The impact of biomass burning on upper tropospheric carbon monoxide : A study using MOCAGE global model and IAGOS airborne data.

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Abstract.

In this paper the fate of biomass burning emissions of carbon monoxide is studied with the global chemistry transport model MOCAGE and IAGOS airborne measurements for the year of 2013. The objectives are firstly to improve their representation within the model and secondly to analyse their contribution to carbon monoxide concentrations in the upper troposphere. At

5   first, a new implementation of biomass burning injection is developed for MOCAGE, using the latest products available in GFAS biomass burning inventory on plume altitude and injection height. This method is validated against IAGOS observations of CO made in fire plumes, identified thanks to the SOFT-IO source attribution data. The use of these GFAS products lead to improved MOCAGE skill to simulate fire plumes originating from boreal forests wildfires. It was also shown that this new biomass burning injection method did not change upper tropospheric carbon monoxide concentrations elsewhere on the globe

10 as the previous one was already satisfying. Then, MOCAGE performances were evaluated in general in the upper troposphere in comparison to the IAGOS database, and were shown to be very good, with very little bias and good correlations between the model and the observations. Finally, we analyse the contribution of biomass burning to upper tropospheric carbon monoxide concentrations. This was done by comparing simulations were biomass were toggled on and off in different source regions of the world to assess their individual influence. The two regions contributing the most to upper tropospheric CO were found to

15 be the boreal forests and equatorial Africa, in accordance with the quantities of CO they emit each year and the fact that they undergo fast vertical transport: deep convection in the tropics and pyroconvection at high latitudes. It was found that biomass burning contributes for more than 11 % on average on the CO concentrations in the upper troposphere, and up to 50 % at high latitudes during the wildfire season.

1 Introduction

The upper troposphere-lower stratosphere (UTLS) is an important layer of the atmosphere subject to strong gradients in both its dynamics and its chemical composition. Understanding upper tropospheric air composition is necessary to better quantify exchanges of air masses between the troposphere and the stratosphere (Gettelman et al., 2011). Ozone (O_3), mostly abundant

5 in the stratosphere, plays a great role on Earth radiative balance (Xia et al., 2018). Its impact on surface temperature for instance, has been shown to be maximum for ozone in the UTLS (Riese et al., 2012) . At this altitude, ozone can originate from stratospheric intrusion, but also be produced from tropospheric precursors such as nitrogen oxides (NO_x) and carbon monoxide (CO). Even though processes influencing UT air composition are numerous, focus is made here on CO, for the reasons detailed below.

10 Carbon monoxide is one of the primary species emitted during incomplete combustion. It is emitted from both anthropogenic and natural sources. In the atmosphere it is also a product of methane (CH_4) oxidation. Since CO is the main sink of tropospheric hydroxyl (OH) radicals, it influences the oxidizing capability of the atmosphere (Lelieveld et al., 2016). CO is also a precursor to a few greenhouse gases, such as Carbon Dioxide (CO_2) and ozone, thus influencing the global radiative budget of the Earth. In the upper troposphere, CO is the main precursor of ozone compared to other volatile organic compounds (VOCs) due to

15 their shorter lifetime (at most a few days). CO can also be a way to discriminate air from the troposphere and the stratosphere, since it is only found in very low amount above the tropopause.

Given its variety of sources, CO can be transported up to the upper troposphere by two different mechanisms : (1) When emission occurs in the PBL (Planetary Boundary Layer), usually from anthropogenic origin or from the least intense fires, CO can be transported in the free troposphere and up to the tropopause thanks to deep convection and to a lesser extent to

20 long range transport, given its 2 months lifetime. (2) For the most active biomass fires, pyroconvection can result in injecting smoke directly above the PBL, and up to 8-9 km for the most extreme cases (Labonne et al., 2007; Kahn et al., 2008). Unlike long range transport, the time scale of these injections is much shorter, and leads to the largest anomalies of CO detected in the troposphere (Petetin et al., 2018). Since this phenomenon occurrence relies heavily on the fire activity, as well as the atmospheric state above the fire, the computational cost to represent this process explicitly in a global chemistry model is high.

25 Biomass burning sources contribute to an important part of CO emitted in the troposphere, and are estimated to range between 350-600 TgCOyr^{-1} (van der Werf et al., 2006), which is comparable to CO emissions from anthropogenic sources, estimated between 450-600 TgCOyr^{-1} (Lamarque et al., 2010).

Satellite observations have greatly improved our understanding of biomass burning fluxes, resulting in the development of many fire emissions database such as GFAS (Global Fire Emissions Database), GFED (Global Fire Emissions Database) or

30 IS4FIRES (Integrated Monitoring and Modelling System for wildland fires). However, biomass burning emissions are still linked to uncertainties compared to other sources of trace gases such as anthropogenic emissions, for three reasons: (1) Their location and time of occurrence, though tied to seasons, is unpredictable. Their detection relies heavily on the frequency and coverage of satellite observations used to make estimates of fire emissions.(2) The nature and quantities of compounds emitted during a fire depend on the nature of the fuel, which can be inferred from a land cover class map. (3) Their intensity can vary as

well, and depending on the intensity of the fire as well as the atmospheric conditions above the fire, pyroconvection can occur. This phenomenon can cause the plume to rise above the PBL, and therefore have an important impact on plume transport. In this study the sensibility to the injection of CO from biomass burning in the troposphere is investigated, through global modelling and in-situ measurements.

5   Different types of measurements are available to study upper tropospheric carbon monoxide. Satellites (IASI for example) provide global measurements, but have been shown to lack vertical resolution in the UTLS, where gradients of CO concentrations are strong (Deeter et al., 2013). Vertical soundings can provide vertically well-resolved data, but concern mostly O3 and lack global coverage. Lastly, aircraft can also be equipped for air composition measurements. On one hand, airborne campaigns provide high quality measurements on a regional scale, but limited in time. On the other hand, the IAGOS infrastructure

10   (www.iagos.org, Petzold et al. 2015) equips commercial air planes with air composition sensors, in order to provide regular sampling over several decades. This dataset has been chosen for this study since it offers a large number of UTLS multi-species measurements (including CO and O3), on an almost global scale since 1994 for O_3 and 2001 for CO.

A challenge to tackle when using the IAGOS dataset is to be able to discriminate the most likely source of the sampled air masses. As explained earlier, CO sources are multiple, and pathways through the troposphere can be various. This is

15   why the analysis of such a large dataset requires other kinds of data, like source appointment, usually obtainable through a Lagrangian backward transport calculation. SOFT-IO (Sauvage et al., 2017) is a recently developed tool coupling backward transport calculation and emission inventories to discriminate sources of CO anomalies encountered by the aircraft. Results from this model are available as added value products for the whole IAGOS dataset. As this study focuses primarily on biomass burning and CO, data from SOFT-IO are used for source attribution and identification of biomass burning plumes. Identified

20   fire plumes in the IAGOS database are then used to assess the ability of global numerical simulations performed by MOCAGE Chemistry-Transport Model (CTM) to model these plumes.

Regarding the representation of biomass burning within a CTM, previous studies have shown the importance of taking into account the variability in the altitude at which emissions are injected (Turquety et al., 2007; Leung et al., 2007). Recent developments have been made in fire emissions inventories such as the Global Fire Emissions Database (GFAS). They now

25   include plume rise parameters, like the injection height or the top of observed plumes. These plume rise parameters are obtained through satellite measurement of the Fire Radiative Power (FRP) and a plume rise model. These parameters have very recently been used in two different global chemistry models. In the C-IFS model, plume rise parameters from the GFAS inventories have been implemented on a daily resolution and validated against aerosol extinction coefficient measurements during two different airborne campaigns (Rémy et al., 2017). Another approach was tested in the GEOS-CHEM model (Zhu et al., 2018). Monthly

30   vertical profiles of emissions were computed using a plume rise model as well as satellite data, and used for biomass burning emissions over the United States of America. The results were validated for various trace gases, including CO and PAN. These two studies show an improvement of modelled atmospheric concentrations of species emitted by fires, and encourage efforts to develop the use of such derived fire products for biomass burning emissions, in particular information on vertical distribution. In this paper is presented an implementation of the latest GFAS daily products regarding plume rise in the MOCAGE CTM. A

35   systematic global validation of biomass burning induced plumes is performed over the year of 2013 thanks to IAGOS airborne

data, and SOFT-IO information to select CO anomalies originating from biomass burning. Additionally, the results are used
to better understand biomass burning impacts on upper tropospheric CO budget, and relevant process for CO injection at this
altitude.

Our objective is to study the impact of biomass burning emissions on upper tropospheric air composition, with a focus
5 on CO exclusively. This work is based on both the IAGOS database and MOCAGE global CTM, presented respectively in
section 2.1.1 and section 2.2.1. In section 2.1.2 are presented the SOFT-IO products, and how they are used in this study to
automatically identify biomass burning plumes within the IAGOS dataset. The role of injection height during biomass burning
events is investigated in section 3 based on the latest plume rise parameters (both plume top altitude and injection height), taken
from the biomass burning inventories GFAS. In section 4.1, MOCAGE results are evaluated in the upper troposphere against
10 IAGOS observations for the reference year. Finally, the impact of biomass burning at a global scale is analysed in section 4.2
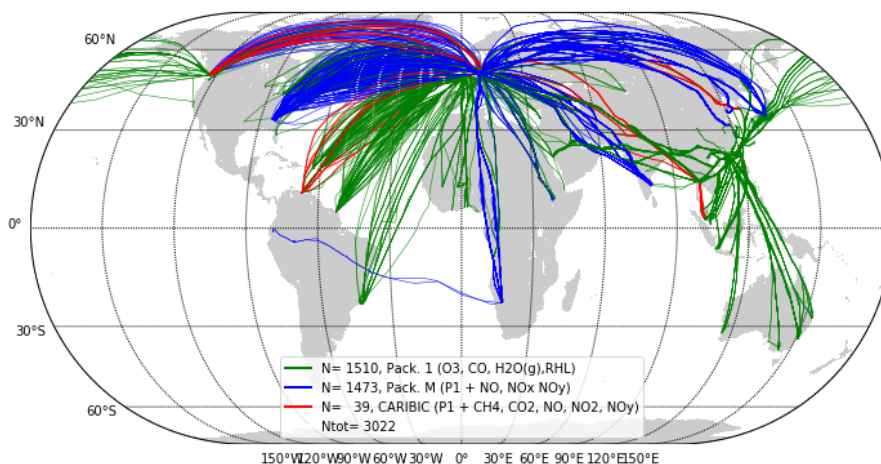by comparing different MOCAGE simulations.

2 Methodology

2.1 Data description

2.1.1 In-situ measurements : the IAGOS database

15 IAGOS (In service Aircraft for a Global Observation System) is a European research infrastructure providing a dense database
of in-situ air composition measurements, using medium to long-haul commercial flights. Initially called the MOZAIC program
(Marenco et al., 1998), it started in 1994 with aircraft equipped for ozone (Thouret et al., 1998) and water vapour measurements
(Helten et al., 1999). Carbon monoxide measurements begun later in Dec. 2001 (Nédélec et al., 2003). These air composition
measurements are complemented with meteorological measurements such as wind direction and speed, and air temperature.

20 The IAGOS program (www.iagos.org, Petzold et al. 2015) started in 2011 with the same objectives as the MOZAIC program,
and the same measurements are performed automatically on board of the equipped aircraft, during the ascent, descent and at
cruise altitude. The resulting joint IAGOS-MOZAIC database will be called hereafter the IAGOS database. To complement
existing features, greenhouse gases (CO_2 and CH_4, Filges et al. (2018)) and aerosols measurements (Bundke et al., 2015) are to
be instrumented as well in the next few years. On average, 7 to 15 aircraft have been instrumented at the same time to perform
25 daily flights, and can result in more than 3000 instrumented IAGOS flights per year. Due to the density of the dataset and its
timespan, it has been used for different kinds of studies : climatology and trends (e.g. Cohen et al., 2018; Gaudel et al., 2018),
chemistry and physics processes (e.g. Ding et al., 2015; Zahn et al., 2014), as well as model evaluation (e.g. Gaudel et al.,
2016; Inness et al., 2013; Duncan et al., 2007).

The data used in this study are the CO measurements. Their precision is estimated to be $\pm$ 5 % $\pm$ 5 ppb, for an integration
30 time of 30 s (Nédélec et al., 2003, 2015). A reference year had to be chosen for computational efficiency when comparing IA-
GOS measurements to model outputs. 2013 was selected as the most recent year with the most validated IAGOS measurements
at the beginning of this study, with a total of 3022 flights (shown in Fig. 1).
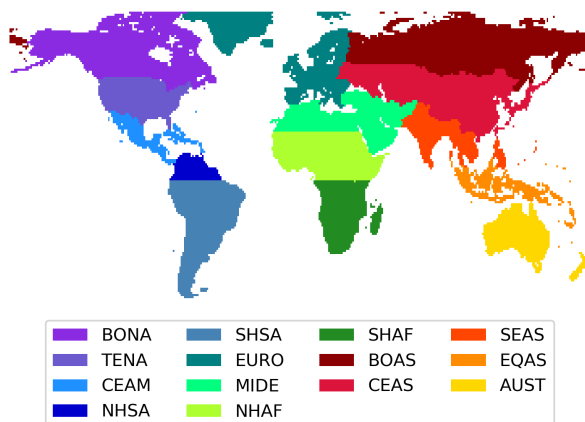
Figure 1. Trajectories of the 3022 IAGOS flights that occurred in 2013.

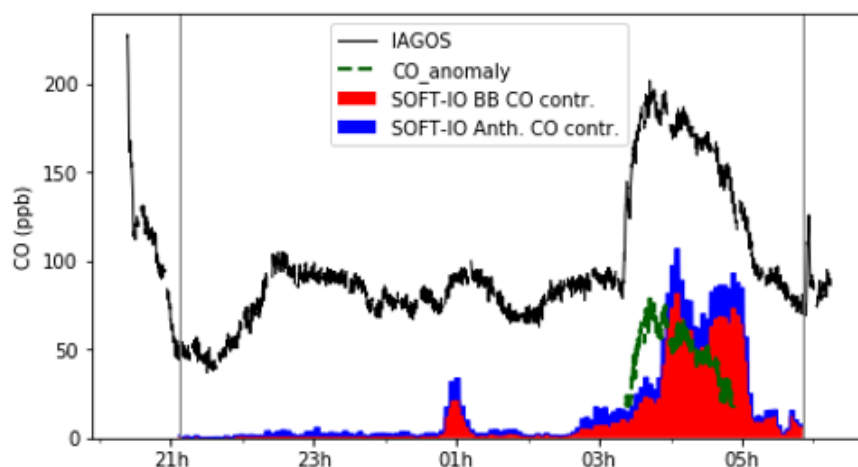### 2.1.2 SOFT-IO products and biomass burning plume identification

SOFT-IO, a tool aiming to identify the most likely source of CO anomalies encountered by the IAGOS equipped aircraft has been recently developed. The complete method description can be found in Sauvage et al. (2017). Here are summarize its main features. The FLEXPART lagrangian transport and dispersion model is ran along each aircraft trajectories at measurement

5   time and location, for a 20 days backward transport trajectories. Coupled to anthropogenic and biomass burning inventories, it results in an estimation of the contribution of recent emissions (less than 20 days) to the observed CO mixing ratios, but does not simulate CO background. Meteorological fields are taken from the operational ECWMF analysis and forecasts. The calculated CO contribution is decomposed into the contribution from the 12 zones defined in GFED (see Fig. 2). The version of SOFT-IO used in this work is the 1.0.0, where outputs are available using the MACCcity anthropogenic inventory, and for

10   both GFAS and GFED biomass burning inventories. For biomass burning, the contribution computed with GFAS emitted CO quantities and injection height are used in this study, out of consistency with MOCAGE set-up (see section 2.2.3). An example of the SOFT-IO contribution products can be seen in Fig. 3.

    SOFT-IO data are used to attribute a source for anomalies encountered by the aircraft. The method used to calculate anomalies is the same as in Sauvage et al. (2017), but only the anomalies measured at cruise altitude are considered, since we focus

15   only on the upper troposphere. CO anomalies C_{UT}^{A} are defined as the difference between the measured CO mixing ratio C_{UT} and the background CO mixing ratio, C_{UTback}. C_{UTback} is evaluated as the regional median seasonal value computed for the whole IAGOS database. Finally, only the anomalies superior to the seasonal value of the third quartile of CO mixing ratio, $C_{UTseason}(Q3)$, are considered. This can be summarized as:

$$C_{UT}^{A} = C_{UT} - C_{UTback} \quad \text{if} \quad C_{UT} > C_{UTseason}(Q3) \tag{1}$$

Figure 2. Geographical regions defined in the Global Fire Emission Database (GFED), adapted from http://www.globalfiredata.org/data.html

Figure 3. Example of SOFT-IO contributions as well as illustration of the method to calculate CO anomalies. The selected flight took off from Frankfurt (Germany) the 13th of January 2013 at 20h23 UTC, and landed the next day in Windhoek (Namibia) the next day at 6h14 UTC. The contribution is here mostly from NHAF. The two vertical grey lines represent the end of ascent and the beginning of the descent of the plane. The CO anomaly (in green) is displayed only when superior to the third quartile of anomalies.

This calculation is only performed when the plane is considered to be below the tropopause, defined here as the 2 PVU (potential vorticity unit) isosurface. The ascent and descent phases of the flight are not considered either. A detailed example of the process is presented in Fig. 3.

Based on both the anomaly calculation and the contribution calculated by SOFT-IO, a method to automatically identify biomass burning related anomalies is used. For each CO anomaly detected during a flight, if the biomass burning contribution

5

Table 1. Number of biomass burning plumes per origin, following SOFT-IO contribution calculations.

Origin	Number of plumes
AFR (NHAF/SHAF)	95 (33/62)
BOREAL (BOAS/BONA)	51 (21/30)
TENA	21
CEAM	14
SHSA	2
SEAS	8
MULTIPLE	29
TOTAL	220

from SOFT-IO is on average higher than 5 ppb and is superior to the anthropogenic contribution from SOFT-IO, the anomaly is selected as a biomass burning plume. In total, 220 plumes were sampled by IAGOS aircraft in 2013, with a majority of them originating from the African continent (either SHAF or NHAF) and boreal latitudes (either BOAS and BONA). A summary of the number of plumes by origin can be found in Table 1.

2.2 Global model

2.2.1 MOCAGE global CTM : general description

MOCAGE is the off-line global CTM (Chemistry-Transport Model), developed at Météo-France (Josse et al., 2004; Guth et al., 2016). It is used for a wide variety of research subjects concerning atmospheric air composition, at global and regional scales. It is also operated daily as part of the European ensemble forecasting system in the CAMS project (Copernicus Atmospheric Monitoring Service), with six other chemistry models (Marécal et al., 2015).

MOCAGE chemistry is based on 2 different schemes. In the troposphere, the RACM (Regional Atmospheric Chemistry Mechanism) described in Stockwell et al. (1997) is used, while the REPROBUS (REactive Process Ruling the Ozone BUdget in the Stratosphere) scheme is used in the stratosphere (Lefèvre et al., 1994). Merged together they allow for the representation of 112 gaseous species, 434 chemical reactions of which 57 are photolysis reactions.

MOCAGE has 47 sigma-hybrid vertical levels, from the surface up to 5 hPa. The vertical resolution ranges from 40 m near the surface, to 400 m in the free troposphere, and 700-800 m in the upper troposphere and the stratosphere.

Since MOCAGE is an off-line CTM, it needs external meteorological forcing (temperature, pressure, humidity, wind, cloudiness and precipitation) from a separate meteorological or climate model. Long-range transport through advection is calculated with a semi-lagrangian scheme (Williamson and Rasch, 1989). Tracers transport through convection and diffusion are computed inside the model, according respectively to Bechtold et al. (2001) and Louis (1979).

Anthropogenic, biogenic, and biomass burning emissions are taken into account using precomputed inventories (choice of inventory is detailed in Sect. 2.2.2). Both anthropogenic and biogenic emissions are injected into the 5 lowest levels of the model, with an exponential decay. Biomass burning injection scheme is detailed in 2.2.3. Nitrogen oxides (NO_x) emitted by lightning are computed according to Price et al. (1997).

5   The following processes concerning gaseous species are also represented in MOCAGE. Dry deposition is taken into account following Wesely (1989). Wet deposition is represented for both convective and stratiform precipitation, according to Mari et al. (2000) for convection, and the works of Liu et al. (2001) and Giorgi and Chameides (1986) for stratiform precipitation. Wet deposition is split into two processes, in-cloud scavenging (also known as rainout), and below-cloud scavenging (also known as washout).

10   MOCAGE has already been used to study the chemical composition or structure of the UTLS, or biomass burning plume transport. As part of the Chemistry and Climate Modelling Initiative (CCMI), it is used to study the impact of climate on tropospheric and stratospheric chemistry, as well as transport of trace gases through the troposphere (Morgenstern et al., 2017; Orbe et al., 2017). Several studies on the extra-tropical UTLS and stratospheric intrusion have also been performed in Barré et al. (2013). It was also used in the framework of the ChArMEx-GLAM airborne campaign, to study the origin of an

15   observed CO anomaly in the upper troposphere sampled over the Mediterranean basin (Brocchi et al., 2017). Indeed, such CO enhancement was a biomass burning plume originating from both Siberian and North American forest fires. In order to correctly represent the plume intercontinental transport in this study, the top of the fire emissions had to be set to an altitude of 10 km in MOCAGE to account for this extreme case of pyroconvection. Furthermore, Guth et al. (2018) highlighted the importance of the variability of biomass burning emissions of pollutants and their direct impact over the Mediterranean basin

20   aerosol budget. They showed that in 2013, important fires in North America were the main contributor to the budget of primary organic aerosols over the Mediterranean basin. Here a different study is carried out, exploring in further detail the UTLS composition and its link to biomass burning emissions.
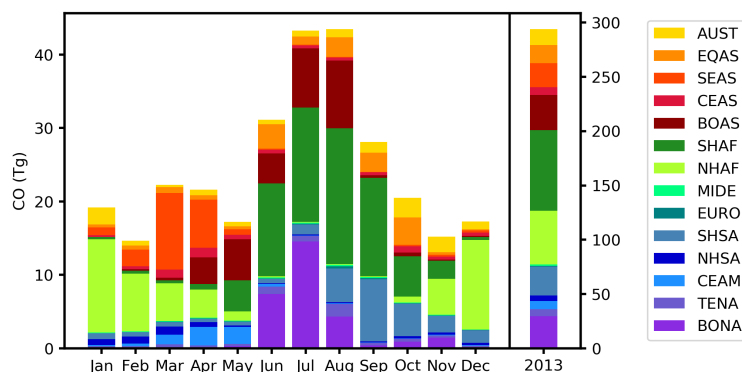
2.2.2   Simulation general set-up

MOCAGE dynamical forcing fields are taken here from meteorological analysis performed by the ARPEGE model (Action de

25   Recherche Petite Echelle Grande Echelle) operated at Météo-France (Courtier et al., 1991). They are available at a 3-hourly time-step, and linearly interpolated at a 1 h time-step, to be coherent with the dynamical time-step of MOCAGE.

For anthropogenic emissions, MACCcity inventory was used at a monthly resolution, complemented by the ACCMIP dataset for aircraft emissions (Lamarque et al., 2010). Concerning biogenic emissions, the MEGAN-MACC inventory is used (Sindelarova et al., 2014).

30   MOCAGE is used in its global configuration, with a resolution of 1°longitude $\times$ 1°latitude (approximately 110 km $\times$ 110 km at the equator and 80 km $\times$ 110 km at mid-latitudes).

The simulation covers the whole 2013 year, with an additional spin-up period of three months (starting the simulation on the 1$^{\text{rst}}$ of October 2012). Hourly outputs from MOCAGE are used, as this is the minimum coherent time step of the model.
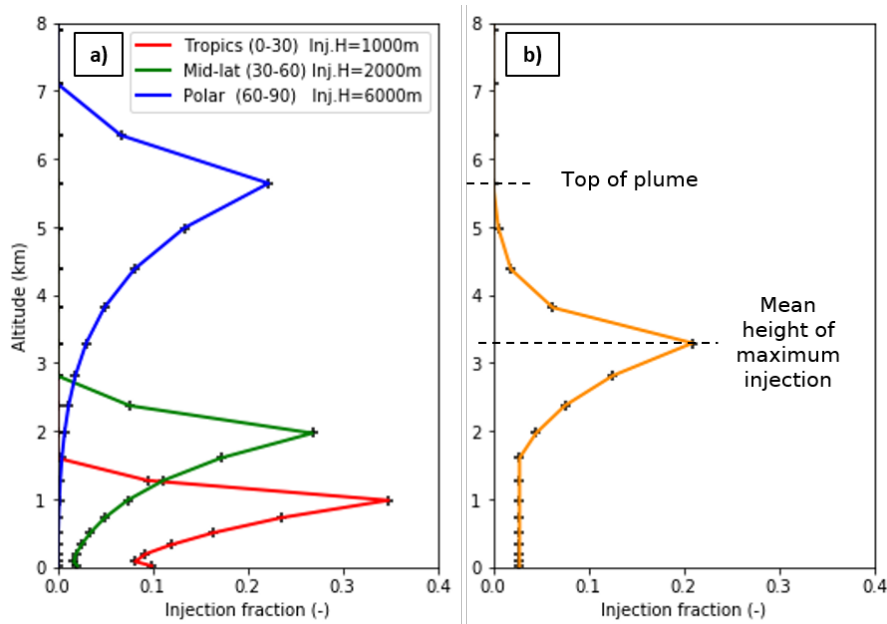
Figure 4. GFAS 2013 monthly and yearly emitted quantities of CO (in Tg) for the 14 geographical areas defined in Fig. 2.

2.2.3 Biomass burning emissions set-up

In MOCAGE, biomass burning emissions are constrained using daily GFAS products. GFAS is an inventory based on the observation of the Fire Radiative Power (FRP) made by the MODIS instrument, on board of the Aqua and Terra satellites. Based on the link made between FRP and fuel consumption, emitted quantities are calculated taking into account the land cover class at the location of the observation. Emitted CO quantities are presented in Fig. 4 for the year of 2013, where emissions have been split according to the geographical areas defined in Fig. 2. With a total of 294 Tg of CO being emitted, 2013 biomass burning yearly emissions are below GFAS overall average. In the work of Petetin et al. (2018), where GFAS CO emissions were assessed over the same continental areas from 2002 to 2017, 2013 appears to be an average year for all regions, except for SHSA being below average and BONA emissions above average.

GFAS is averaged from its native resolution (0.1°longitude × 0.1°latitude) at MOCAGE resolution (1°longitude × 1°latitude) to set the location and quantities emitted for each day. But since no information on vertical repartition of the emissions of fires was available until v1.2 released in 2017, prescribed profiles of injection were used so far (see Fig. 5). The choice was made that the injection height depends on the latitude of the fire. Injection fraction is then calculated above and below the injection height with an exponential decay. The top of the plume is set to 2 model levels above the maximum of injection. The pre-scribed injection heights are 1 km, 2 km and 6 km, respectively applied for tropical latitudes, mid-latitudes and polar latitudes. These injection profiles were developed for the AMMA model inter comparison project (Williams et al., 2008, 2009) and are derived from the work of Lavoué et al. (2000). While it is a good approximation of the potential effect of pyroconvection on fire emissions, it does not take into account the difference between the active front the fire and the smoldering areas, where pyroconvection rarely occurs. A first simulation using this representation of fire emissions is performed, reference throughout this paper as the BASE simulation.

As we want to investigate the role of the variability of injection height in the representation of biomass burning, a new way to set the injection profile in MOCAGE is developed. Instead of the prescribed injection height as a function of latitude, 2 products from GFAS v1.2 (Rémy et al., 2017) are used: the mean height of maximum injection (or injection height), and the

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Figure 5. Injection profile schemes: a) BASE injection profiles with latitude dependent injection height b) INJH injection profile using GFAS plume rise parameters.

plume top altitude. The injection height corresponds to the altitude at witch detrainment is at its maximum within the plume, and thus injection of smoke at its maximum. The plume top altitude is simply the highest altitude reached by the plume. These products are derived from the measured Fire Radiative Power (FRP) from the MODIS instrument, and a Plume Rise Model (PRM) that takes into account the atmospheric state above the fire. They are available daily for most of GFAS fire pixels.

5　These two plume rise parameters are used in MOCAGE to constrain the injection profile, as presented in Fig. 5. The injection profile is still set at its maximum at the GFAS injection height, and the GFAS plume top altitude is used instead of the fixed two model levels above the injection height. A second simulation (referred to as INJH hereafter) using this new representation of biomass burning emissions is performed, identical in every other aspect to the BASE simulation. This new representation of fire injection will be compared to the one in the BASE simulation, using observations of biomass burning plumes from the

10　IAGOS dataset, in section 3.

　　A third simulation (named NOBB) is performed where emissions from biomass burning are simply turned off (not just for CO, but all concerned species). It will be used to study biomass burning impact on CO budget in the UT, presented in section 4.2. A summary of the three simulations and their specificities can be found in Table 2.

Table 2. Simulations description summary

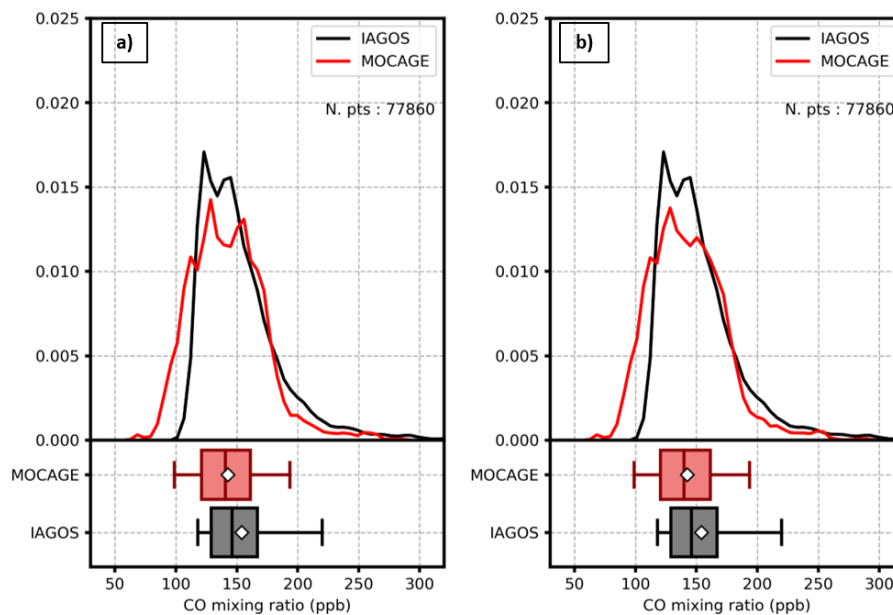| Sim. name	BB emission inventory	Injection scheme
NOBB	None	None
BASE	GFAS	Prescribed profile with zonal injection height
INJH	GFAS	Prescribed profile with injection height and top of plume from GFAS

3 Sensitivity to injection profile

3.1 Effect on biomass burning plumes

For the 220 plumes detected in the IAGOS database over 2013, CO mixing ratios measurements inside the plumes are compared with the MOCAGE model with the following methodology. Hourly results from the global model are interpolated on each plane

5 trajectory at the measurement time and location to perform a collocated comparison. The interpolation is bi-cubic in space and linear over time. This results in a consistent dataset between the IAGOS CO measurements and each MOCAGE experiment, where each data value shares the same space and time coordinates.

To perform the IAGOS-MOCAGE comparison, the probability density function (PDF) for each dataset is computed , as well as box-and-whiskers plots. The results are first investigated for the plumes originating from the African continent and boreal

10 forests, as they are the most sampled by IAGOS aircraft in 2013, and the main contributors to biomass burning emissions.

Looking at plumes originating from the African continent (either from NHAF or SHAF, see 2), both the PDF and the box plots are similar between IAGOS and MOCAGE for the two simulations. Both MOCAGE simulations well reproduce the CO mixing ratios distribution when compared to the IAGOS measurements, with a slight negative bias of -11 ppb between the means. The 95th quantile in particular is 26 ppb lower in both MOCAGE simulations compared to the IAGOS measurements.

15 Comparing both simulations against IAGOS observations, it appears that using plume rise parameters does not improve the representation of the biomass burning plumes, as no significant difference can be seen between the two datasets. Indeed, this could be expected, because pyroconvection rarely drives fire emissions above 3-4 km into the troposphere for African fires. Looking at the vertical repartition of GFAS plume rise parameters in the INJH simulation in Fig. 7, it appears that the bulk of fire emissions occurs in the BPL, with a mean altitude of injection on around 2 km. This value is not far from the injection

20 height for tropical latitudes in the BASE method of 1 km injection height. This difference is not significant because most of emissions still occurs in the PBL and plumes that reach the cruise altitude of the aircrafts (10-11 km) in the TTL are mostly transported thought deep convection. Hence, the injection profile difference on CO emissions altitudes is not noticeable at this range of altitudes.

However, looking at Fig. 8, where the same statistical comparison is performed for boreal biomass burning plumes, the PDF

25 of CO mixing ratios in the BASE simulations shows a strong underestimation when compared to the IAGOS one. In particular, the strongest enhancements are not represented at all in MOCAGE, with a difference of more than 60 ppb for the 95th quantile
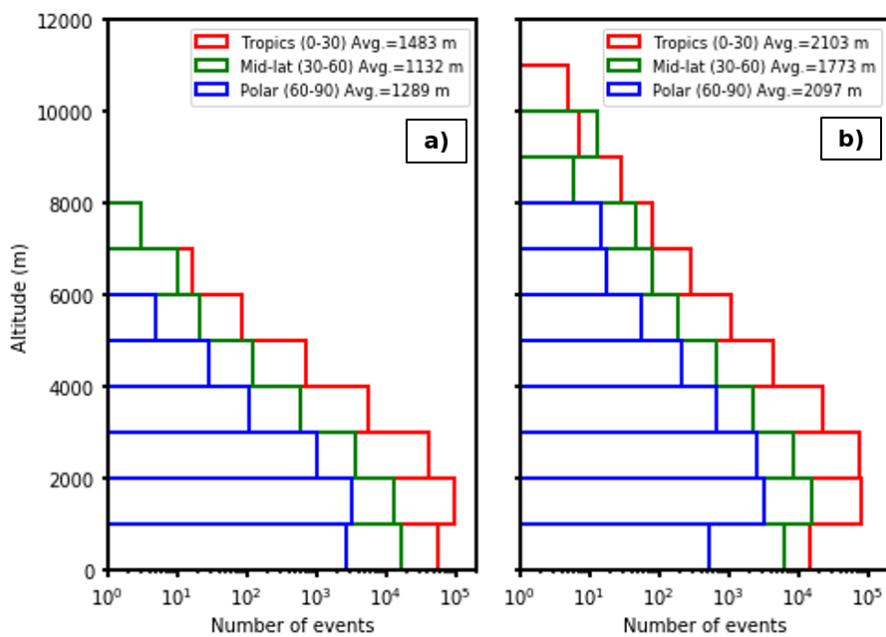
Figure 6. Probability density functions and box plots comparing IAGOS CO mixing ratios (in black) inside identified African Biomass burning plumes and corresponding MOCAGE simulated CO mixing ratios (in red) from the **a)** BASE simulation **b)** INJH simulation. Whiskers on box plots represent the 5th and 95th quantiles and the black diamond is the arithmetical mean.
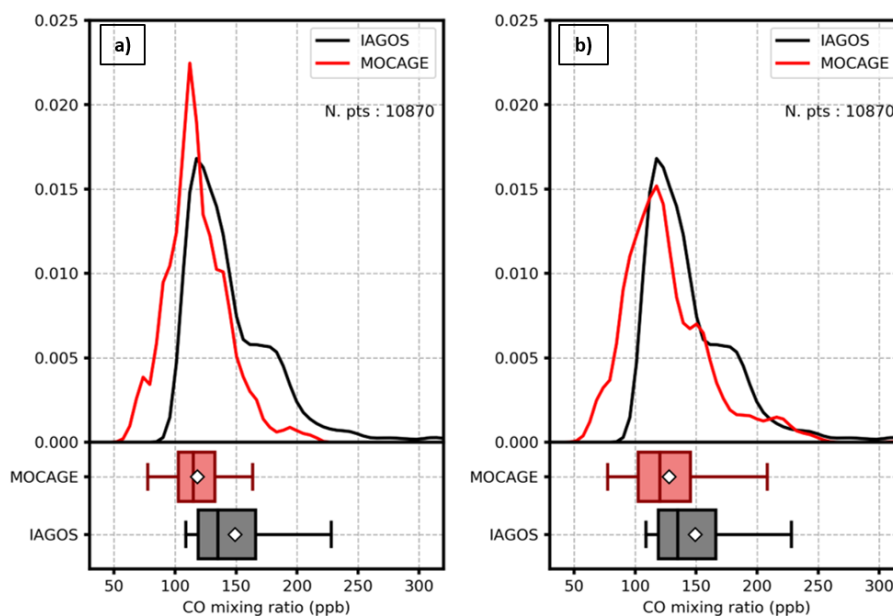
between the BASE simulation and IAGOS CO mixing ratios. Using injection heights from GFAS improves the simulation results. The INJH repartition of simulated CO mixing ratios is a lot more similar to the observed one when compared to the BASE simulation. There is a slight negative bias of around 20 ppb between the modelled plumes and the measurements, but the strongest anomalies above 200 ppb are now represented. Still, a significant part of the simulated CO mixing ratios fall in the

5   50-100 ppb range, the background level of CO concentrations in the UT. This is a least partially due to the numerical diffusion, in addition to the vertical resolution of MOCAGE, which is 700-800 m at the considered range of altitude.

In Fig. 9 the same comparison is performed but for plumes originating from other regions than AFR or BOREAL. Again, MOCAGE seems to be able to correctly represent part of the CO enhancements inside biomass burning plumes, with little difference being made by the injection method globally. This time a more significant part of simulated CO mixing ratios fall

10   towards background values for the upper troposphere as two modes appear in the MOCAGE PDF. It could be explained as the plumes being misrepresented more often in the model as they are sampled further from their origin. Lastly, considering the dataset regardless of the origin of the plume (not shown), African emissions contributes to more than half of the data points, hence conclusions are the same as for Fig. 6.
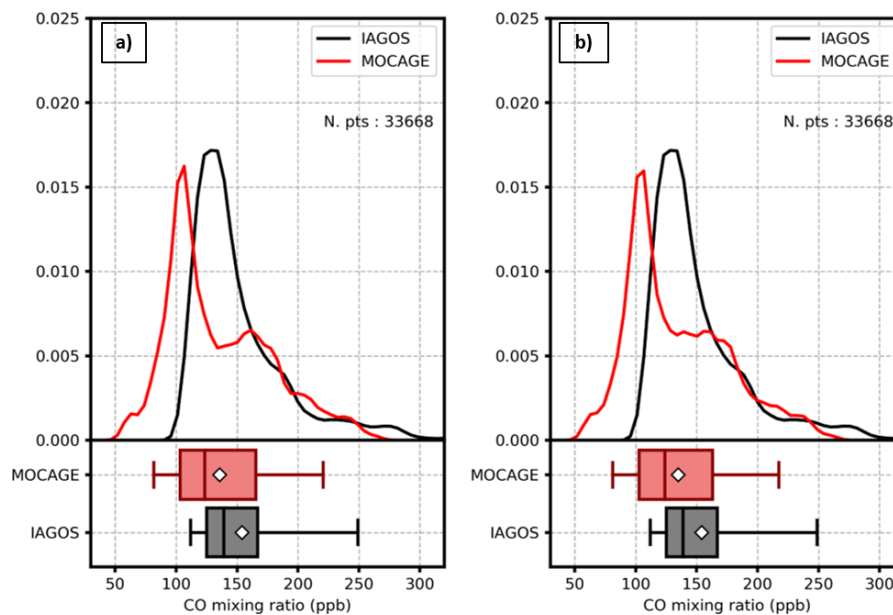
In order to complement the analysis, statistical scores have also been computed for each of dataset of plumes, for the

15   two different MOCAGE simulations against the IAGOS biomass burning plume measurements. The chosen metrics are the Spearman's correlation coefficient, the Modified Normalized Mean Bias (MNMB) and the Fractional Gross Error (FGE). Those

Figure 7. Vertical distribution of GFAS plume rise parameters over 2013 at a 1°longitude × 1°latitude resolution. **a)** Injection Height and **b)** Plume top altitudes

Figure 8. Same as Fig. 6, but for biomass burning plumes originated from boreal regions.

Figure 9. Same as Fig. 6, but for biomass burning plumes originated from other regions than AFR or BOREAL.

three metrics have been chosen to better represent an atmospheric chemistry model performance against in-situ data and have been used in recent studies (Elguindi et al., 2010; Wagner et al., 2015). Each metrics calculation is detailed in the Appendix A. Spearman's correlation coefficient ranges from -1 (perfect anti-correlation) to 1 (perfect correlation). MNMB ranges from -2 to 2, with 0 being the target for no bias. FGE ranges from 0 to 2 with 0 being the target as well. The statistical

5   evaluation of the simulations is presented in Table 3.

   Looking at MNMB and FGE across the different origins, both scores are within acceptable range for each MOCAGE simulation. The MNMB is negative for all set of plumes, meaning that the model slightly under-represents CO enhancement inside biomass burning plumes. This can easily be explained as the model being diffusive, considering the size of each grid cell. Spearman's correlation is acceptable across the different plumes origin, though not great either. This can be explained by

10   the plumes being slightly offset spatially and/or temporally in the model, due to uncertainties in the dynamics as well as both the vertical and horizontal resolution.

   For all origins except the boreal regions, metrics are the same regardless of the method of injection of biomass burning emissions. This is consistent with the comparison presented earlier in this section. For plumes originating from boreal regions, both the correlation and the MNMB are significantly improved using the injection height and plume top estimation from GFAS.

15   Again, this is consistent with comments made on Fig. 8. It also brings the MNMB closer to values considered for other plumes, making the representation of biomass burning plumes in the model more consistent overall.

Table 3. Comparison of scores between MOCAGE simulation and IAGOS measurements for plume identified thanks to SOFT-IO

Origin (N. plumes)		BASE	INJH
AFR (95)	Sp. Corr.	0.45	0.46
	MNMB	-0.07	-0.08
	FGE	0.17	0.17
BOREAL (51)	Sp. Corr.	0.60	0.67
	MNMB	-0.21	-0.15
	FGE	0.24	0.22
OTHERS (74)	Sp. Corr.	0.61	0.61
	MNMB	-0.14	-0.14
	FGE	0.23	0.23
ALL (220)	Sp. Corr.	0.54	0.55
	MNMB	-0.10	-0.10
	FGE	0.19	0.19

To summarize, the latitude dependent injection profile historically used in MOCAGE is well adapted to represent most of the enhancement of CO related to biomass burning plumes in the UT. However, using GFAS products to recompute injection profile for each fire event improved the representation of plumes originating from boreal regions.
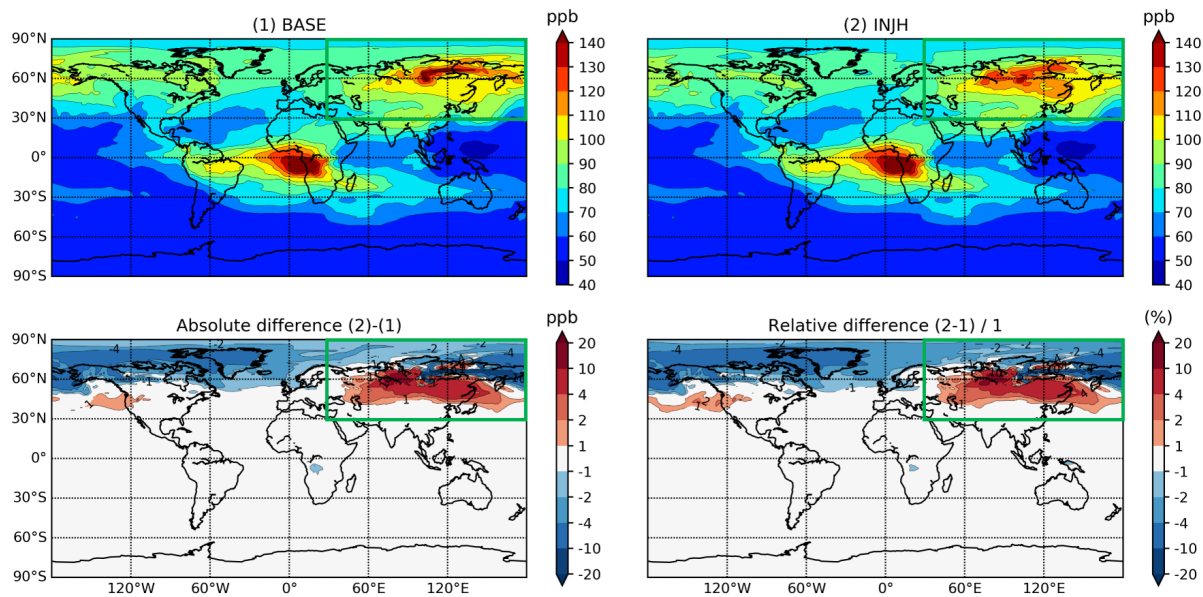
3.2   Global impacts of injection profile

As we want to investigate the impact of using GFAS plume rise parameters on carbon monoxyde mean repartition in the upper troposphere, a global comparison is performed between the BASE and INJH MOCAGE simulation. The results are monthly averaged CO, but only the month of August 2013 is shown hereafter as it is the month where most of the impact can be seen.
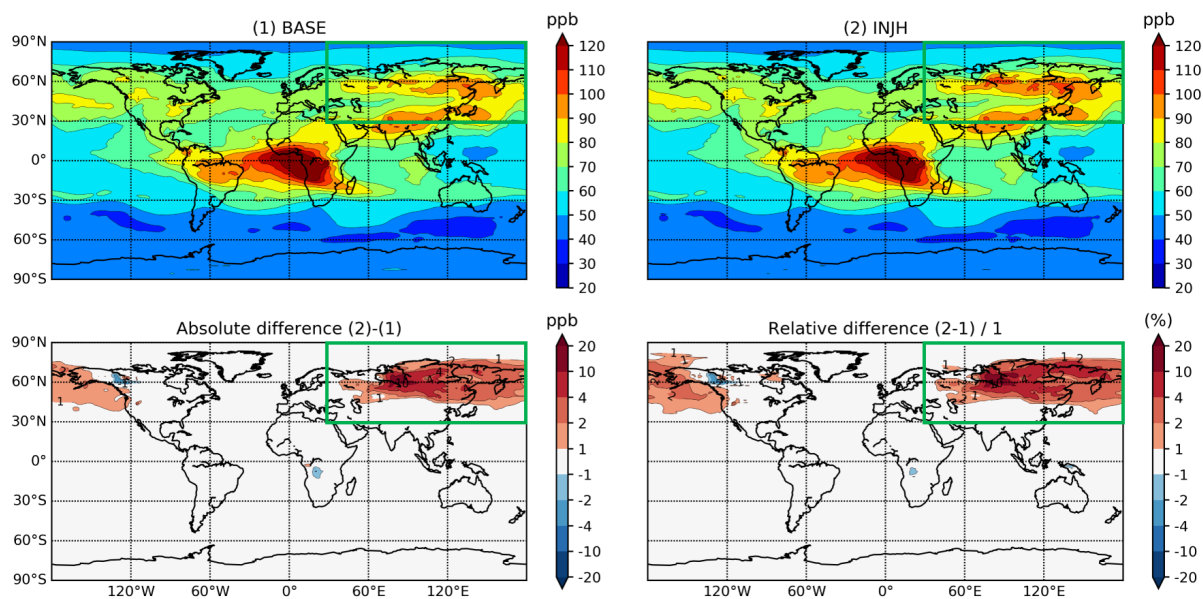
In Fig. 10 is displayed the August 2013 CO vmr mean for both BASE and INJH MOCAGE simulations at 350 hPa, as well as absolute and relative differences between the two fields. At this pressure (an altitude around 8 km), most of the difference between the two experiments is situated in the northern hemisphere, at mid to high latitudes, and especially above the Asian boreal forests. The 60°N parallel stands out as the demarcation line between different injection height (from mid-latitudes to high-latitudes) in the BASE simulation. It results in more CO being found at this altitude South of the 60°N parallel, up to 15 pbb, and less North, up to -25 ppb.

The same comparison is shown on Fig. 11, but interpolated at 250hPa. At this pressure level (around 10 km altitude), the only noticeable difference is also located above the Asian boreal regions, where using GFAS plume rise parameters leads to more injection of CO. This is due to the few events of extreme pyroconvection that are characteristic of high-latitudes forest fires (Nedelec et al., 2005).
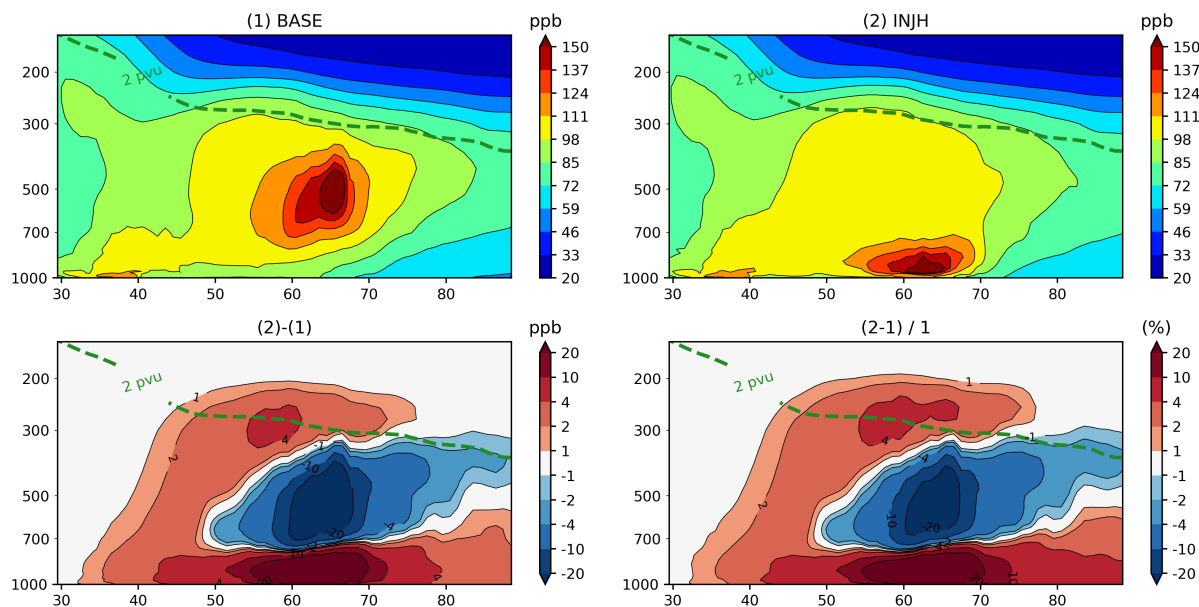
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Figure 10. Monthly mean differences of CO mixing ratios between the BASE and IJNH MOCAGE simulations on 350 hPa iso-surface for the month of August 2013. The green box indicate the area used for the zonal mean.

Figure 11. Same as in Fig.11 but at 250 hPa.

Figure 12. Zonal mean of CO mixing ratios for the month of August 2013. The mean is calculated between 30N-90N and 30E-180E. Doted green line indicates the 2 pvu iso-surface

It is also noticeable that no other area of the globe shows differences regarding the injection method used for biomass burning emissions. Despite finding injection heights and plume top altitudes reaching the upper troposphere all over the globe (see Fig 7), only the events in the Asian boreal forests seem to have emissions reaching the top of the troposphere consistently enough to influence the monthly concentration of CO.

5   Knowing that most of the impact of the INJH method of injection is over the Asian boreal forests during the fire season (i.e. boreal summer), a zonal mean is performed over the box defined in Fig. 10. This zonal mean is presented in Fig. 12, for the month of August 2013 as well. In the BASE simulation, the maximum of CO mixing ratios is well defined and located in the free troposphere, around 600 hPa and between 60°N and 70°N. This is certainly the result of the fixed injection profile used in this method. While it allows CO to be injected in the free troposphere, it probably misrepresents its vertical distribution, as the

10   mean GFAS injection height at these latitudes is around 2000 m. The difference between the INJH and the BASE simulation further confirms this. In the INJH simulation, CO in much more abundant in the PBL than in the BASE simulation. It also leads to an enhancement of CO mixing ratios above the dynamical tropopause identified as the 2 pvu iso-surface, probably as a result of the highest injection occurring above 6 km.

As shown in section 3.1, the INJH simulation showed better results for the simulation of biomass burning plumes originating

15   from boreal forests. This new method of injection of fire emissions also results in a more realistic repartition of CO over the Asian boreal region, but little to no impact elsewhere over the globe. The GFAS plume rise parameters have helped improving

17

Table 4. Statistical scores comparing the INJH MOCAGE simulation against the IAGOS measurements. The annual mean of each score is written in bold, followed by the seasonal mean in brackets as follows (DJF | MAM | JJA | SON).

	From mid to high latitudes	Tropics
MNMB	**0.02** (0.02 | 0.05 | 0.01 | 0.01)	**0.03** (0.04 | 0.06 | 0.02 | 0.01)
FGE	**0.18** (0.15 | 0.19 | 0.20 | 0.19)	**0.17** (0.16 | 0.17 | 0.17 | 0.16)
Correlation	**0.84** (0.85 | 0.87 | 0.82 | 0.82)	**0.75** (0.76 | 0.73 | 0.74 | 0.76)

the representation of biomass burning emissions in MOCAGE, and this configuration of the model will now be used to assess the impact of biomass burning on the budget of CO in the upper troposphere.

4 Global impact of biomass burning on UT air composition

4.1 MOCAGE evaluation against IAGOS

5 Before assessing the impact of biomass burning on upper tropospheric CO, the INJH MOCAGE simulation is compared against the IAGOS database for the year of 2013. The objective is to validate the ability of MOCAGE to represent realistic CO in UTLS altitude range. For this purpose we calculate statistical scores for the IAGOS measurements made during the cruise part of the flights to avoid sampling the free troposphere and the PBL happening during ascent or descent of the plane. Statistical scores used are the MNMB and FGE described in Appendix A for the same reasons given in Sect. 3.1. The results are presented in

10 Table 4, where scores have been detailed for mid-to-high latitudes (above 30°N) and the tropics (below 30°N).

Overall the statistics are good regardless of the considered latitude, with both MNMB and FGE being close to zero, indicating a good representation of CO mixing ratios by MOCAGE. Correlation is also high with a mean value over 0.75 in the tropics and 0.84 at higher latitudes. The higher correlations in the mid to high latitudes is due to the aircraft sampling both the UT and the LS whereas the flights at tropical latitudes only sample upper tropospheric air and thus a smaller range of concentrations. It also

15 indicates that MOCAGE seems to correctly represent the transition between tropospheric and stratospheric air. Additionally, there is no evidence of an annual cycle in the bias or errors concerning CO, as all the statistical scores are nearly constant through the different seasons.

To complement the validation of MOCAGE behavior in the UTLS, vertical profiles are computed using measurements during the ascent and descent phase of the aircraft. Airport clusters are formed by selecting nearby airports in order to take into account

20 a larger number of flights. The airports considered in each cluster are described in Table 5, as well as the total number of flights taking of or landing at each of these airports or cluster of airports. These profile are calculated by averaging measurements on fixed pressure levels (a total of 16 levels logarithmically spaced between the ground and the highest pressure reached during the ascent). Only measurements within 400 km of each airport of the considered cluster are used for the profiles, to keep the profiles representative of a geographical region.

Table 5. Description of used airport clusters for vertical profiles, as well as the total number of vertical profiles.
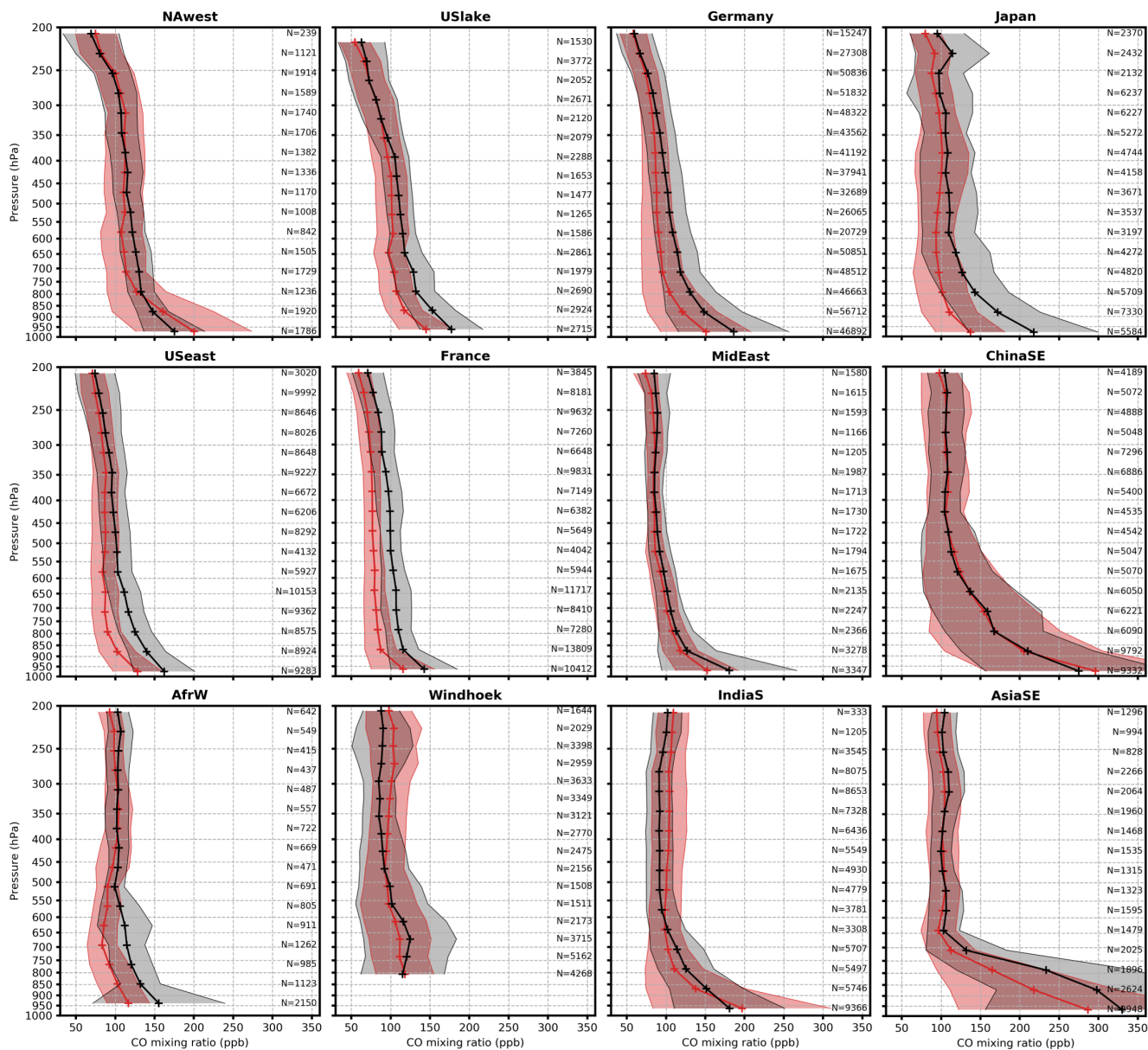
Cluster's name	Airports	N. profiles
NAwest	Seattle, Vancouver	215
USlake	Chicago, Detroit, Toronto	89
USeast	New York City, Boston, Washington D.C., Philadelphia	265
France	Paris (CDG and ORY)	318
Germany	Frankfort, Düsseldorf, Munich	1567
MidEast	Dubai, Abou Dabi, Muscat, Riyadh, Kuwait	158
Windhoek	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

The 12 mean profiles are presented in Fig. 13. Profiles are presented from the ground to 200 hPa (12 km), but MOCAGE performances will mostly be discussed in the upper troposphere (300-200 hPa), which is the region of interest in this study.

Looking at profiles representative of mid-latitudes (i.e. NAwest, USlake, USeast, France, Germany, and Japan), they show similarities when comparing MOCAGE CO mean mixing rations to IAGOS. Around 500 hPa, MOCAGE CO mixing ratios are underestimated by 10 ppb or more, but get closer to the IAGOS mean profile upper in the atmosphere, except for the France cluster. In this specific cluster MOCAGE keeps underestimating upper tropospheric CO by 10 ppb. It could be due to an overestimation of the oceanic influence of less polluted air in MOCAGE. At mid latitudes, both the vertical gradient of CO in the UTLS and the concentrations are captured by the model.

For the six other clusters, all located within the tropics, only the troposphere is sampled as the tropopause is above any commercial flight levels in these regions, resulting in almost no vertical gradient in CO concentrations. In the AfrW, AsiaSE, MidEast and ChinaSE clusters, MOCAGE mean vertical profiles matches very well the IAGOS ones with a similar variability. At the Windhoek and InsiaS clusters however, MOCAGE tends to overestimate CO mixing ratios above 400 hPa by almost 20 ppb. MOCAGE mean profile still falls within the standard deviation around IAGOS mean profile. These overestimations of upper tropospheric CO could be a conjunction of an overestimation of surface emissions (especially for IndiaS, where MOCAGE overestimates surface CO), and an overestimation of convective transport, since both of these profile show a deficit of simulated CO in the PBL.

To summarize, MOCAGE chemistry transport model, when compared to IAGOS in-situ observations, is able to reproduce realistic UTLS CO mixing ratios.

Figure 13. Comparison between IAGOS (in black) and MOCAGE INJH (in red) 2013 mean vertical CO profiles at the 12 airport clusters defined in table 5. The shaded grey and red areas represent respectively the standard deviation around the IAGOS and the MOCAGE mean. The number of data points used for each each pressure level mean is indicated on the right side of each profile.

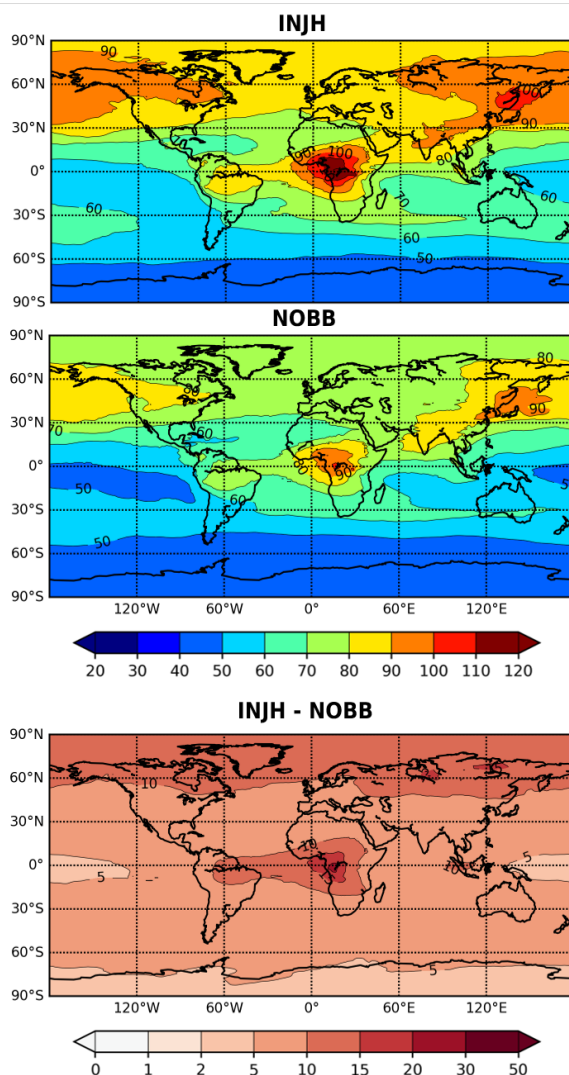## 4.2 Global impact of biomass burning on upper tropospheric air composition

In order to assess the impact on global CO concentration in the upper troposphere over the globe, the INJH and NOBB MOCAGE experiments are compared. As we want to investigate the contribution of biomass burning emissions on upper tropospheric composition, an upper tropospheric layer is defined in the MOCAGE simulation results. This layer is defined

5   relative to the dynamical tropopause, ranging between -15 hPa to -65 hPa below the 2 pvu iso-surface, as done in previous studies analysing the UTLS with IAGOS data (Thouret et al., 2006; Cohen et al., 2018). Around the equator, where the potential vorticity rises exponentially, the dynamical definition is replaced by the 380 K potential temperature iso-surface whenever the 2 pvu iso-surface gets higher. The MOCAGE results are then averaged in this layer. The estimation of the contribution of biomass burning is calculated by subtracting the results of the NOBB simulation to the INJH one.

10   In Fig. 14 are displayed the MOCAGE yearly CO mixing ratios for 2013 in the upper troposphere for the INJH and the NOBB experiments, as well as the calculated difference between the two. A similar map as the INJH but with IAGOS data could not be produced as a year of IAGOS flight is not enough to establish a representative climatology. However, the map looks consistent with the 15 year IAGOS CO climatology shown in Cohen et al. (2018). It appears that biomass burning emissions contribute globally to upper tropospheric CO. The averaged contribution can be estimated around 8 ppb for 2013,

15   which is between 10 and 15 % of the CO mixing ratios at this altitude. Looking at the difference between the two simulations, 2 regions stand out with above average contribution of biomass burning, equatorial Africa and Boreal latitudes, with locally more than 15 ppb of CO of difference between the two simulations. This is expected as these two regions represent respectively 42 % and 20 % of CO emissions in 2013, and transport pathways above these regions also favours injection of CO in the upper troposphere. As mentioned in Sect. 3.1, African emissions are driven to the upper troposphere thanks to deep convection, while

20   Boreal fires emissions are transported directly high in the troposphere by pyroconvection.

In Fig. 15 is presented the monthly mean evolution of the absolute difference of UT CO mixing ratios between the INJH and the NOBB simulations. It is worth noting that at all times of the year, biomass burning contributes to simulated UT CO mixing ratios, by more than 5 ppb. In an expected way, the contribution of biomass burning is higher directly above the most active open fire areas. Equatorial Africa is impacted all year round, north of the equator from December to May, and south of the

25   equator from June to November, with a contribution of more than 15 ppb of CO. Boreal fires have the strongest impact during the two most active months of forest fires (July and August), with an enhancement of up to 67 ppb of CO at boreal latitudes, which is approximately half of the CO in this region. Other regions being impacted are the Amazon forest during its fire season (September to November), and Indonesia in October. Even though the strongest impacts are local, the relatively long lifetime of CO (2 months) makes biomass burning emissions impacting UT air composition on a global scale.

30   In order to clarify how each region affects the upper tropospheric CO, different variations the INJH simulation were performed. In each one, the biomass burning emissions were activated only for one region of the globe with significant amount of CO emitted from biomass burning (see Fig. 4). The chosen regions are the following :

- AFR (NHSA and SHSA), with 124 TgCO in 2013 (42 %)
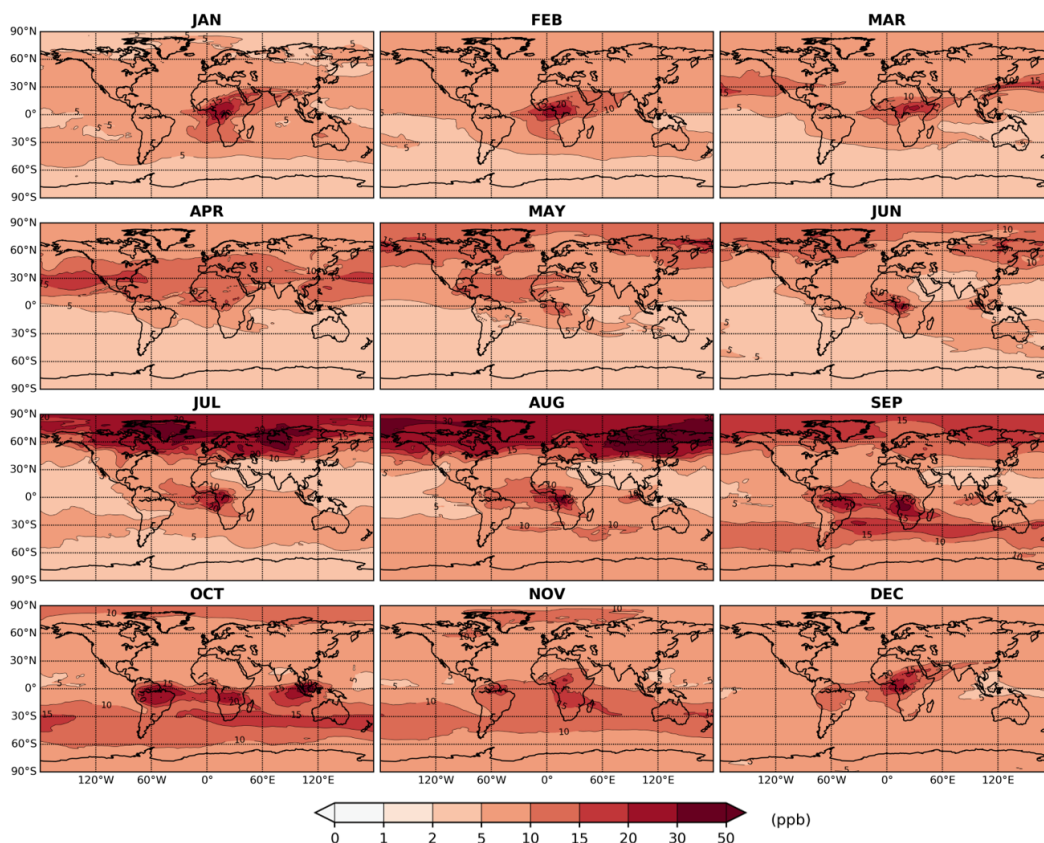
- BOREAL (BOAS and BONA), with 62 TgCO in 2013 (21 %)

Figure 14. Map of 2013 upper tropospheric mean mixing ratios of CO for the INJH and NOBB simulations, as well as the absolute difference between the two.

- AMAZON (NHSA and SHSA), with 32 TgCO in 2013 (11 %)
- EQAS, with 17 TgCO in 2013 (6 %)

The resulting yearly means of upper tropospheric CO are presented in Fig. 16. As expected, impacts are greatest for African
5   and Boreal fire emissions, since both of these regions are the main contributors to annual emissions overall. Due to atmospheric mixing, African emissions have a wider and global impact, with an enhancement between 2 and 5 ppb on upper tropospheric CO. Boreal fire emissions also have a strong impact with an enhancement of 5 to 10 ppb, but limited to boreal latitudes.
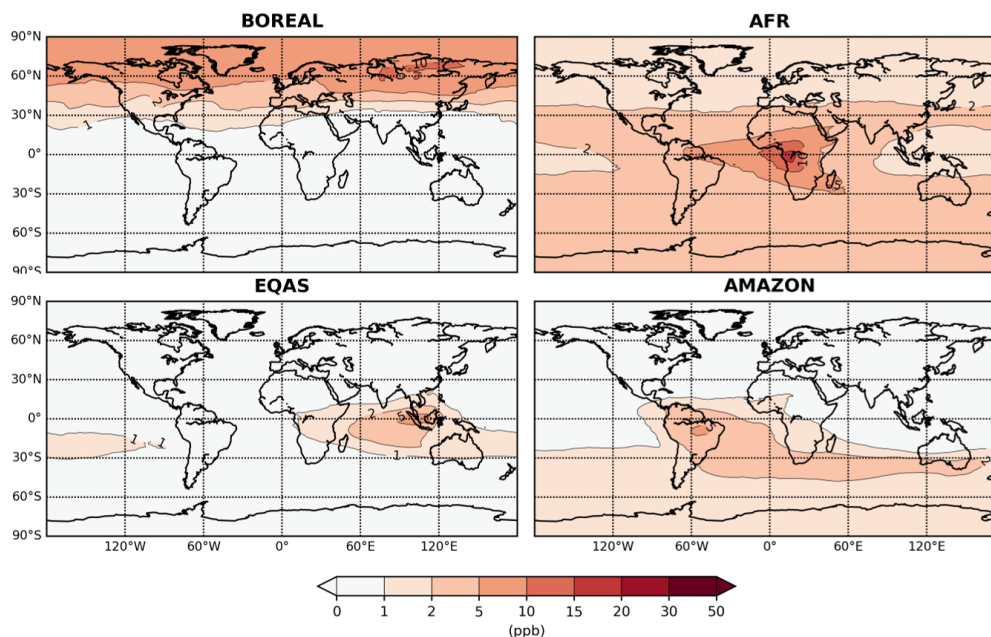
Figure 15. Maps of the 2013 monthly mean difference in upper tropospheric CO mixing ratios in ppb between the INJH and the NOBB
MOCAGE simulations.

Emissions from the Amazon forest, despite being slightly lower than usual in 2013 (Petetin et al., 2018), impact the upper
troposphere by more than 5 ppb directly above the regions, but also a good part of the southern hemisphere through general
circulation. Emissions over the maritime continent (EQAS) mostly have a local impact on the upper troposphere, for around 5
ppb of CO.

5   Three additional simulations were performed where emissions were limited to the SEAS or AUST regions, as well as a
last one for the 5 remaining regoins (TENA, CEAM, EURO, MIDE and CEAS), since they account for similar amount of
CO emissions in 2013 (respectively for 22 Tg, 15 Tg and 23 Tg) as the EQAS region. The results are not shown since no
noticeable effect was seen on upper tropospheric CO. However they allowed for a verification of this method by adding each
CO contribution field and comparing them to the one presented in Fig. 14. Yearly mean results (not shown) matched closely
10 within 1 ppb. Australian fires, despite being active almost all year long, might not have fast enough transport pathway to
the upper troposphere, as convection is less prevalent than in equatorial Asia, and fires are more spread out geographically.
Australian forest are also not known for their pyroconvective events. SEAS emissions on the other hand, despite being more

Figure 16. Maps of upper tropospheric CO yearly mean differences between the four sensitivity tests of the INJH simulation and the NOBB simulation, in ppb.

important than in EQAS, does not impact the upper troposphere. This is probably due to the time of occurrence in 2013 of the fires. SEAS fires mostly occur from February to May, before the beginning of the south asian monsoon, whereas EQAS fires occurs from June to October, covering the whole monsoon season, when fast vertical transport is the most effective through deep convection. In conclusion, biomass burning emissions contribute globally to the upper tropospheric CO budget. The impact is stronger above the areas with the strongest fire emissions, but is non negligible elsewhere around the globe due to atmospheric transport.

5 Summary and conclusions

Biomass burning is a major source of carbon monoxide in the atmosphere, This study aimed at improving the understanding of biomass burning emissions contribution to upper tropospheric carbon monoxide, using global simulations performed with MOCAGE chemistry-transport model. We have chosen that this study would span over a single year, a compromise between the need to study seasonality of biomass burning emissions and computational efficiency as simulations were carried out globally at a fine resolution and with frequent outputs. The year of 2013 was chosen as it was the most recent year representative of average biomass burning emissions in all regions of the globe, as well as the largest number of validated IAGOS flights.

Firstly, GFAS latest products (injection height and plume top altitudes) were used in MOCAGE to improve the injection scheme for biomass burning emissions (INJH simulation), as they are expected to better represent the characteristics of wildfire

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plumes and injection in the upper part of the troposphere dur to pyroconvection. To validate the results of this simulation, IAGOS in-situ observations were used, in addition with SOFT-IO ancillary data on CO anomalies source attribution. Thanks to the SOFT-IO contribution estimations, 220 plumes were selected from the IAGOS flights of 2013, so as to validate the use of GFAS injection height and plume top altitude in MOCAGE. Results were analysed accordingly to the region of origin of

5 the plume. The original plume representation in MOCAGE performs fairly well. However, the new injection scheme greatly improves the modelled plumes from boreal wildfires. It was also shown the prevalence of pyroconvection on CO vertical repartition at northern high latitudes, but not elsewhere, despite higher occurrence of wildfires. To go a step further, it would be interesting to investigate the sensitivity to the shape of injection profile would impact vertical repartition of CO. Based on the good results obtained with MOCAGE, we encourage further work on the use of products such as GFAS injection height

10 for biomass burning emission in other models, especially when studying long-range transport of boreal plumes that can often reach the upper troposphere through pyroconvection only.

Secondly, the INJH simulation was evaluated using all the available CO IAGOS measurements of 2013 before being used to study the contribution of biomass burning to upper tropospheric CO. The extensive IAGOS database has been used in two ways :

15   – Vertical profiles at different airport clusters were used to validate CO levels as well as its gradient in the UT at specific locations.

    – Overall statistical scores calculation in the UT confirmed MOCAGE is well suited tu study upper tropospheric CO.

Finally, the main objective of this study was achieved, by comparing different simulations in which biomass burning emissions were activated or not in different regions of the globe in order to give an estimation of their contribution to upper

20 tropospheric CO. On average the contribution is 8 ppb in 2013, but gets up to 20 ppb above the regions with highest emissions (i.e. equatorial Africa and the boreal forests). Other regions where emissions have a significant impact are the Amazon forest and equatorial Asia. Overall the quantity of emitted CO seems to be the primary factor driving the upper tropospheric contribution, followed by the existence of fast vertical transport above or close to the burning areas (either deep convection or pyroconvection).

25 The conclusions of this work are of course limited to the year of 2013, and since it was a year with no regions with particularly high emissions of CO from wildfires, estimations might be on a lower hand of what they could be. It would nonetheless be interesting to see how it translate for other years with higher emissions and extend these simulations to other years to evaluate the role of inter-annual variability. It is possible notably since the IAGOS database expands over more than 15 years for CO. Before this, the first step will be to extend the analysis of the numerical simulations used in this paper, looking at wildfires

30 impact on upper tropospheric chemistry. The ozone budget especially will be investigated since CO is its main precursor at this altitude.

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Data availability. The IAGOS time series (https://doi.org/10.25326/06), vertical profiles (https://doi.org/10.25326/07) and the SOFT-IO ancillary data (https://doi.org/10.25326/3) used in this study can all be found on the IAGOS data portal (https://doi.org/10.25326/20)

Appendix A: Statistical score used for evaluation

The metrics used in this study for evaluation of modelled variables against in-situ observations are the modified normalized

5 mean bias (MNMB), the fractional gross error (FGE), as well as Pearson's linear correlation coefficient and the Spearman's rank correlation coefficient. The MNMB and the FGE have been chosen over the standard mean bias (MB) and root mean square error metrics (RMSE),as they have been prescribed for atmospheric air composition studies where differences between a model and observations can be larger than for other meteorological fields (Seigneur et al., 2000).

For a dataset of N observations points o_i and co-located modelled value f_i, the MNMB is defined as :

$$10 \quad MNMB = \frac{2}{N} \sum_i^N \left(\frac{f_i - o_i}{f_i + o_i} \right) \tag{A1}$$

It ranges from -2 to 2 and behaves symmetrically in regards of underestimation and overestimation.

$$FGE = \frac{2}{N} \sum_i^N \left| \frac{f_i - o_i}{f_i + o_i} \right| \tag{A2}$$

The FGE ranges from 0 to 2, and unlike the RMSE it does not overweight data points with the highest observed values within a given dataset.

15 In order to asses the model capability to reproduce the observed spatio-temporal pasterns, two types of correlation coefficient were used. Pearson's linear correlation coefficient is defined as follows :

$$r = \frac{1}{N} \frac{\sum_i^N (f_i - \overline{f})(o_i - \overline{o})}{\sigma_f \sigma_o} \tag{A3}$$

with $\overline{f}$ and $\overline{o}$ the mean value of the forecast and the observed values, as well as σ_f and σ_o the associated standard deviation. This coefficient ranges between -1 for a perfect anti-correlation to 1, a perfect correlation. However, it assumes a linear relation

20 between the forecast and the observations. This is not always the case for a global atmospheric composition model, at this is why Spearman's rank correlation coefficient r_s has also been used. It is defined as the following :

$$r_s = \frac{cov(rg_f, rg_o)}{\sigma_{rg_f} \sigma_{rg_o}} \tag{A4}$$

where rg_f and rg_o are the rank of the forecast and observed values within the dataset, with σ_{rg_f} and σ_{rg_o} being their associated standard deviations. Spearman's rank correlation is also bounded between -1 and 1, but does not make the assumption that the

25 forecast and the observed values are linearly related. It particularly is valuable for the study of heavy-tailed distribution of observations, or distributions where strong outliers are expected.

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Author contributions. MC, VM, VT and BJ designed the study and the model experiments. Simulations were carried out by MC with help from BJ. The IAGOS and SOFT-IO data were provided by VT and BS, with guidance on how to use them. Manuscript was written by MC and reviewed by VM, and commented, edited and approved by all the authors.

Competing interests. The authors declare that they have no conflict of interest

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