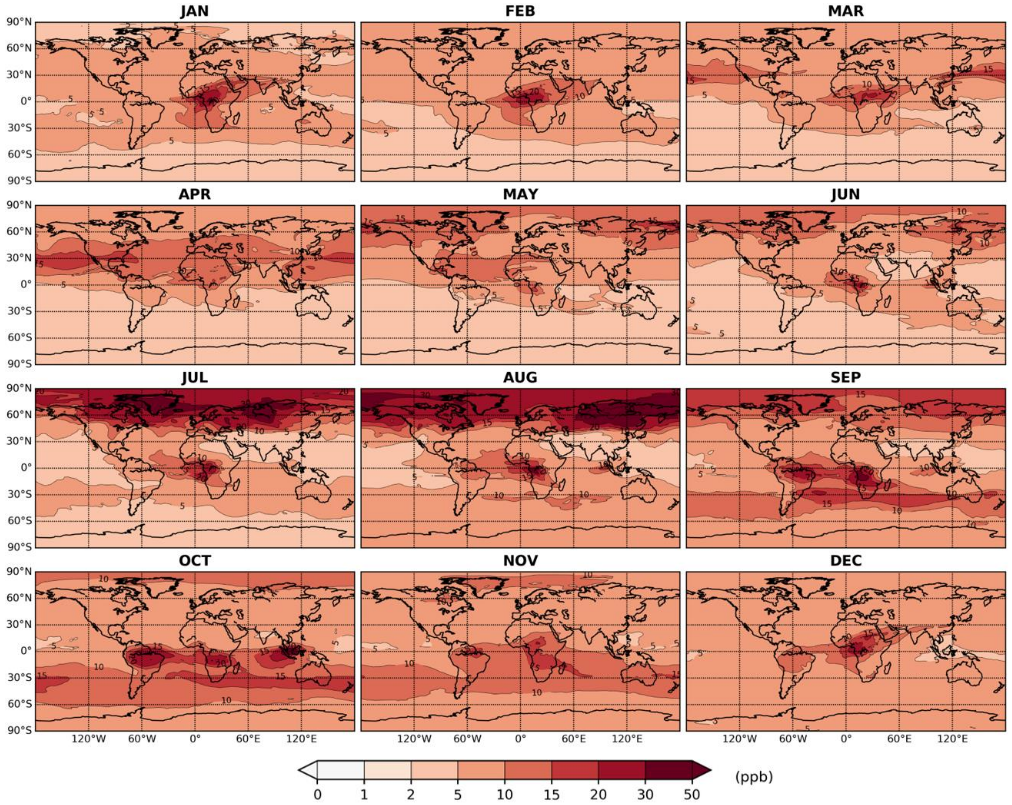
Results are averaged in an upper tropospheric layer defined with respect to the dynamical tropopause (-15 to -65 hPa below 2 pvu). Contribution from biomass burning is estimated by subtracting CO field from a reference simulation (INJH) to the one where BB emissions are turned off (NOBB).



2013 annual mean upper tropospheric CO mixing ratios

Looking at yearly results, impacts are global, with an average contribution of 8 ppb.

Impacts are stronger over equatorial Africa and boreal regions.



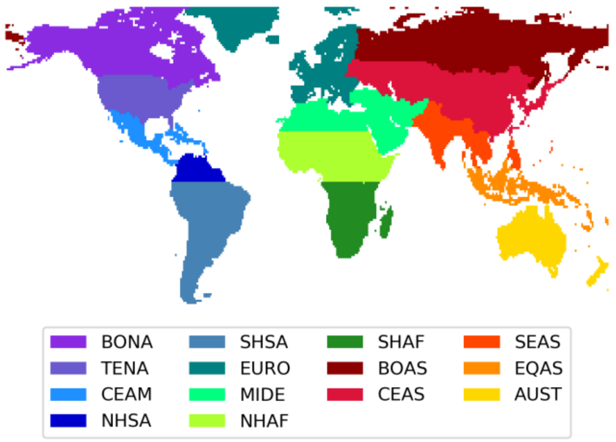
2013 monthly mean upper tropospheric CO mixing ratios

Strong seasonal impacts, following time of occurrence of fire :

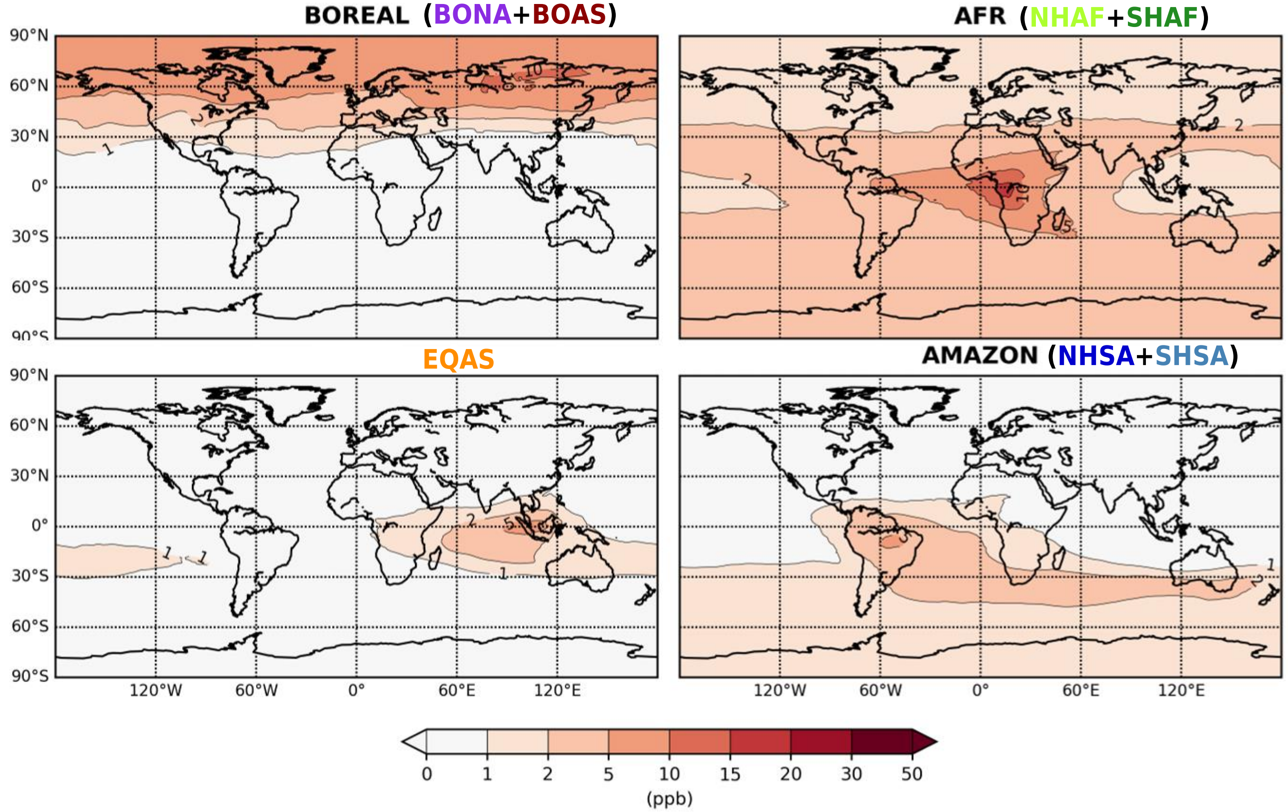
- Following ITCZ over equatorial Africa
- Summer maxima (up to 60 ppb) over boreal Asian forest in July and August

Global Results : CO

In order to evaluate contribution from different regions of the globe, sensitivity experiments are performed. Emissions are only activated in chosen regions.



- Confirmation that African fires are the biggest contributors with CO being redistributed globally.
- Boreal fires strong impact due to **pyroconvection** but limited to northern hemisphere. Southern hemisphere impacted by Amazon fires
- Strong impact over maritime continent despite lower emitted quantities : **strong convective transport** associated to monsoon.



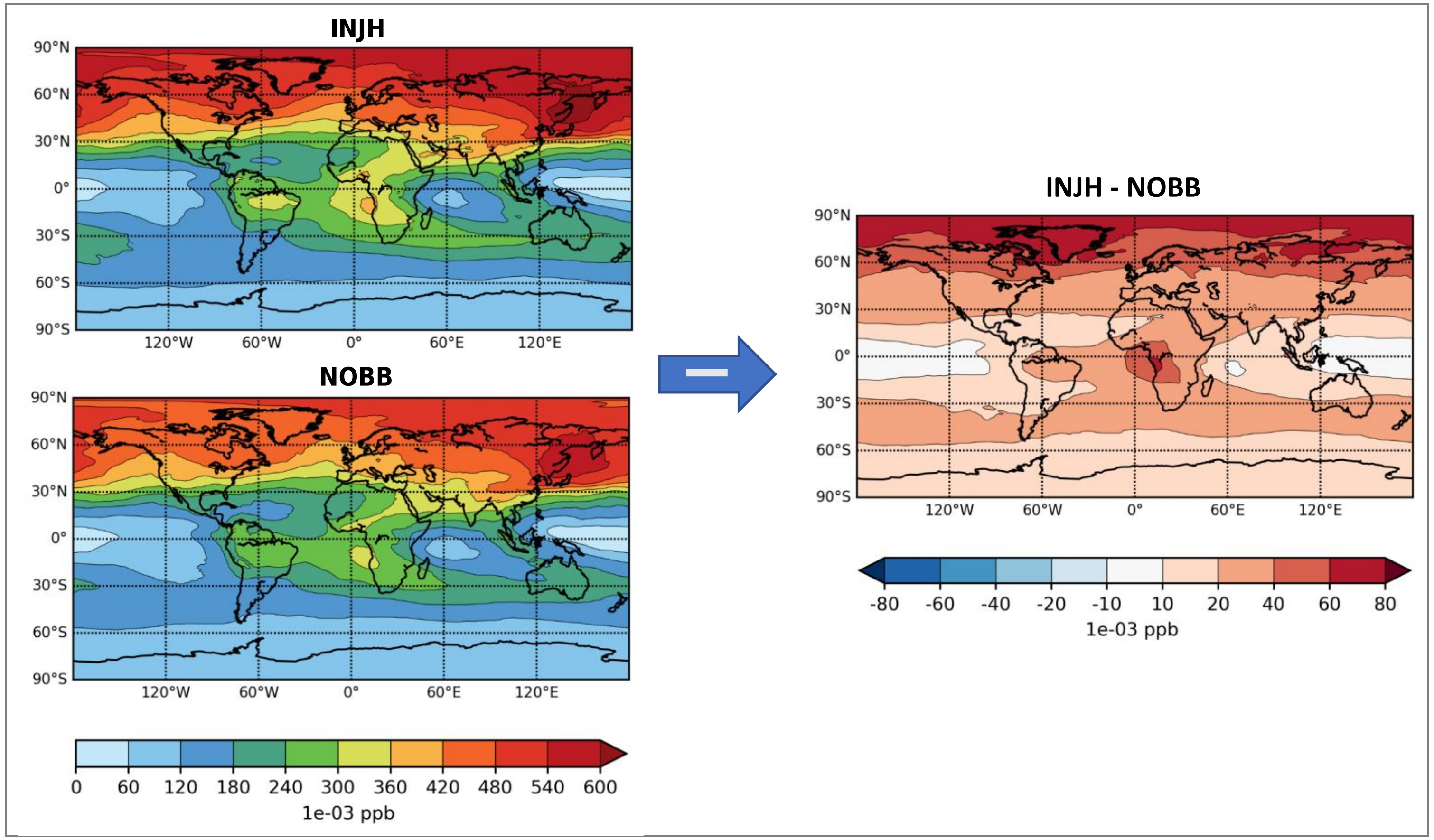
2013 annual mean upper tropospheric CO mixing ratios

Additional information can be found in the following paper :

Cussac, M., Marécal, V., Thouret, V., Josse, B., and Sauvage, B.: The impact of biomass burning on upper tropospheric carbon monoxide: A study using MOCAGE global model and IAGOS airborne data, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-1143, in review, 2020.

Impacts on O_3 production :

Biomass burnings also impacts other O_3 precursors. NO_x are too short-lived to be directly impacted, but PAN (peroxyacetyl nitrates) has a greater lifetime in the UT, and is the main reservoir species in the upper troposphere :

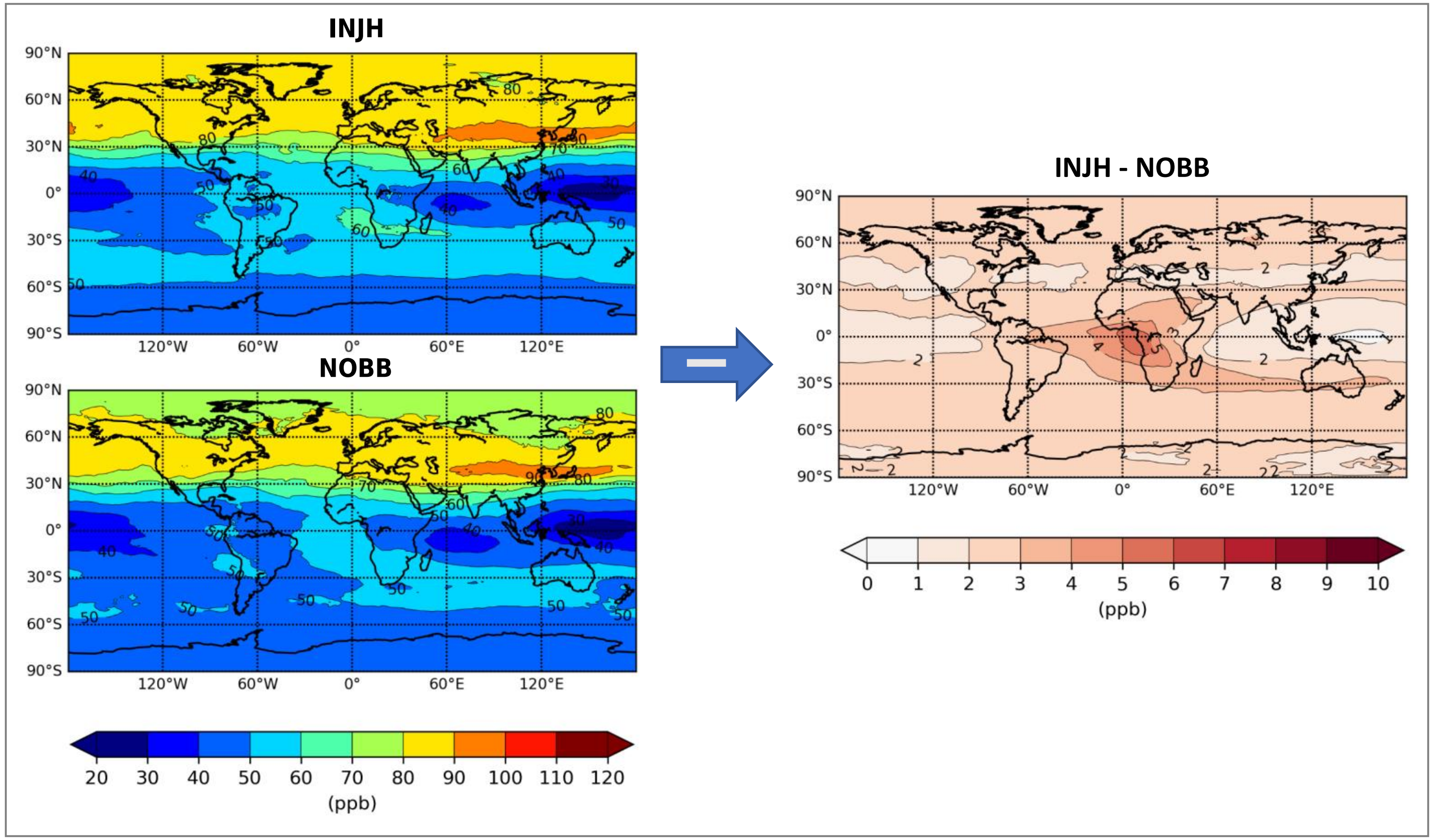


2013 annual mean upper tropospheric PAN mixing ratios

Its response is similar to the one of CO with greater impacts at boreal latitudes where its lifetime is the greatest. Globally biomass burning increases PAN mixing ratios by 10 to 15 % in the upper troposphere.

Impacts on O_3 production :

Finally, we investigate upper tropospheric O_3 and its production:

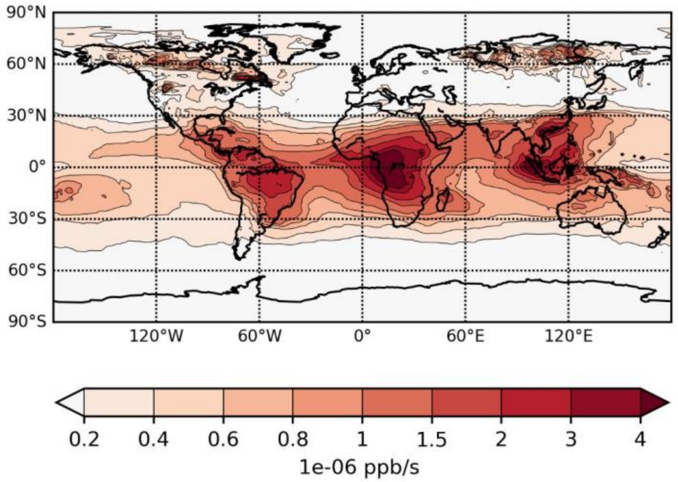


2013 annual mean upper tropospheric O_3 mixing ratios

O_3 increases mostly above equatorial Africa, from 1-5 ppb (2-10 %) :

- Maximal actinic flux all year round around the equator.
- Chemical regime isn't NO_x limited (NO_x are available from lightning).

=> Increase in O_3 chemical net production, mostly in the tropics :



Conclusions & perspectives

- Global simulations at a fine resolution were carried out to evaluate BB contribution to UT air composition :
  - → IAGOS airborne data very valuable for a global validation in the UT and LS, and showed the good performances of MOCAGE in the UT.
  - → Looking at direct impact of BB emissions on UT CO showed a global impact, with major contributions from African and Boreal fires.
  - → O_3 production is also impacted, mostly around the equator.
  - → Despite CO increase over boreal forest, O_3 production increase isn't as strong, probably due to NO_x limited chemical regimes.

- Perspectives :
  - → Complement the CO contribution with indication transport and local production from other COVs
  - → Extend the analysis on chemical regimes for O_3 production with various indicators ($HCHO/No_y$, H_2O_2/NO_z, …)
  - → Perform a similar study but on anthropogenic emissions

Thanks for your attention !

Any questions ?

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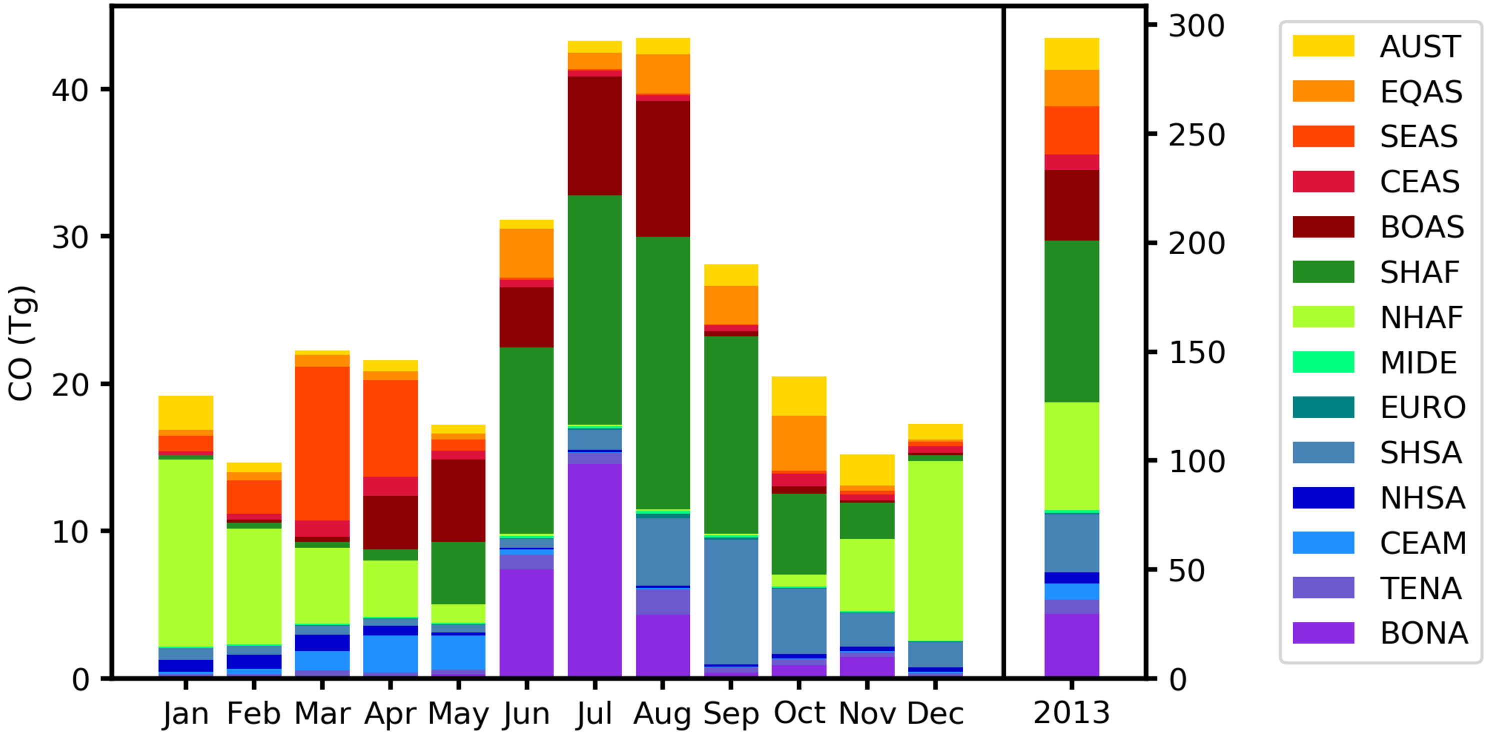
Supplementary materials



GFAS 2013 monthly and yearly emitted quantities of CO (in Tg) for the 14 geographical areas defined earlier.