

The authors would like to thank the reviewer for their helpful comments and suggestions. Please find below a discussion addressing the general comments followed by individual responses to the specific comments.

General comments:

My major issue with this paper is with the discussion (or lack thereof) of potential biases. Many of the conclusions of this work rely on the quality the absolute values of the emission factors calculated - for example, they are directly compared to emission factors derived from in situ measurements. The emission factors measured in this work are derived from OMI NO₂ observations, the TM5 model, and GFED3 dry matter consumption estimates. However, there is limited consideration of how known or potential biases in these three elements (OMI, TM5, GFED3) might impact the final values. Furthermore, all three have been shown to have substantial biases (or potential biases) at the relevant spatial scales. Modeled NO₂ columns are subject to bias due to their low resolution in conjunction with nonlinear NO_x chemistry (e.g. Valin et al., 2011). The instantaneous dilution of NO_x throughout a large grid cell can cause the NO_x lifetime to be under- or overestimated (depending on the concentration), which could result in a bias in β (a value used in this work and derived from TM5). Such a bias could have very important implications, but only minimal, non-quantitative discussions of inaccuracy in the model chemistry are included. It has also been shown that it is possible, even likely, that satellite NO₂ retrievals exhibit biases over fires (e.g. Bousseret, 2013; Mebust et al., 2011). Given the clearly inaccurate NO₂ a priori profiles in the case of NO₂ retrievals over actively burning fires along with heavy aerosol loading, the possible existence of a bias needs to be discussed, and yet this issue is not once mentioned in the paper. Finally, potential biases in GFED are mostly ignored, with the exception of an underestimation in mass burned from agricultural fires. I am less familiar with the potential biases in GFED but undoubtedly there are at least a few. I am particularly interested in the possibility that there are systematic problems with the way GFED defines fuel type, and whether the derived fuel types in this work were compared to another land cover product or otherwise verified. Overall, the paper would strongly benefit from a thorough discussion of biases that might impact the derived emission factors (and relevant associated literature). Following on that discussion, new analyses should be added to the manuscript to show how the biases do or do not affect the conclusions.

We will add to the manuscript a synopsis of the following analysis and discussion of possible biases in the observations and chemical transport model, and their effect on our general conclusions.

1. TM5 Chemistry:

Measurements of the components of NO_y for the Amazon are rare, therefore validation of model nitrogen chemistry remains elusive for this region. However, recent model improvement and sensitivity studies give some insight into possible biases in the TM5 v3 simulations of tropospheric NO₂.

In Williams et al. (2012) the authors implemented an on-line photolysis algorithm in TM5 v3 and found that a revised JNO₂ value increased NO₂ concentrations above 800 hpa, adding approximately 0.2×10^{15} molecules/cm² to the column over South America. Williams et al. (2012) also estimated that OH concentrations near the surface should be increased by 15%, implying that the conversion of NO₂ to HNO₃ via reaction with OH is biased low and the lifetime of NO_x is overestimated in TM5 v3.

However, recent laboratory (Mollner et al., 2010) and aircraft (Henderson et al., 2012) observations indicate that the Sander et al. (2006) rate constant for this reaction, widely used in global chemical transport models, could be biased high by approximately 15-20%. Therefore, in the current configuration of TM5 v3, these two biases would largely offset each other, leaving a 5-10% high bias in the loss of NO₂ to HNO₃, the predominant chemical sink of NO₂, and therefore a high bias in β .

Because HO₂ loss to aerosols, a very uncertain but potentially important HO_x sink, is not included in TM5 v3 the low bias in β for grid cells affected by biomass burning is likely greater than 5-10%. We therefore make an estimate based on the discussion above and the sensitivity simulations by Stavrou et al. (2013) of increasing the reaction probability of HO₂ uptake on aerosols to 1, that β is overestimated by 25% and that NO₂ tropospheric columns are biased low in TM5 v3 by 0.2×10^{15} molecules/cm² + 0-20%. This estimate is conservative as HO₂ uptake on aerosols equal to 1 is an upper limit for metal-doped aerosols.

From the August and September monthly means shown in Figure 1, it is apparent that along the arc of deforestation TM5 v3 with GFED v3 emissions overestimates NO₂ columns, while background concentrations are underestimated. This is consistent with our findings that NO_x emission factors for deforestation burning are on average too high as well as the indications that the model NO₂ lifetime may be too short.

2. Model Resolution and Sampling Errors:

If the resolution of a NO_x emission source is significantly smaller than the resolution of the chemistry transport model, biases can occur in the estimate of NO_x emissions from model simulations because emissions from the point source will be artificially diluted to the coarse resolution model grid cell (Valin et al., 2011; Vinken et al., 2011). This dilution effect leads to errors in model NO₂ concentration and NO_x lifetime.

This issue is most relevant for the estimate of NO_x emissions from fires in July, when burning is more spatially heterogeneous, and takes place in relatively pristine regions. In August and September, hundreds of active fires occur within a 1°x1° grid cell, thus mimicking a spatially homogeneous source at the resolution of the model. This is a consequence of the limited time period during which ambient conditions favor burning in South America, and the concentration of

agricultural development along the forest boundary.

At the beginning of the dry season in July, background NO₂ concentrations are low. Thus, an increase in the NO_x concentration will decrease the NO₂ lifetime. The instant dilution of the emissions will dampen this effect leading to an overestimation of the NO₂ concentration in the model and an underestimation of β . For 1°x1°, Valin et al. (2011) estimate the bias in the column is on the order of 25%. Although adding to the uncertainty in the value of the fire NO_x emission factor for July, the overestimation of the model NO₂ concentration and lifetime implies that for our analysis the increment in NO_x emissions needed to resolve the model-observation mismatch should be larger and supports our findings that for woodland, and less significantly for savanna burning, NO_x emission factors decrease from July to August.

3. NO₂ Tropospheric Column Retrievals:

Incorrect assumptions about the NO₂ profile shape can lead to biases in the OMI air mass factor (AMF) used to convert tropospheric slant column densities to vertical column densities. The coarse resolution of the TM4 model that provides the a priori NO₂ profile for the DOMINO retrieval may underestimate the surface level concentrations. Recent work by Bousserez (2013) shows the effect on the air mass factor of assuming an a priori NO₂ profile without fire emissions. They find that AMFs would be biased low by as much as 50%, but on average 20-30%. This of course represents the upper limit for this bias, as the TM4 model a priori profiles do account for fire emissions. Therefore, a reasonable estimate for this bias is 10-15% on average.

High aerosol loadings may also contribute to biases in NO₂ tropospheric columns retrieved by DOMINO (Leitão et al., 2010). If aerosols are well mixed with the NO₂ plume, enhanced light scattering will increase the sensitivity to NO₂ in this layer. AMFs would be underestimated and NO₂ tropospheric columns would be overestimated if this effect were not taken into account. If aerosols exist as a layer above the NO₂ plume, they effectively shield the lower atmosphere, reducing the sensitivity to NO₂. Ignoring this effect would overestimate the AMFs and underestimate NO₂ tropospheric columns.

Observations from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument that has an overpass time within 15 minutes of OMI can provide some information regarding biomass burning aerosol layer heights over South America. However, CALOP's narrow 335 m footprint does not allow for extrapolation of daily observations to biome scales. Torres et al. (2013) developed a climatology of aerosol layer heights from 2.5 years of CALOP observations, and found that they ranged from 2-4 km during the South American burning season. Individual orbital tracks show spatial heterogeneity of elevated and vertically well-mixed aerosol.

If we assume that a homogeneous elevated aerosol layer develops over the region

shielding the surface emissions, OMI-DOMINO v2 NO₂ tropospheric columns could be underestimated by 50% (Lin et al., 2013). This number represents an upper limit, and is probably not representative of the typical aerosol induced retrieval error as some aerosol correction occurs via increased cloud fractions in the DOMINO retrieval (Boersma et al., 2011).

4. GFED v3 Dry Matter Consumption Estimates:

Known issues of underestimated burned area from increased cloud cover and small fires, particularly for agricultural burning, were addressed several times in the manuscript. Considering the comparison of modeled CO total columns to MOPPITT observations minimized biases in GFED v3 dry matter consumption. The analysis showed that simulation errors were within the uncertainty of the satellite observations for the July through September time period considered in this study. The remaining two months of the burning season were not considered because of known low biases. Therefore, we are confident that the GFED v3 fuel consumption estimates aggregated to the monthly biome scale are not significantly biased, but have roughly 20% uncertainty.

A detailed description of how fuel consumption is assigned a biome or burning type is given in van der Werf et al. (2010). Briefly, the 0.5° x 0.5° burned area dataset developed by Giglio et al. (2010) uses 500 m burned area estimates in combination with the 2001 1 km MOD12Q1 land cover map with the UMD classification scheme and MOD44 vegetation continuous field (VCF; fraction tree, herbaceous, and bare cover) to calculate the fraction of burned area in each 0.5° grid cell from different land cover classes and fraction tree cover bins. The fraction tree cover determines the area of herbaceous landscape and wooded landscape burned in each grid cell for a land cover class.

It is clear from the description in the manuscript of our analysis that the assigned biome/burning types in this work aggregate the sub-grid scale heterogeneity of the landscape in a 1°x1° TM5 grid cell. However, the high-resolution land cover maps drive the spatial patterns of biome/burning types. Comparison to other land cover products is not possible because the biome assignments made here are not based on the relative spatial extent of live biomass, but on the relative amount of combusted biomass. For example, grassland may make up the majority of the area of a grid cell, but the mass of woody fuel burned from a forest that covers only a small fraction of the grid cell could be an order of magnitude higher than the mass of herbaceous fuel burned in the grassland.

5. Analysis of Biases:

In Figure 1 below we show the estimated NO_x emission factors for the following scenarios:

- (1) Constant Background Emission: Assume that background NO_x emissions are accurate, and only fire emissions account for the model-observations bias – i.e. the analysis described in the manuscript.
- (2) Constant Background Emission + Bias: Assume that background NO_x

emissions are accurate, only fire emissions account for the model-observations bias, and:

- a. β should increase by 25% in August and September
 - i. 5-10% to account for biases in OH concentration (Williams et al., 2012)
 - ii. 10-15% to account for HO₂ loss to aerosols (Stavrakou et al., 2013)
 - b. β should increase by 50% in July
 - i. 5-10% to account for biases in OH concentration (Williams et al., 2012)
 - ii. 10-15% to account for HO₂ loss to aerosols (Stavrakou et al., 2013)
 - iii. 25% to account for resolution effects (Valin et al, 2011)
 - c. TM5 NO₂ columns should increase by 0.2×10^{15} molecules/cm² + 20%
 - i. 0.2×10^{15} molecules/cm² to account for offline photolysis (Williams et al., 2012)
 - ii. 20% to account for underestimated lifetime (Mollner et al., 2010; Williams et al, 2012; Stavrakou et al., 2013)
 - d. OMI NO₂ columns should increase by 60%
 - i. 50% for aerosol shielding (Lin et al., 2013)
 - ii. 10% for a priori profile error (Bousserez, 2013)
- (3) Background Fraction Constant: The fraction of fire emissions to background emissions is accurate.
- (4) Background Fraction Constant + Bias: The fraction of fire emissions to background emissions is accurate, and the bias assumptions from (2) are included.

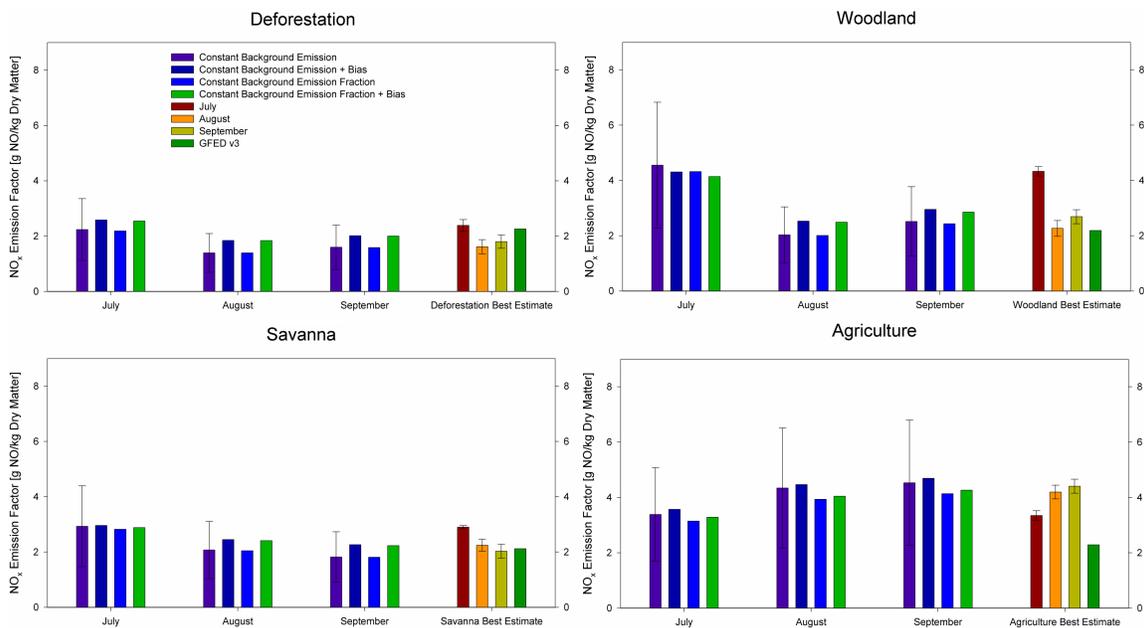


Figure 1. NO_x emission factor estimates considering two different assumptions for (a) the background NO_x emission contribution to the NO_2 tropospheric column, and (b) biases in model and observed NO_2 column. Error bars for the ‘Constant Background Emission’ case represent the 50% combined uncertainties of the modeled NO_2 columns, observed NO_2 column, and GFED estimated dry matter emissions. Error bars on the best estimate NO_x emission factors represent the standard deviation of the estimates from the four scenarios.

The four NO_x emissions factors calculated with different underlying assumptions for background NO_x emissions and biases in model and observed NO_2 tropospheric columns are well within the estimated margin of uncertainty of our original calculations. From the spread of the NO_x emission factors calculated from the four scenarios we can estimate that underlying biases in the chemical transport model and observations contribute at most 10% uncertainty to our estimates. The mean of the 4 scenarios can be considered a best estimate, and these differ by 10% or less from our original calculations.

While the current observation, chemical transport modeling, and bottom up emissions estimates may have substantial uncertainty, which makes calculating a definitive emission factor impossible, globally averaged fire emission factors derived from in situ observations likewise have large uncertainties. While these results do not close the spread in observed emission factor values, it does show that an analysis of a large number of fires aggregated over time and space yields NO_x emissions factors that are generally consistent with point observations of a few fires.

On another note, this paper has a pretty limited scope in that it covers a single year and South America only. The authors do a good job of setting up the basis for a South America-focused analysis (starting P22761 L14), but I don't feel as though there is a good sense of why the analysis is restricted to a single year, especially since the requisite datasets are available over several years. The authors do mention that 2005 is a drought year (P22764 L6), but that alone does not justify using only a single year of data - in fact it makes it impossible to determine if the results shown here are generally true or are drought specific phenomena. I recognize that to do this analysis for additional year(s) would be a substantial amount of additional work, but I do think there needs to be a clearer, stronger argument as to why this would be outside the scope of this particular paper and how the reader should view the generality of the results presented herein.

Interannual variability will be a part of a separate follow-up analysis. This will require consideration of changes in ambient conditions, fuel load, as well as political and economic incentives. Therefore, we have chosen to focus on differences between biomes for one region in a year in which significant deforestation burning occurred and OMI had the best spatial coverage.

Specific comments:

P22761 L8: This is not how I understood the conclusions of McMeeking et al. (2009). The molar ratio NO_x/NH_3 (intended to account for fuel nitrogen) was loosely linearly fit with MCE, but with substantial variability and inconsistent in slope with previous work that examined NO_x/NH_3 ratios as a function of MCE. That certainly does not indicate that fuel nitrogen and MCE together explain all of the variability in NO_x emission factors.

In McMeeking et al. (2009) the authors presented NH_3 and NO_x emission ratios as well as MCE measured during laboratory burns. In Figure 10, there is indeed scatter in the regression of NH_3/NO_x vs MCE. However, the authors state: “Most of the samples that deviated from the linear fit corresponded to burns with low NO_x emissions and high uncertainties in the calculated NH_3/NO_x molar ratios.”

In addressing the comparison of the slope of NH_3/NO_x vs MCE to the previous work of Goode et al. (2000) who also measured emissions ratios in laboratory burns, the authors state: “Goode et al. [2000] treated all NO_x emissions as NO because NO_2 mixing ratios were below their instrument’s detection limits. The high- NO_x FLAME data agreed with the Goode et al. [2000] fit if $\text{NH}_3:\text{NO}$ molar ratios are considered.”

The studies that were inconsistent with the results presented in McMeeking et al. (2009) used open-path and aircraft instrumentation to sample burning in the field. Discrepancies in instrumentation and experimental design (i.e. sampling differences, field vs laboratory burning) must be considered before comparing the regressions directly. Taken individually, the results of each experiment show that the NH_3/NO_x emission ratio typically increases linearly with MCE.

P22761 L12: How does this MCE compare to typical fire MCEs? Are typical MCEs variable with fuel type? I know this comes up later but it is strange to bring up a number without providing any context for how it compares to a normal fire.

The mention of MCE here is to inform the reader on the value of MCE above which flaming combustion and therefore higher NO_x emission factors occur. Typical values of MCE for different biomes can be found in Table 1, which the reader is referred to at several points later in the manuscript.

P22767 L11: I am concerned about the assumption that CO is an appropriate proxy for dry matter burned. First, I would disagree that the CO emission factor is constant to within 20%—there are ample observations (e.g. Yokelson et al. 2008) that indicate that CO emission factors (even specifically for “tropical” fires) can vary by a factor of 2 depending on the fire and the type of combustion. And even savanna fires vs. tropical forest fires (both types of fires analyzed in this work) have very different (i.e. >20%) CO emission factors in Akagi et al. (2011), which the authors cite as indicating variability is less than 20%. Further, in the discussion of observations of seasonal variability in NO_x emissions factors the authors suggest that MCE effects explain some of the variability, but this would imply some amount of seasonal variability in CO emissions too as they are also dependent on MCE. So I would like to see some discussion of this effect. If MCE

decreases across the season (one of the authors' propositions) then the CO emission factor would increase, which might help explain why TM5 under-predicts observed CO in October (and to a lesser extent, September).

The large spread in CO emission factors reported in Yokelson et al. (2008) Table 4 occurs between aircraft, ground based, and laboratory observations. The large variability in MCE presented in Table 3 of Yokelson et al. (2008) are for laboratory fires, which are not representative of actual burning conditions.

In Table 2 of Yokelson et al. (2007), the standard deviation of CO emission factors from 9 aircraft observed fires was 24%. In Akagi et al. (2011), the estimated natural variability of each emission factor is given in parenthesis in Table 1. The estimated variability for CO emission factors for tropical forest and savanna burning are 29% and 27%, respectively. These values are based on considering results from 10 measurement campaigns – not all of which are specific to South America. In van Leeuwen and van der Werf (2011) an analysis of 6 CO emission factor scenarios was carried out. The scenarios represented the predicted variability in CO emission factors due to the effect of seasonal, interannual, and spatial variability in combustion efficiency driven by ambient conditions. These sensitivity studies indicated that for South America, the average CO emission factor seasonal variability was approximately 10%, while the spatial variability is 0-30% with the highest variability occurring in forested areas. While individual fires may have large differences in CO emission factors, at the biome level (relevant for this work) an estimate of 20% spatiotemporal variability for CO emission factors is reasonable.

According to MOPITT observations, an 80-100% increase in total CO fire emissions would be required to resolve the CO concentration discrepancy in October. While CO emission factor variability may explain a fraction of the October CO underestimate, using active fire observations Randerson et al. (2012) show that in October increased cloud cover during this time period leads to a significant underestimation in burned area.

P22767 L22: How is the model being temporally sampled? I'm assuming it's not a daily average - but is it a snapshot in time (from the timestep following the OMI overpass) or an average over some time period? Given the possibility for temporal differences in the OMI overpass (for example, two different swaths), does this mean that at times the model output is coming from different model times in different grid cells? How does this sampling strategy work with the 3-hr resolution of the GFED emissions? Why not take a 3 hour window of the model corresponding to the GFED emission resolution that overlaps with the OMI observations?

The hourly model output is sampled before and after the overpass time of OMI (approximately 13:30 local time) and averaged. This window also corresponds to the middle of the peak in fire emissions. The resolution of GFED and ECMWF is 3-hourly, but the TM5 chemistry time step is shorter and dynamic. While the approximation of constant fire emissions during the 3-hour time step will introduce uncertainty, it is random and second order compared to the combined uncertainty in

model chemistry, observed NO₂ column, and estimated dry matter combusted.

P22768 Paragraph 2: I'm not sure I get the argument here. The way I read the written summary of the other paper, it sounds like the model matches the observations, and when the fire emissions in the model are doubled, the column doubles - I don't understand why that indicates that the chemistry and transport is "reasonable". All it says is that increasing NO₂ emissions in the model increases the column at roughly the same rate - to say it in terms of the method described in the next section, β (over the whole region) is approximately 1. To me, that doesn't say anything about whether the chemistry is correct. (Here might be a good place to talk about potential chemistry biases.)

Please see the discussion on Page 1.

P22769 L1: This essentially describes why the possibility of NO₂ lifetime biases in the model needs to be discussed. If the lifetime is incorrect β will be incorrect.

Please see the discussion on Page 1.

P22769 L3: I would clarify that EOMI represents the fire emissions (not all emissions) of NO₂ derived from OMI, and that it is calculated assuming that the entire discrepancy between OMI and TM5 is from inaccurate fire emission factors and none of the discrepancy is from inaccurate biogenic/anthropogenic/lightning emissions or errors in GFED mass burned. (The authors might also want to discuss this assumption, whether it is valid, and how it affects the results.)

Please see the discussion on Page 1.

P22770 L20: Please include the number or percentage of cells that changed to agricultural fires after using this filter.

The number of grid cells for the 3 months increases from 51 to 144.

P22770 L24: Please provide a number after "few and sporadic".

The number of grid cells that were forest fire dominated for July, August, and September were 16, 19, and 29, respectively.

P22771 L16: This is under normal NO₂ circumstances, not for heavily fire-influenced pixels. Also, how do the typical column densities in this analysis compare to this value (2×10^{15})? Keep in mind that in a 1x1 degree grid cell at this column density, approximately 50% of OMI pixels in that grid cell are higher than that.

Boersma et al. (2011) estimated that each individual DOMINO retrieval has an uncertainty of 1.0×10^{15} molecules/cm² + 25%. Thus for higher NO₂ column concentrations, the total relative uncertainty is lower than the conservative 75% we have given. As can be seen from Figure 1, the typical observed NO₂ tropospheric

column density in areas with high fire activity is $2-3 \times 10^{15}$ molecules/cm².

P22772 and on (Section 5): The authors make a lot of comparisons between different emission factors in this section, and it gets fairly confusing. It would be better to avoid putting the specific numbers directly into the text (since they are all in Table 1) and instead refer to the table and state the major point - e.g. “this value falls into the range of previous measurements and is slightly larger than the mean”. Also, I’m a little concerned about the discussion of seasonal differences. The figure is not that compelling - it’s not clear that the observed differences are statistically significant given the large error bars, and it looks like the entire “seasonal” difference is due to much higher values in July which suggests that there could simply be some substantial bias in that month. The authors should discuss the uncertainties and provide some sort of statistical assessment of how confident the monthly differences are.

See the above discussion regarding model and observation biases.

A statistical assessment of the monthly differences could be misleading, as there is significant uncertainty in the uncertainty estimates themselves. However, our analysis shows that by incorporating the OMI derived monthly variability in emission factors, systematic errors in the simulated NO₂ tropospheric columns are reduced (see Figure 2 below).

P22773 L11: The findings might support this, but they do not prove it. There are other possible explanations, including fuel nitrogen effects, and these should be discussed or at least mentioned.

We are careful to write that our findings suggest a shift to lower MCE from burning more woody fuel, not that it is proof. An increased contribution from woody fuel and the effect on MCE is convolved with a shift to lower fuel nitrogen content, as the C/N ratio of wood is 50 times higher than leaf litter. We will mention this in the manuscript.

P22773 L20: Again, it is not proven that the MCE is exceptionally low in this August, although though the emission factor is. Clarify the uncertainty around this statement.

We acknowledge this in the manuscript and state on P22773 L24: “Analysis of multiple years of data is needed to confirm this.”

P22774 L7: I would not use the word “likely” given the several problems identified with the assessment of agricultural fires - including the identified low bias in GFED, the small number of analyzed cells which implies that the values are highly uncertain, and the fact that the agriculturally influenced cells likely are influenced by a larger fraction of anthropogenic and biogenic emissions and thus the authors’ assumption that the model correctly captures these other types of emissions induces more uncertainty. These factors should also be mentioned.

We will mention that elevated background concentrations in agricultural areas driven by anthropogenic and biogenic emissions is an additional source of uncertainty.

See the discussion for P22774 L11 following the next paragraph.

P22774 L11: I cannot see how this analysis could simultaneously suggest that agricultural emission factors should be doubled and GFED dry matter consumption should be also increased. The author's values for emission factors are based on GFED dry matter consumption so any low bias in GFED dry matter will automatically produce a higher emission factor. If GFED is biased low by 55% then the emission factor that was derived from that will be biased high in response.

The underestimation of agricultural fires reported in Randerson et al. (2012) is for regional total annual burned area and emissions. Thus, the agricultural fires analyzed in this study occurring in the peak of the fire season when burned area estimates are the most reliable are not necessarily biased low. We acknowledge that the results provided in this work for agricultural burning are more uncertain than for the other biomes, but feel it is important to highlight the potential air quality implications of this source and the need for further analysis.

P22774 L20: How does this analysis differ for SCIAMACHY vs. OMI? Specifically, how did you sample the model in time given that the SCIAMACHY overpass is at a different time of day?

The hourly model output is sampled at the overpass time of SCIAMACHY (approximately 10:00 local time).

P22776 L4: It is certainly possible that these discrepancies are due to within-biome variability in emission factors, but it is also possible (and I would argue likely at least in part) that they are due to sources of error or uncertainty in the analysis, including all those I've already mentioned or others (for example, uncertainties in scaling 3hr emissions).

Remaining scatter between model and observed NO₂ tropospheric columns can be expected, as other model errors will not be corrected. However, the simulations using revised emission factors results in unbiased and symmetrically distributed errors, indicating a decrease in underlying systematic errors (see Figure 2 below). This discussion will be added to the manuscript.

P22776 Section 6: Please review the conclusions to make sure that they reflect any changes made - for example, statements on agricultural emission factors in conjunction with GFED underestimates.

Change made.

P22788 (Figure 1): This figure strongly suggests the possibility for both model and observational biases. It looks like it is nearly uniformly true that in high NO₂ areas, the model overestimates the observations, and in low NO₂ areas the model underestimates the observations. Given this consistency, I would hesitate to call these process-based (i.e. differences in emission factors) without a thorough analysis of the effects of biases both in OMI and in the model itself.

[Please see the discussion on Page 1.](#)

P22791 (Figure 4): I don't understand - why does biomass burning in July only either stay exactly the same or increase by 30-40%? That discontinuity, to me, suggests some sort of computational error or similar problem.

[In July, the 3 relevant burning types are deforestation, savanna, and agricultural burning \(Figure 2\). From Table 1, you can see that the OMI derived EF is within 10% of the GFED v3 value, while for savanna and agricultural fires they are more than 30% higher than the GFED v3 value.](#)

P22793 (Figure 6): This figure isn't very compelling. I know that the RMSE decreases when using OMI derived emissions in many of the cases, but visually they look nearly identical (OMI derived vs. initial). Rather than the figure you could just include the RMSE values in a table.

[We have changed the figure to Figure 2 on the following page and will include the following discussion:](#)

[“Figure 6 shows the probability distribution of the model bias for observed daily NO₂ concentrations over grid cells where fire emissions dominate over NO_x emissions from other sectors. In general, for all fire types the RMSE as well as the mean and standard deviation of the bias decreased, but the largest changes occurred in deforestation dominated grid cells. Moreover, the skew towards positive bias is reduced for the simulations using OMI derived NO_x emission factors. That the bias is more symmetric indicates that underlying systematic errors in the simulation are reduced.”](#)

[“Although the spread in the bias decreases, the standard deviation of the bias is still larger than the detection limit of the observations. Continued significant errors in the simulation can be expected, as other model errors will not be corrected.”](#)

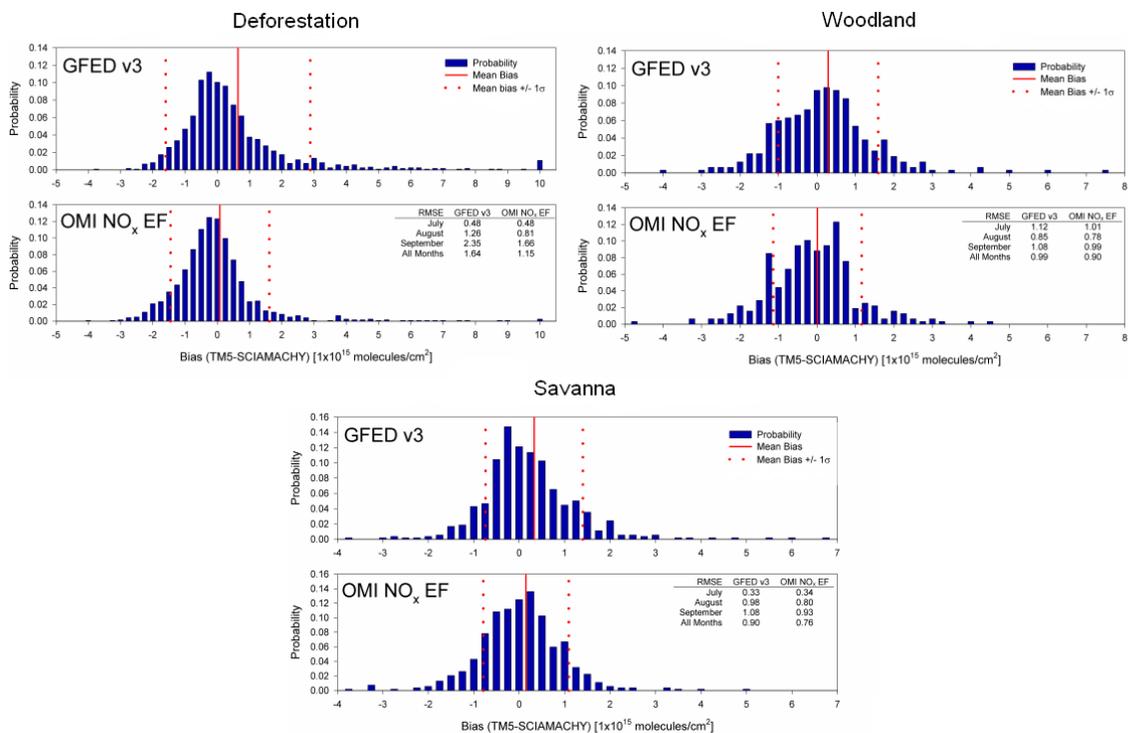


Figure 2. The probability distribution of the bias between TM5 and SCIAMACHY daily NO₂ tropospheric columns.

Technical/stylistic comments:

P22759 L4: I understand that deforestation and agricultural fires are important in this region and that is why fire is discussed as a “tool” but it seems incomplete to not at least mention wild fires in addition to intentional ones.

We will change the first sentence to mention the contribution from accidental burning.

“The spatiotemporal patterns of fire in the tropics are driven by human-triggered fires both intentional and accidental, as fire is a widely used tool to manage landscapes and clear land for new uses.”

P22760 Eq. 1: Notation in this equation is inconsistent – in one case B is superscripted and in the other it is contained in parentheses. Use a consistent notation. Also, the B and subsequent list of fire types needs to be further separated from the rest of the equation - it looks like it is a variable being multiplied.

The notation in Eq. 1 is intentionally different from the notation in Eq. 4. In Eq. 1, in each grid cell the total fire dry matter emission is the sum of the sub grid components from each biome. In Eq. 4, each grid cell has been assigned a dominant biome according to the description in Section 4.

P22760 L10: This sentence is awkward and could be reworded.

Change made.

“Including temporal variability in emission factors has not been possible because of the paucity in emission factor observations.”

P22760 L15: I would use “scaling” or “conversion” rather than “partitioning”.

Change made.

P22760 L20: The description of prompt NO_x is unnecessarily long. I would just say that “Laboratory studies indicate that emitted nitrogen-containing chemical species are accounted for by volatilized nitrogen from the fuel (refs)”.

The authors appreciate the helpful suggestion. However, we choose to keep the discussion of all known pathways of NO_x formation during combustion, particularly because there are few publications that give a complete overview of these processes for open burning of biomass.

P22761 L24 & P22764 L6: These two semicolons should be commas.

Change made.

P22762 through the end of the introduction: This section is really long and hard to read because it is so detailed. I understand the desire to explain fire characteristics for each type of fire, but there should be more focus on the details that are important to your later discussion and conclusions. I would suggest pulling out the specific values that are already included in your table (Table 1) unless there is a very compelling reason to include them (such as they are necessary to make a comparison point), and shortening the in-text discussion of each fire type as much as possible.

The authors value this feedback, but choose to keep the introduction as is. The current knowledge is scattered over many publications spanning over 20 years, and we would like to synthesize the current understanding of fire characteristics particular to South America. Many readers will likely not be aware of the heterogeneity of landscapes and the various roles of fire in this region.

P22762 L24: These 2 paragraphs on RSC interrupt your description of the four fire types and thus it seems out of place. It would be helpful to move this up somewhere to a section where you are talking about MCE.

The authors value this suggestion, but choose to keep the discussion of residual smoldering combustion after the description of deforestation burning, as this is the relevant biome and fire process for this type of combustion.

P22764 L13: You don't need this sentence - don't need to justify your choice to validate using an independent dataset.

We choose to keep this sentence, as we would like to signal to the reader that this analysis is to come.

REFERENCES:

- Akagi, S. K., Yokelson, R. J., C, W., Alarado, M. J., Reid, J. S., Karl, T., Crounse, J. D. and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, *Atmospheric Chemistry and Physics*, 11, 4039–4072, 2011.
- Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitão, J., Richter, A., Zhou, Y. and Brunner, D.: An improved tropospheric NO₂ column retrieval algorithm for the Ozone Monitoring Instrument, *Atmospheric Measurement Techniques*, 4, 1905–1928, doi:10.5194/amt-4-1905-2011, 2011.
- Bousserez, N.: Space-based retrieval of NO₂ over biomass burning regions: quantifying and reducing uncertainties, *Atmospheric Measurement Techniques Discussions*, 6, 6645–6684 [online] Available from: <http://www.atmos-meas-tech-discuss.net/6/6645/2013/amtd-6-6645-2013.pdf> (Accessed 14 December 2013), 2013.
- Giglio, L., Randerson, J. T., Werf, G. R., Kasibhatla, P. S., Collatz, G. J., Morton, D. C. and DeFries, R. S.: Assessing variability and long-term trends in burned area by merging multiple satellite fire products, *Biogeosciences*, 7, 1171–1186 [online] Available from: <http://www.biogeosciences.net/7/1171/2010/>, 2010.
- Goode, J. G., Yokelson, R. J., Ward, D. E., Susott, R. A., Babbitt, R. E., Davies, M. A. and Hao, W. M.: Measurements of excess O₃, CO₂, CO, CH₄, C₂H₄, C₂H₂, HCN, NO, NH₃, HCOOH, CH₃COOH, HCHO, and CH₃OH in 1997 Alaskan biomass burning plumes by airborne Fourier transform infrared spectroscopy (AFTIR), *Journal of Geophysical Research*, 105(D17), 22147–22166, doi:10.1029/2000JD900287, 2000.
- Henderson, B. H., Pinder, R. W., Crooks, J., Cohen, R. C., Carlton, A. G., Pye, H. O. T. and Vizuete, W.: Combining Bayesian methods and aircraft observations to constrain the HO₂ + NO₂ reaction rate, *Atmospheric Chemistry and Physics*, 12(2), 653–667, doi:10.5194/acp-12-653-2012, 2012.
- Leitão, J., Richter, A. and Vrekoussis, M.: On the improvement of NO₂ satellite retrievals–aerosol impact on the airmass factors, *Atmospheric ...*, 2010.
- Lin, J. T., Martin, R. V., Boersma, K. F., Sneep, M., Stammes, P., Spurr, R., Wang, P., van Roozendaal, M., Clémer, K. and Irie, H.: Retrieving tropospheric nitrogen dioxide over China from the Ozone Monitoring Instrument: effects of aerosols, surface reflectance anisotropy and vertical profile of nitrogen dioxide, *Atmos. Chem. Phys. Discuss.*, 13(8), 21203–21257, doi:10.5194/acpd-13-21203-2013, 2013.

McMeeking, G. R., Kreidenweis, S. M., Baker, S., Carrico, C. M., Chow, J. C., Collett, J. L., Jr, Hao, W. M., Holden, A. S., Kirchstetter, T. W., Malm, W. C., Moosmüller, H., Sullivan, A. P. and Wold, C. E.: Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory, *Journal of Geophysical Research*, 114, D19210, doi:10.1029/2009JD011836, 2009.

Mollner, A. K., Valluvadasan, S., Feng, L., Sprague, M. K., Okumura, M., Milligan, D. B., Bloss, W. J., Sander, S. P., Martien, P. T., Harley, R. A., McCoy, A. B. and Carter, W. P. L.: Rate of Gas Phase Association of Hydroxyl Radical and Nitrogen Dioxide, *Science*, 330(6004), 646–649, doi:10.1126/science.1193030, 2010.

Randerson, J. T., Chen, Y., Werf, G. R., Rogers, B. M. and Morton, D.: Global burned area and biomass burning emissions from small fires, *Journal of Geophysical Research*, 117, G04012, doi:10.1029/2012JG002128, 2012.

Sander, S. P., Friedl, R. R., Ravishankara, A. R., Golden, D. M., Kolb, C. E., Kurylo, M. J., Molina, M. J., Moortgat, G. K., Finlayson-Pitts, B. J., Wine, P. H., Huie, R. E. and Orkin, V. L.: Chemical Kinetics and Photochemical Data

for Use in Atmospheric Studies

, 6 ed., Jet Propulsion Laboratory, Pasadena, California. 2006.

Stavrakou, T., Müller, J. F., Boersma, K. F., van der A, R. J., Kurokawa, J., Ohara, T. and Zhang, L.: Key chemical NO_x sink uncertainties and how they influence top-down emissions of nitrogen oxides, *Atmospheric Chemistry and Physics*, 13, 9057–9082, doi:10.5194/acp-13-9057-2013, 2013.

Torres, O., Ahn, C. and Chen, Z.: Improvements to the OMI near-UV aerosol algorithm using A-train CALIOP and AIRS observations, *Atmospheric Measurement Techniques*, 6(11), 3257–3270, doi:10.5194/amt-6-3257-2013, 2013.

Valin, L. C., Russell, A. R., Hudman, R. C. and Cohen, R. C.: Effects of model resolution on the interpretation of satellite NO₂ observations, *Atmospheric Chemistry and Physics*, 11(22), 11647–11655, doi:10.5194/acp-11-11647-2011, 2011.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y. and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), *Atmospheric Chemistry and Physics*, 10, 11707–11735, doi:10.5194/acp-10-11707-2010, 2010.

van Leeuwen, T. T. and van der Werf, G. R.: Spatial and temporal variability in the ratio of trace gases emitted from biomass burning, *Atmospheric Chemistry and Physics*, 11, 3611–3629, doi:10.5194/acp-11-3611-2011, 2011.

Vinken, G. C. M., Boersma, K. F., Jacob, D. J. and Meijer, E. W.: Accounting for non-linear chemistry of ship plumes in the GEOS-Chem global chemistry transport model,

Atmospheric Chemistry and Physics, 11(22), 11707–11722, doi:10.5194/acp-11-11707-2011, 2011.

Williams, J. E., Strunk, A., Huijnen, V. and van Weele, M.: The application of the Modified Band Approach for the calculation of on-line photodissociation rate constants in TM5: implications for oxidative capacity, *Geoscientific Model Development*, 5, 15–35, doi:10.5194/gmd-5-15-2012, 2012.

Yokelson, R. J., Christian, T. J., Karl, T. G. and Guenther, A.: The tropical forest and fire emissions experiment: laboratory fire measurements and synthesis of campaign data, *Atmospheric Chemistry and Physics*, 8, 3509–3527, 2008.

Yokelson, R. J., Karl, T., Artaxo, P., Blake, D. R., Christian, T. J., Griffith, D. W. T., Guenther, A. and Hao, W. M.: The Tropical Forest and Fire Emissions Experiment: overview and airborne fire emission factor measurements, *Atmospheric Chemistry and Physics*, 7, 5175–5196, 2007.