

Reviewer 2

General Comments

This manuscript presents IASI measurements of formic acid between 2008 and 2014, and uses these data to determine enhancement ratios from biomass burning emissions over seven regions. HCOOH and CO total columns, MODIS fire counts, and ECMWF surface wind speeds are combined to identify enhancements due to biomass burning. Correlations between HCOOH and CO total columns are used to calculate the enhancement ratio in each region. These results suggest that production of HCOOH by Siberian forest fires may be underestimated by 60%, and provide some insights into sources and sinks of HCOOH in other regions studied.

The manuscript provides a useful contribution to the field, but is somewhat qualitative and speculative in places, as noted by the other reviewer. It also has many distracting grammatical errors and should be carefully reviewed and revised to correct these and to improve the clarity of the writing. I recommend publication in ACP after the comments below are addressed.

The authors would like to thank reviewer 2 for his careful reading of the manuscript and for his thorough review. A detailed point by point reply (in blue) is provided hereafter.

As suggested by the first reviewer, an additional work has been done by using backward trajectories from HYSPLIT and land cover information from MODIS.

Moreover the text has been revised. Thus, in addition to our answers, we suggest that the reviewer reads the revised manuscript since Sect. 5.1 and 5.2 were largely rewritten.

A lot of MODIS hotspots have been studied in this paper, in total it represents 9628 hotspots. It is difficult to calculate backward trajectories for each hotspot, especially as different altitude ranges need to be tested since the vertical sensitivity of IASI (CO & HCOOH) is located in the free troposphere.

However, illustrative tests were done to show the distinct origins of the air masses at different locations, periods of the year and altitudes of the plume. Specifically, 5 hotspots have been chosen randomly for each region and 3 different altitudes have been selected: 500 m (thus close to the surface), 2000 m and 5000 m (representing the free troposphere). In total, this represents 105 trajectories.

These trajectories show that the air masses initialized at 500 and 2000m are mainly influencing by air masses close to the surface, confirming an origin near the source of our IASI fire-affected columns. It also shows the difficulty to estimate the origin of the air masses without an accurate knowledge of the altitude of the plumes.

These trajectories were plotted through the HYSPLIT online service:

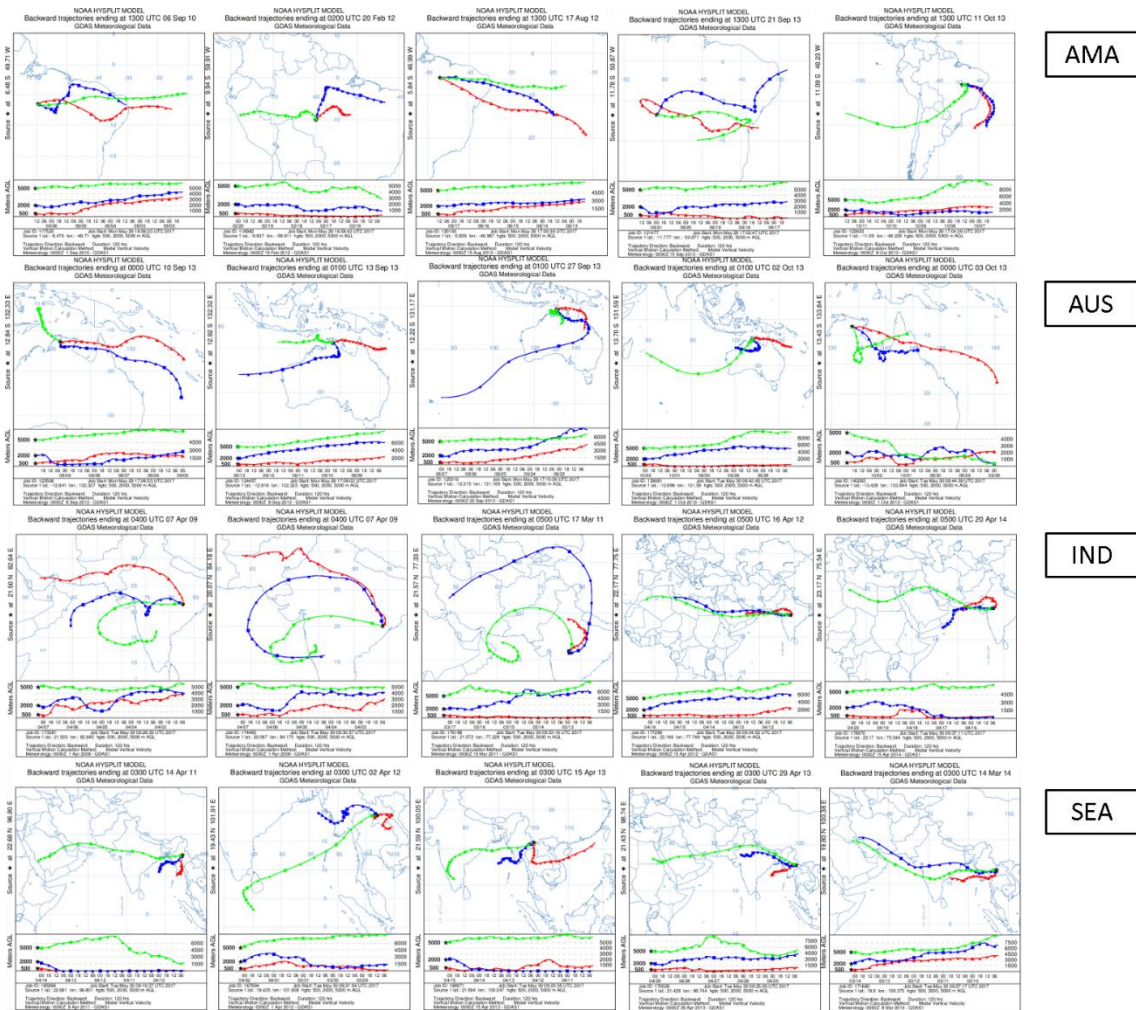


Fig 1. 5-day backward trajectories from HYPPLIT online service calculated at 3 altitudes: 500 m (red), 2000 m (blue) and 5000 m (green), for 5 hotspots chosen randomly over the 7 regions studied in the paper. The parameters characterizing each MODIS hotspots are summarized in the following table. The meteorological fields are from GDAS at $1^\circ \times 1^\circ$ horizontal resolution.

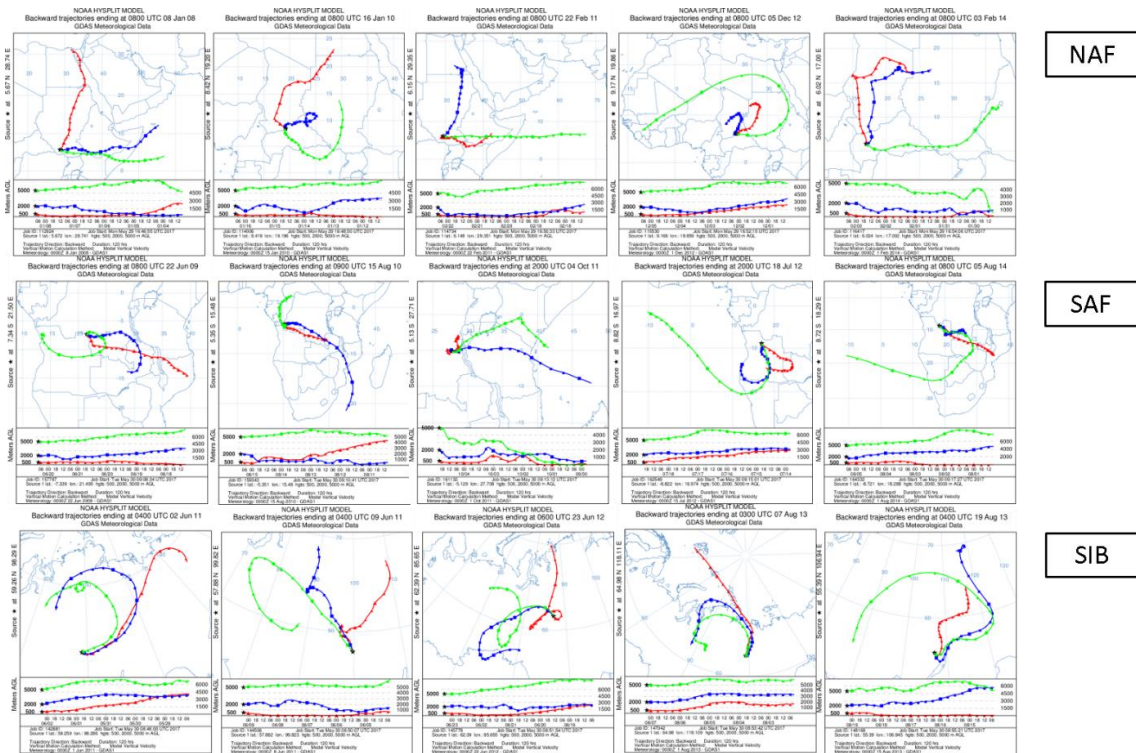


Fig 1. Continue

Tab. Characteristic of each MODIS hotspot used for the trajectories plotted in the previous figure. The dates, the time recorded by the instrument and the coordinates for each hotspot are written.

AMA
20100906 / hour (UTC)=13 / lat=-6.476 - lon=-49.71
20120220 / hour (UTC)=2 / lat=-9.937 - lon=-59.911
20120817 / hour (UTC)=13 / lat=-5.839 - lon=-46.987
20130921 / hour (UTC)=13 / lat=-11.777 - lon=-50.871
20131011 / hour (UTC)=13 / lat=-11.09 - lon=-48.229
AUS
20130910 / hour (UTC)=0 / lat=-12.841 - lon=132.327
20130913 / hour (UTC)=1 / lat=-12.916 - lon=132.323
20130927 / hour (UTC)=1 / lat=-12.215 - lon=131.169
20131002 / hour (UTC)=1 / lat=-13.696 - lon=131.59
20131003 / hour (UTC)=0 / lat=-13.428 - lon=133.844
IND
20090407 / hour (UTC)=4 / lat=21.503 - lon=82.645
20090407 / hour (UTC)=4 / lat=20.067 - lon=84.175
20110317 / hour (UTC)=5 / lat=21.572 - lon=77.328
20120416 / hour (UTC)=5 / lat=22.166 - lon=77.749
20140420 / hour (UTC)=5 / lat=23.17 - lon=75.544
SEA
20110414 / hour (UTC)=3 / lat=22.681 - lon=96.801
20120402 / hour (UTC)=3 / lat=19.435 - lon=101.908
20130315 / hour (UTC)=3 / lat=21.594 - lon=100.047
20130329 / hour (UTC)=3 / lat=21.426 - lon=98.744
20140314 / hour (UTC)=3 / lat=19.8 - lon=100.375

NAF
20080108 / hour (UTC)=8 / lat=5.672 - lon=28.741
20100116 / hour (UTC)=8 / lat=8.419 - lon=19.196
20110222 / hour (UTC)=8 / lat=6.148 - lon=29.351
20121205 / hour (UTC)=8 / lat=9.168 - lon=19.859
20140203 / hour (UTC)=8 / lat=6.024 - lon=17.002
SAF
20090622 / hour (UTC)=8 / lat=-7.339 - lon=21.495
20100815 / hour (UTC)=9 / lat=-5.351 - lon=15.48
20111004 / hour (UTC)=20 / lat=-5.129 - lon=27.708
20120718 / hour (UTC)=20 / lat=-8.822 - lon=16.974
20140805 / hour (UTC)=8 / lat=-8.721 - lon=18.288
SIB
20110602 / hour (UTC)=4 / lat=59.259 - lon=98.286
20110609 / hour (UTC)=4 / lat=57.882 - lon=99.823
20120623 / hour (UTC)=6 / lat=62.39 - lon=85.655
20130807 / hour (UTC)=3 / lat=64.98 - lon=118.109
20130819 / hour (UTC)=4 / lat=55.39 - lon=106.945

We have added the sentences (in bold) in our Section 5.1.2:

“A few backward trajectories (along 5 days, not shown) have been calculated for our hotspots with the online version of the HYSPLIT atmospheric transport and dispersion modeling system (Rolph, 2017). These trajectories, initialized at different altitudes, confirm a main origin close to the surface of our IASI fire-affected columns. It is however impossible to properly compare the origin of the air masses with previous studies as our studied period (2008-2014) or our studied fires do not necessarily match with plumes described in other publications. It is also difficult to estimate the age of our studied air masses by gathering the plumes during a 7-yr period and without an accurate knowledge of the altitude of the plumes.”

And:

“One possible explanation is the multi-origin of the plumes studied by Rinsland et al. (2006), since, based on their backward trajectories, their plumes could be influenced by biomass burning originating from Southern Africa and/or from Southern America. The travel during the few days across the Atlantic Ocean may explain the change in their ER_(HCOOH/CO).”

With the corresponding reference:

Rolph, G.D.: Real-time Environmental Applications and Display sYstem (READY) Website (<http://www.ready.noaa.gov>). NOAA Air Resources Laboratory, College Park, MD, 2017.

We also have discovered that the information about the type of vegetation was available in the MODIS products.

In “Section 3. MODIS” we have added this paragraph:

“To characterize each MODIS hotspot by the type of fuel burned, the Global Mosaics of the standard MODIS land cover type data product (MCD12Q1) in the IGBP Land Cover Type Classification (Friedl et al., 2010; Channan et al., 2014) with a 0.5° × 0.5° horizontal resolution has also been used (<http://glcf.umd.edu/data/lc/>). As the annual variability in this product is limited (not shown) and since the period available (from 2001 to 2012) does not fully match

the period of the IASI mission, only the data for 2012 have been used. Whitburn et al. (2017) have also used this MCD12Q1 product to determine their IASI-derived NH₃ enhancement ratios by vegetation types.”

With the corresponding references:

Channan, S., Collins, K., and Emanuel, W. R., Global mosaics of the standard MODIS land cover type data. University of Maryland and the Pacific Northwest National Laboratory, College Park, Maryland, USA, 2014.

And

Friedl, M. A., Sulla-Menashe, D., Tan, B., Schneider, A., Ramankutty, N., Sibley, A. and Huang, X., MODIS Collection 5 global land cover: Algorithm refinements and characterization of new datasets, 2001-2012, Collection 5.1 IGBP Land Cover, Remote Sensing of Environment, 114 , 168–182, doi:10.1016/j.rse.2009.08.016, 2010.

And

Whitburn, S., Van Damme, M., Clarisse, L., Hurtmans, D., Clerbaux, C., and Coheur, P.-F.: IASI-derived NH₃ enhancement ratios relative to CO for the tropical biomass burning regions, Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-331>, in review, 2017.

We have also added this sentence in Section 4.2:

“The classification of the vegetation from the MODIS product has also been used for a detailed analysis of the enhancement ratios for these regions (Fig. 1).”

And Fig. 1 has been modified as below:

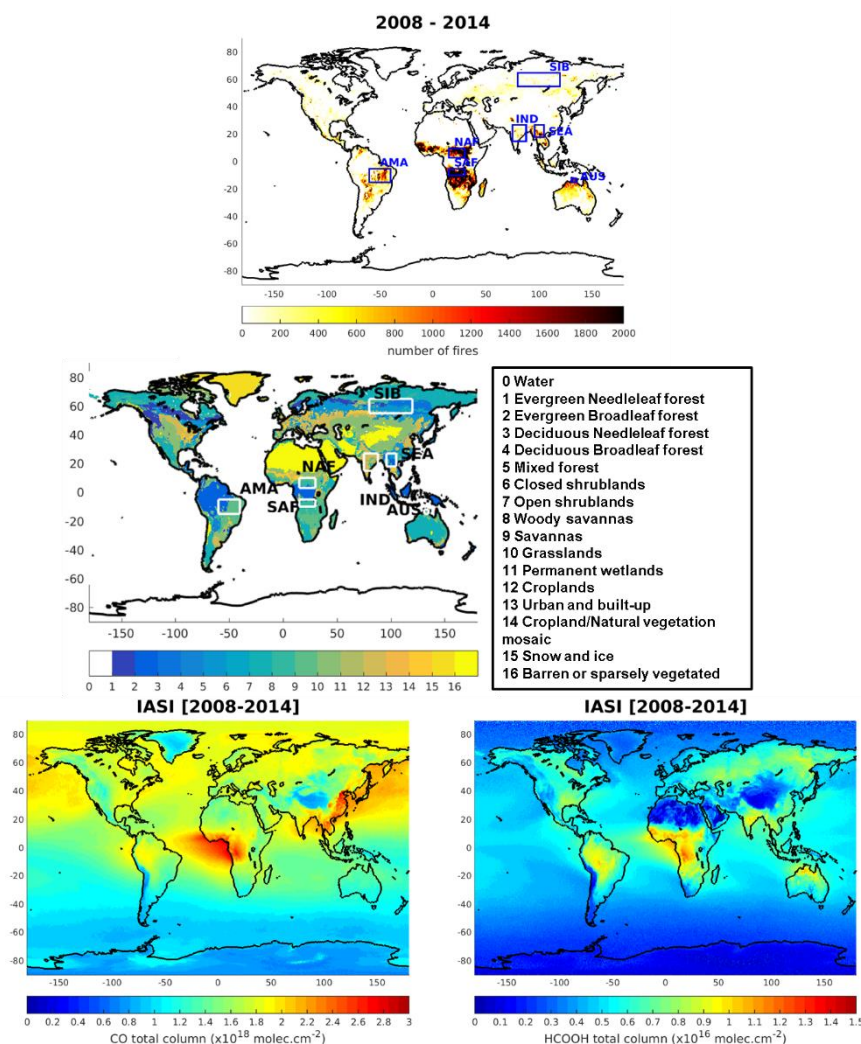


Figure 1: Top: Number of MODIS fire hotspots with a confidence percentage higher or equal to 80%, averaged on a $0.5^\circ \times 0.5^\circ$ grid, for the period between 2008 and 2014. The blue boxes are the regions studied in this work. Middle: Classification of the land cover type from MODIS on the same grid and highlighting the studied regions in white. Each number corresponds to the type of vegetation. Only the data between 64°S and 84°N are available. Bottom: The IASI CO total column distribution (left) and the IASI HCOOH total column distribution (right), averaged between 2008 and 2014 and on the same grid.

Section 5.2 was also rewritten and now named “5.2. Analysis based on the type of vegetation” since ER (HCOOH/CO) by biome have also been added in Table 3 as below:

Table 3. Enhancement ratio of HCOOH relative to CO (mol/mol) with its standard deviation and enhancement ratio of HCOOH relative to CO (mol/mol) by biome with its standard deviation calculated in this work. For each enhancement ratio by biome, the correlation coefficient and the number of MODIS hotspots are provided. The enhancement ratios are compared to emission ratios calculated from emission factors given in the literature for the seven studied regions. For the calculation of these emission ratios, the emission factors of CO for the corresponding fuel type given in Akagi et al. (2011) are used. Emission ratios of HCOOH relative to CO (mol/mol) calculated from the emission factors of HCOOH given in Akagi et al. (2011) for the corresponding fuel type are also provided.

Region	Enhancement Ratio to CO (mol/mol) – this work	Enhancement Ratio to CO (mol/mol) ¹ by biome ² – this work	Emission Ratio to CO (mol/mol) calculated from EF _{HCOOH} given in literature and using EF _{CO} from Akagi et al. (2011)	Instrument used
AMA	$7.3 \times 10^{-3} \pm 0.08 \times 10^{-3}$	$6.3 \times 10^{-3} \pm 0.22 \times 10^{-3}$ (Evergreen Broadleaf forest, r=0.81, n = 454) $3.0 \times 10^{-3} \pm 0.81 \times 10^{-3}$ (Open shrubland, r=0.91, n = 5) $7.0 \times 10^{-3} \pm 2.47 \times 10^{-3}$ (Woody savanna, r=0.63, n = 14) $7.6 \times 10^{-3} \pm 0.09 \times 10^{-3}$ (Savanna, r=0.79, n = 3909) $8.4 \times 10^{-3} \pm 0.39 \times 10^{-3}$ (Grassland, r=0.88, n = 143) $4.6 \times 10^{-3} \pm 0.35 \times 10^{-3}$ (Cropland, r=0.88, n = 54)	1.8×10^{-3} – Tropical forest (Yokelson et al., 2007 ; 2008) ³ 2.7×10^{-3} – Savanna (Yokelson et al., 2007 ; 2008) ³ 2.0×10^{-3} – Savanna (Akagi et al., 2011) 5.2×10^{-3} – Tropical forest (Akagi et al., 2011)	Airborne FTIR (Yokelson et al., 2007) ; laboratory (Yokelson et al., 2008) catalogue
AUS	$11.1 \times 10^{-3} \pm 1.37 \times 10^{-3}$	$5.7 \times 10^{-3} \pm 2.55 \times 10^{-3}$ (Woody savanna, r=0.6, n = 11) $11.2 \times 10^{-3} \pm 1.49 \times 10^{-3}$ (Savanna, r=0.65, n = 80)	2.0×10^{-3} – Savanna (Akagi et al., 2011)	catalogue
IND	$6.8 \times 10^{-3} \pm 0.44 \times 10^{-3}$	$6.6 \times 10^{-3} \pm 0.77 \times 10^{-3}$ (Woody savanna, r=0.65, n = 103) $6.2 \times 10^{-3} \pm 0.62 \times 10^{-3}$ (Cropland, r=0.58, n = 198) $8.8 \times 10^{-3} \pm 1.19 \times 10^{-3}$ (Cropland/Natural vegetation mosaic, r=0.85, n =23)	2.0×10^{-3} – Savanna (Akagi et al., 2011) 2.7×10^{-3} – Extratropical forest (Akagi et al., 2011) 6.0×10^{-3} – Cropland (Akagi et al., 2011)	catalogue
SEA	$5.8 \times 10^{-3} \pm 0.15 \times 10^{-3}$	$5.6 \times 10^{-3} \pm 0.20 \times 10^{-3}$ (Evergreen Broadleaf forest, r=0.83, n = 334)	2.0×10^{-3} – Savanna (Akagi et al., 2011) 2.7×10^{-3} – Extratropical forest (Akagi et al., 2011) 6.0×10^{-3} – Cropland (Akagi et al., 2011)	catalogue

		$6.3 \times 10^{-3} \pm 0.66 \times 10^{-3}$ (Mixed forest, $r=0.76$, $n = 70$)		
		$6.2 \times 10^{-3} \pm 0.38 \times 10^{-3}$ (Woody savanna, $r=0.86$, $n = 99$)		
		$7.1 \times 10^{-3} \pm 0.99 \times 10^{-3}$ (Cropland/Natural vegetation mosaic, $r=0.84$, $n = 23$)		
NAF	$4.0 \times 10^{-3} \pm 0.19 \times 10^{-3}$	$3.4 \times 10^{-3} \pm 0.63 \times 10^{-3}$ (Evergreen Broadleaf forest, $r=0.52$, $n = 78$)	2.0×10^{-3} – Savanna (Akagi et al., 2011)	catalogue
		$3.3 \times 10^{-3} \pm 0.28 \times 10^{-3}$ (Woody savanna, $r=0.44$, $n = 569$)		
		$4.4 \times 10^{-3} \pm 0.29 \times 10^{-3}$ (Savanna, $r=0.59$, $n = 441$)		
		$22.6 \times 10^{-3} \pm 11.06 \times 10^{-3}$ (Cropland/Natural vegetation mosaic, $r=0.67$, $n = 7$)		
SAF	$5.0 \times 10^{-3} \pm 0.13 \times 10^{-3}$	all hotspots are woody savanna	3.3×10^{-3} – Tropical forest (Sinha et al., 2004) ⁴ 4.8×10^{-3} – Savanna (Sinha et al., 2004) ⁴	Airborne FTIR
			4.1×10^{-3} – Tropical forest (Yokelson et al., 2003) 6.0×10^{-3} – Savanna (Yokelson et al., 2003)	Airborne FTIR
			13×10^{-3} – Tropical forest (Rinsland et al., 2006) 19.2×10^{-3} – Savanna (Rinsland et al., 2006)	ACE-FTS
			2.0×10^{-3} – Savanna (Akagi et al., 2011) 5.2×10^{-3} – Tropical forest (Akagi et al., 2011)	catalogue
SIB	$4.4 \times 10^{-3} \pm 0.09 \times 10^{-3}$	$4.0 \times 10^{-3} \pm 0.31 \times 10^{-3}$ (Evergreen Needleleaf forest, $r=0.63$, $n = 245$)	2.7×10^{-3} – Boreal forest (Akagi et al., 2011)	catalogue
		$3.6 \times 10^{-3} \pm 0.16 \times 10^{-3}$ (Deciduous Needleleaf forest, $r=0.66$, $n = 659$)		
		$3.4 \times 10^{-3} \pm 0.18 \times 10^{-3}$ (Mixed forest, $r=0.57$, $n = 759$)		

$6.6 \times 10^{-3} \pm 0.48 \times 10^{-3}$
(Open shrubland,
 $r=0.76$, $n = 143$)

$6.0 \times 10^{-3} \pm 0.41 \times 10^{-3}$
(Woody savanna,
 $r=0.76$, $n = 155$)

$3.8 \times 10^{-3} \pm 0.65 \times 10^{-3}$
(Permanent wetland,
 $r=0.6$, $n = 63$)

¹ Only the enhancement ratio to CO calculated from a scatterplot with a correlation coefficient higher than 0.4 are reported.

² The type of vegetation is defined by the land cover type data product (MCD12Q1).

³ The EF_{HCOOH} were corrected based on the comment from Yokelson et al. (2013) (EF_{HCOOH} used: 0.281 for Yokelson et al. (2007); 0.2767 for Yokelson et al. (2008)).

⁴ The mean of both EF_{HCOOH} values provided in Sinha et al. (2004) were used for our $EmR_{HCOOH/CO}$ calculation

We have added these sentences at the end of the Section 5.2:

“In addition to the $EmR_{(HCOOH/CO)}$ calculated from the EF_{HCOOH} given in the literature, a classification for our $ER_{(HCOOH/CO)}$ has also been done, based on the data from the MCD12Q1 product. As each hotspot is associated with a land cover value defined by the MCD12Q1 product, enhancement ratios by biome have been calculated. The limitations of this dataset are its coarse resolution ($0.5^\circ \times 0.5^\circ$) and the lack of seasonal variation. It gives however a supplementary information on the type of fuel burned identified by MODIS. The corresponding $ER_{(HCOOH/CO)}$ are provided in Table 3. Only the values calculated from a scatterplot with a correlation coefficient higher than 0.4 are reported.”

And

“In general, the $ER_{(HCOOH/CO)}$ calculated for a specific biome varies with the regions. This shows that the type of vegetation is not the only factor influencing the $ER_{(HCOOH/CO)}$. The ongoing chemistry within a plume is important and the age of the air masses impact the level of HCOOH and CO in the plumes.”

We have also added these sentences in the abstract:

“An additional classification of the enhancement ratios by type of fuel burned is also provided, showing a diverse origin of the plumes sampled by IASI, especially over Amazonia and Siberia. The variability in the enhancement ratios by biome over the different regions show that the levels of HCOOH and CO do not only depend on the fuel types.”

And in the conclusion:

“Finally, the estimation of the $ER_{(HCOOH/CO)}$ calculated by the type of vegetation burned, as referenced in the MODIS product, varies with the regions. This shows that other parameters than the type of fuel burned also influence the $ER_{(HCOOH/CO)}$.”

Specific Comments

Page 1, line 1 – The title is awkwardly phrased. Why just a “Possibility” for IASI to detect HCOOH in biomass burning plumes? “document” should be replaced by “measure” or “detect”. A better title might be something like: “Detection of HCOOH from biomass burning plumes by the Infrared Atmospheric Sounding Interferometer”

The title is now:

“Determination of enhancement ratios of HCOOH relative to CO in biomass burning plumes by the Infrared Atmospheric Sounding Interferometer (IASI).”

Page 1, lines 25-27 – Make clear whether this underestimation for Siberian forest fires is in the IASI HCOOH or other studies or both. This seems rather speculative based on the results presented in the paper.

This information has been deleted from the abstract but we have added these sentences (in bold) in the conclusions:

“The underestimation by 60% over Siberia is consistent with conclusions given in R’Honi et al. (2013). **The calculation of the $ER_{(HCOOH/CO)}$ by biome shows that Siberian plumes are related to the burning of six different vegetation classes. The underestimation reported is thus difficult to confirm without the use of a chemical transport model.**”

We have also written in Section 5.2:

“These hypotheses in biased emissions and/or secondary production need, however, to be verified with modeling studies.”

Page 1, lines 27-29 – Rewrite this last sentence for clarity.

Done. Now it reads:

“In comparison **with referenced emission ratios**, it is also shown that the selected agricultural burning plumes captured by IASI over India and Southern East Asia correspond to recent plumes where the chemistry or the sink does not occur.”

Page 5, line 185 – Why is 1.44 m/s used as a threshold?

As explained in the following sentence: "This value of 1.44 m/s for the surface wind speed corresponds to the 25th percentile of the distribution of the three regions characterized by the lowest surface wind speed (Fig. 3)."

Page 6, lines 210-212 – Please clarify this discussion. It is not clear how a better detection limit “minimizes the bias with the lowest columns”, nor what suggests “a negligible effect of the low column biases”.

We agreed it was confusing. Now the sentences are (the modifications are highlighted in bold):

“Nevertheless, in order to investigate the possible impact of the overestimation in the lower columns **and the underestimation in the higher columns** on the calculated ratios, a test was performed, by using only HCOOH columns with a thermal contrast larger than 10K. Indeed, the increase in the thermal contrast (i.e. the temperature difference between the surface and the first layer in the retrieved profile) leads to **reducing** the detection limit as shown in Pommier al. (2016). **This enhancement of the detection level helps to minimize the bias in the retrieved total columns as explained in Crevoisier et al. (2014). For the analysis performed here, similar slopes and correlation coefficients were generally calculated**, suggesting a negligible effect of this parameter on the biases. The only exception is an increase in $ER_{(HCOOH/CO)}$ over Siberia ($6.5 \times 10^{-3} \pm 0.19 \times 10^{-3}$ mol/mol when using only IASI measurements with TC above 10K against 4.4×10^{-3} mol/mol $\pm 0.09 \times 10^{-3}$ in Table 2). **It is worth noting that only 48% of the selected scenes remain over Siberia when applying this filter on thermal contrast (60% for SEA, 77% for AMA, 80% for SAF, 83% for AUS and NAF, and 89% for IND). This implies that the statistics on the fire emissions in the higher latitudes of Siberia is dominated by measurements with a low thermal contrast and thus with HCOOH total columns with higher uncertainties. However, the limited changes in slopes and correlation coefficients give us confidence that the results presented in Table 2 are representative.**”

We also have added this reference:

Crevoisier, C., Clerbaux, C., Guidard, V., Phulpin, T., Armante, R., Barret, B., Camy-Peyret, C., Chaboureau, J.-P., Coheur, P.-F., Crépeau, L., Dufour, G., Labonnote, L., Lavanant, L., Hadji-Lazaro, J., Herbin, H., Jacquinet-Husson, N., Payan, S., Péquignot, E., Pierangelo, C., Sellitto, P., and Stubenrauch, C.: Towards IASI-New Generation (IASI-NG): impact of improved spectral resolution and radiometric noise on the retrieval of thermodynamic, chemistry and climate variables, *Atmos. Meas. Tech.*, 7, 4367-4385, doi:10.5194/amt-7-4367-2014, 2014.

Page 6, para 3 – This is a long paragraph, written in a way that is hard to follow. Please revise for clarity. e.g., lines 224-228 – Explanations are also not clear here. Please explain why the results suggest that the plume “encountered a limited secondary production or a low sink as deposition or reaction with OH” and why the faster decay of HCOOH relative to CO, suggests rapid advection of the plumes.

The section has been rewritten. Please refer to the revised manuscript.

About lines 224-228, now it reads (the changes are in bold):

“Since these $ER_{(HCOOH/CO)}$ from previous studies and the $EmR_{(HCOOH/CO)}$ from Sinha et al. (2003) agree with our $ER_{(HCOOH/CO)}$, and since HCOOH has a short lifetime, this may suggest that the selected plumes measured by IASI from 2008 to 2014 and those sampled by Vigouroux et al. (2012) and Coheur et al. (2007), encountered a limited secondary production or a low sink as deposition or reaction with OH in the troposphere during their transport. **To quantify the role of the chemistry or of the deposition within the plumes, a modeling work should be performed. This is however beyond the scope of this paper.**

Another important point is that the decay of HCOOH is faster than for CO. **As our $ER_{(HCOOH/CO)}$ is similar to the $ER_{(HCOOH/CO)}$ from the other studies and to the $EmR_{(HCOOH/CO)}$ given in Sinha et al. (2003), this could suggest that all these plumes (from our study, from Vigouroux et al. (2012) and Coheur et al. (2007)) are rapidly advected in the troposphere”.**

And line 237 – How would the impact of the difference in the geometry of sampling be accounted for in a proper comparison between ACE-FTS and IASI?

The sentences were confusing. The sentences have been changed as below:

“It is worth noting that the ACE-FTS instrument used in their study works in a limb solar occultation mode. This means that the atmospheric density sampled by the instrument is larger than the one measured by the nadir geometry with IASI. However, the difference in geometry cannot explain why we find an agreement with the ACE-FTS measurements values reported by Coheur et al. (2007) and a disagreement with those from Rinsland et al. (2006). Part of the difference could be associated with the difference in the assumptions used in both retrievals (e.g. the a priori).”

Line 239 – Where were the plumes sampled by Yokelson et al.?

The plumes were over Zambia, Zimbabwe and South Africa. This information is now included in the sentence (in bold):

“The $ER_{(HCOOH/CO)}$ from our work is also 15% lower than the $EmR_{(HCOOH/CO)}$ in Yokelson et al. (2003) ($5.9 \times 10^{-3} \pm 2.2 \times 10^{-3}$ mol/mol) **who calculated their value within plumes over Zambia, Zimbabwe and South Africa.**”

Page 7, lines 243-244 – What was the approach developed by Chaliyakunnel et al. (2016) to determine pyrogenic ER(HCOOH/CO)? It is not clear what is meant “by reducing the impact of the mix with the ambient air”.

An explanation of their approach is now added (in bold):

“To do so, they calculated the ER_(HCOOH/CO) in the vicinity of fire count from MODIS (averaged in a cell having the resolution of the GEOS-Chem model, i.e. 2°× 2.5°) and they differentiated this value with a background ER_(HCOOH/CO) defined by the concentrations distant from these fires. They concluded that their most reliable value on the amount of HCOOH produced from fire emissions was obtained for African fires.”

Page 7, lines 269-271 – Revise this poorly written paragraph. It is not clear what is meant by either sentence.

As explained as introduction of our answers, section 5.2 was rewritten and the title of section has been changed.

To answer the question about lines 269-271, the new lines are (the changes are in bold):

“5.2. Analysis based on the type of vegetation

We have complemented our comparison of the enhancement ratios **by comparing our ratios to emissions ratios calculated** from emission factors found in literature. The main argument to perform such comparison is the lack of measurements of enhancement ratios over IND and SEA. **Furthermore, such comparison from emission factors facilitates an analysis based on hypothesis about the type of vegetation burned.”**

Page 7, lines 275-279 – Why can't the decay be taken into account by considering the exponential decrease between emission and detection using relative lifetimes, e.g., Viatte et al. (2015) and references therein?

Each MODIS hotspot is characterized by a mean CO total column and a mean HCOOH total column. These averages are calculated along 5 hours. During 5 hours, the chemistry may already occur and it is the reason why we have written that the decay of these compounds could not be taken into account in our methodology.

Moreover, without to know the accurate altitude of the plumes, it is challenging to calculate the age of the air masses.

Sections 5.1 and 5.2 – Both sections discuss enhancement ratios and emission ratios, including comparisons with other studies, e.g., on page 8, there is additional discussion of ER although the title suggests that Section 5.2 is about EmR. These sections could be more clearly differentiated.

Both sections have been changed.

Now there are:

- 5. Analysis of the data over the fire regions
 - 5.1. Determination of the enhancement ratios
 - 5.1.1 General analysis
 - 5.1.2 Analysis over each region
 - 5.2. Analysis based on the type of vegetation

Page 9, lines 358-359 – Arguably, such an intercomparison could have been included in this study.

An inter-comparison has to be done but it is beyond the scope of this paper. It will be a subject for a next study. It is the reason why it was mentioned in the conclusion.

Technical Corrections

Page 1, line 19 – add comma after “(MODIS)”

Done

Page 1, line 26 – add comma after “forest fires”

The sentence has been deleted.

Page 1, line 34 – delete “for”

Done

Page 2, line 46 – Rewrite this sentence. Not clear what is meant by “as on the oxidizing power...”

A complementary information is now provided. The changes are shown in bold:

“... on the oxidizing **capacity** of the atmosphere (**i.e. the chemistry of OH in cloud water - Jacob, 1986; the heterogeneous oxidation of organic aerosols - Paulot et al., 2011**)”

and we have added this reference:

Jacob, D.: Chemistry of OH in remote clouds and its role in the production of formic acid and peroxymonosulfate, *J. Geophys. Res.*, 91, 9807–9826, 1986.

Page 2, line 55 – “hence depend on”

Corrected

Page 2, line 67 – change “as with” to “including” or “such as”

“Including” is now used.

Page 2, line 69 – delete “with the”

Done

Page 2, line 70 – “Atmospheric Chemistry Experiment – Fourier Transform Spectrometer (ACE-FTS)”

Changed

Page 2, line 72 – I think this means “(MIPAS) limb instrument, which is sensitive to altitudes down to ~ 10 km” (rather than only sensitive at 10 km)

Grutter et al. (2010) – the cited reference – shows distributions and time-series at 10 km. Most of their profiles start at 8 km, and thus we kept the sentence:

“... Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) limb instrument which is sensitive to around 10 km (Grutter et al., 2010).”

Page 2, line 74 – “compared to ground-based and airborne”

Corrected.

Page 2, line 75 – “allows observation of remote regions”

“which allows observing remote regions” is now changed by “allows observation of remote regions”.

Page 2, line 77 – “ratios of HCOOH relative to CO over”

Changed.

This change has also been done in the title of the tables.

Page 3, lines 93-94 – add space before K, as done for other units like km, cm⁻¹, etc.
Done

Page 3, line 97 – Isn't the lifetime of CO closer to two months than several weeks?
The lifetime depends on the season and on the location. We clarified this point by changing “several weeks” by “a few weeks to a few months depending on latitude and time of year.”

Page 3, line 113 – “in more detail”
Corrected

Page 3, lines 117-118 – “which is less than 35% for total columns smaller than...”
Changed

Page 4, line 123 – “hotspots”
“s” has been added.

Page 4, line 123 – MODIS has already been defined
That is correct. Thank you for noticing it.

Page 4, line 129 – “which, for each detected fire pixel, includes the ...”
Changed.

Page 4, line 132 – Last sentence doesn't need to be a separate paragraph.
Changed.

Page 4, line 141 – “most active in terms of actual fires but are still of interest. The first ...”
These four sentences about importance of biomass burning in India and Siberia could also be rewritten for clarity.

The sentence has been changed as:
“Among these regions, India and Siberia do not represent the most active regions in terms of number of fires. It seemed however important to also investigate them.”

Page 4, line 144 – “over some years, such as during summer 2010”
Changed

Page 4, line 154 – “(correlation coefficient, r , from”
Changed

Page 4, line 155 – “the impact of sources other than biomass burning”
Changed.

Page 4, line 156 – “also have”
Changed

Page 4, line 160 – “The large region selected over Siberia”
Changed

Page 4, line 161 – “other regions, such as polluted”

Changed

Page 5, line 170 – add comma after “criteria”

Done

Page 5, line 171 – “in Table 1. The smaller correlation coefficients, i.e., less than 0.7, are found”

Changed

Page 5, line 172 – “the HCOOH and CO columns”

Changed

Page 5, line 178 – “assign” rather than “attribute” ?

Changed

Page 5, line 179 – ECMWF has already been defined

It was not defined previously, except in the abstract. Thus we have decided to keep the definition in this line.

Page 5, line 182 – “(r close to 0.8)”

“r” has been added.

Page 5, line 183 – Clarify that the low mean and median refer to surface wind speed. Also rewrite the sentence on line 184 for clarity.

The sentence has been changed as:

“IND has also a low mean and median **surface wind speed** but the distribution of **this** surface wind speed **over IND** is more spread out than for **AMA, SEA and SAF.**”

Page 5, line 186 and elsewhere through the manuscript– “in Table 2” ? Does ACP accept Tab. as an abbreviation for Table?

“Tab” has been changed by “Table” everywhere through the manuscript.

Page 5, line 197 – “than using only the columns”

Changed

Page 5, line 198 – “for each measurement pair”

Corrected

Page 6, line 201 – “so comparison with previous work is ... over another”

Corrected.

Page 6, line 203 – should globally be generally?

“globally” was changed by “generally”.

Page 6, line 206 – “The effects of both biases are, however, limited”

The sentence has been changed as requested.

Page 6, line 211 – “an improved [or a lower?] detection limit”

That’s correct; in this case, improved means lower. To clarify it, we have changed “improve” by “reduce”. Now it reads:

“Indeed, the increase in the thermal contrast (i.e. the temperature difference between the surface and the first layer in the retrieved profile) leads to **reducing** the detection limit as shown in Pommier al. (2016).”

Page 6, line 222 – “same plume as”
Corrected

Page 6, line 231 – trajectories
Corrected

Page 6, line 235 – “reasons for the agreement”
The sentence has been changed:
“However, the difference in geometry cannot explain why we find an agreement with the ACE-FTS measurements values reported by Coheur et al. (2007) and a disagreement with those from Rinsland et al. (2006).”

Page 6, lines 241-242 – “Conversely, the ... from IASI is twice that of Chaliyakunnel”
Changed. The sentence is now:
“**Conversely**, the $ER_{(\text{HCOOH}/\text{CO})}$ retrieved **from IASI is twice that of** Chaliyakunnel et al. (2016) ($2.6 \times 10^{-3} \pm 0.3 \times 10^{-3}$ mol/mol).”

Page 7, line 247 – No need for a new paragraph here.
We have preferred to keep this new paragraph since it corresponds to the analysis of the results over NAF and the previous paragraphs are about SAF.

Page 7, line 248 – “worth noting”
“Reminding” is replaced by “noting” as requested.

Page 7, line 251 – “and that of Paton-Walsh (2005) may be explained”
Changed. The sentence is now: “The difference between our work and **that of Paton-Walsh (2005) may be explained** by the different origin of the probed plume.”

Page 7, line 254 – quantify “quite uncertain”
The value is now given in text:
“... a quite uncertain value is reported ($4.5 \times 10^{-3} \pm 5.1 \times 10^{-3}$ mol/mol),...”

Page 7, line 280 – “For both the IND”
The comma has been deleted as requested.

Page 8, line 287 – Equation
Changed

Page 8, line 289 – “composed of tropical”
Page 8, line 292 – “composed of cropland”
Both were corrected.

Page 8, line 293 – “characterized by an”
Corrected.

Page 8, line 300 – “(2004) both used the same”

Changed.

Page 8, line 307 – “twice the value” [also specify whether ER or EmR from Akagi]
We agreed that this sentence was confusing. We changed “value” by “EmR”. Now it reads:
“Over Northern Africa, our $ER_{(HCOOH/CO)}$ **is twice as large as** the **EmR** $_{(HCOOH/CO)}$ provided by Akagi et al. (2011), probably due to the lower correlation found in our scatterplot.”

Page 8, line 308 – “It is highly”
Changed.

Page 8, line 314 – “forest fire plumes”
Changed.

Page 9, line 336 – “difficulties ... are”
Corrected

Page 9, line 338 – “using satellite, airborne, or FTIR measurements”
Changed

Page 9, line 346 – “A very good agreement was found” in what? Specify.
The information has been added (in bold):
“A very good agreement **in** $ER_{(HCOOH/CO)}$ was found over Amazonia, especially with the work done by Chaliyakunnel et al. (2016) who determined pyrogenic $ER_{(HCOOH/CO)}$.”

Page 9, line 349 – Replace “delicate” with a better description.
We replaced “delicate” by “complicated”:
“The analysis over Australia is however **complicated** as our $ER_{(HCOOH/CO)}$ approximately corresponds...”

Page 9, line 355 – “a modelling study could be”
“work” has been replaced by “study”.

Page 9, line 357 – times
“s” has been added.

Page 9, line 358 – “instruments such as”
“Such” has been added.

Page 10, line 367 – Isn’t IASI an instrument, not a mission?
There are 3 similar instruments. So IASI is both an instrument and a long term mission.

Page 10, line 372 – “for free access”
Corrected

Page 10, lines 385 and 387 – Inconsistent formatting of references for the same journal.
Thanks for this observation. The doi was missing for the first reference. Now the references are:
Andrews, D. U., Heazlewood, B. R., Maccarone, A. T., Conroy, T., Payne, R. J., Jordan, M. J. T., and Kable, S. H.: Photo-tautomerization of acetaldehyde to vinyl alcohol: a potential route to tropospheric acids, Science, 337, 1203–1206, doi:10.1126/science.1220712, 2012.

Beirle, S., K. F. Boersma, U. Platt, M. G. Lawrence, and T. Wagner : Megacity emissions and lifetimes of nitrogen oxides probed from space, *Science*, 333, 1737–1739, doi:10.1126/science.1207824, 2011.

Page 17, caption line 2 – “over the 7 [seven] regions studied. IASI data are”
7 has been replaced by “seven”.

Page 17 – Tab. or Table ?

As for your previous comment, “Tab” has been changed by “Table” everywhere through the manuscript.

Pages 18 and 19, table headings – “HCOOH/CO Enhancement/Emission Ratio ...” would be a better title

The table headings are:

Table 2. Enhancement ratio of HCOOH relative to CO (mol/mol) with its standard deviation compared to enhancement ratios of HCOOH relative to CO and emissions ratios of HCOOH reported in the literature for the seven studied regions.

Table 3. Enhancement ratio of HCOOH relative to CO (mol/mol) with its standard deviation and enhancement ratio of HCOOH relative to CO (mol/mol) by biome with its standard deviation calculated in this work. For each enhancement ratio by biome, the correlation coefficient and the number of MODIS hotspots are provided. The enhancement ratios are compared to emission ratios calculated from emission factors given in the literature for the seven studied regions. For the calculation of these emission ratios, the emission factors of CO for the corresponding fuel type given in Akagi et al. (2011) are used. Emission ratios of HCOOH relative to CO (mol/mol) calculated from the emission factors of HCOOH given in Akagi et al. (2011) for the corresponding fuel type are also provided.

Page 18, Table 3 – Left justify all the table entries

Done

Page 19, caption line 2 – “in the literature”. Also, rewrite the full caption for conciseness and clarity, e.g., HCOOH/CO enhancement ratio, etc.

See our answer about the table headings.

Page 20, caption line 3 – “column distribution ... column distribution”

Corrected

Page 21, Figure 2 and page 22, Figure 4– Preferable to have units on the y-axis labels, rather than just in the caption.

Figs 2 and 4 now include units, as hereafter:

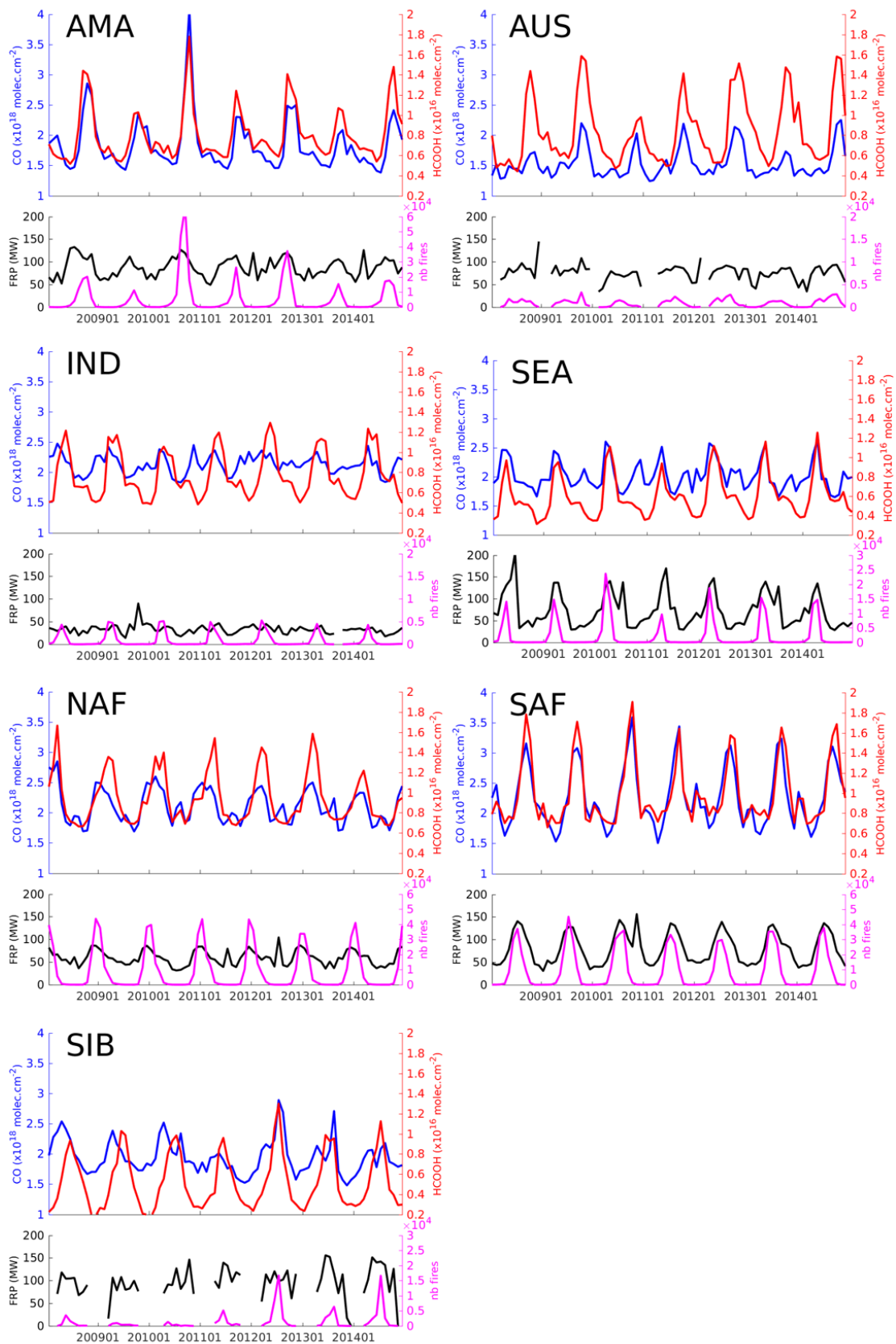


Figure 2: Time-series from 2008 to 2014 of the monthly means of IASI CO (blue) and HCOOH (red) total columns in 10^{18} molec./ cm^2 and in 10^{16} molec./ cm^2 , respectively, FRP (black) in MegaWatts and the number of fires (magenta) from MODIS over the seven regions (AMA=Amazonia, AUS=Australia, IND = India, SEA = Southern East Asia, NAF= Northern Africa, SAF= Southern Africa, SIB= Siberia).

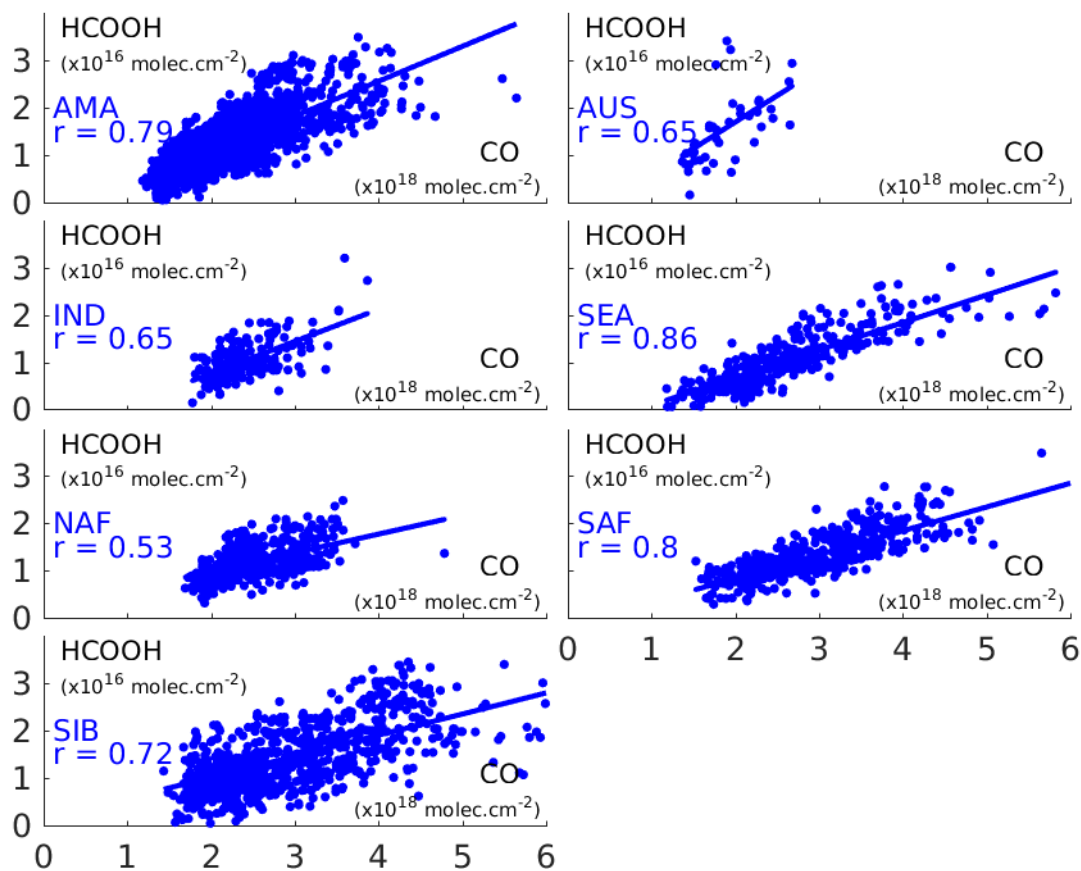


Figure 4: Scatterplots between the IASI fire-affected HCOOH total columns (in 10^{16} molec./cm 2) and the CO total columns (in 10^{18} molec./cm 2) over the seven regions (AMA=Amazonia, AUS=Australia, IND = India, SEA = Southern East Asia, NAF= Northern Africa, SAF= Southern Africa, SIB= Siberia).The linear regression is represented by the blue line and the correlation coefficient is also provided for each region.

Page 22, Figure 3, caption line 4 – Clarify text describing the percentiles

The sentences “The whiskers encompass values from 25th-1.5×(75th-25th) to the 75th+1.5×(75th-25th). This range covers more than 99% of a normally distributed data set.” Have been changed by:

“The whiskers encompass values from 25th-1.5×(75th-25th) to the 75th+1.5×(75th-25th). This range of values corresponds to approximately 99.3% coverage if the data are normally distributed.”