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

The methodology used depends on the assumption that biases between TM5 and OMI-observed NO2 result from emission errors. The case made for discounting errors in other aspects of the model NO2 budget (e.g. chemistry) is not particularly strong. In particular, what is included in the model in terms of organic nitrate chemistry (e.g. PAN precursors, isoprene nitrates)? This is likely important in determining the model NO2 lifetime in this region of enhanced biogenic emissions. e.g. a balance between HNO3 production and PAN production likely has strong implications for the NO2 distribution. An evaluation of model NOy speciation would be useful in terms of stating or quantifying uncertainties in the NO2 distribution. Huijnen et al., (2010) gives some detail of the TM5 chemistry, but only a limited evaluation in this region.

In TM5, PAN originates from the reaction of C_2O_3 and NO_2, and ORGNTR (lumped alkyl nitrates) are modeled as the product of the reaction of NO_3 with isoprene and the conversion of NO with XO2N, the NO to alkyl operator as described in Gery et al., (1989). For our purpose, where OMI has a local overpass time of 13:30, NO_x, HNO_3, and ORGNTR are the most relevant NO_y species, as the PAN lifetime is temperature dependent and thus will quickly decompose back into NO_2 in the tropical boundary layer.

Recent debate regarding isoprene chemistry therefore adds uncertainty to model concentrations of background NO_2 as laboratory observations show that the RO_2+ NO branching ratio should be lower and OH recycling by RO_2 isomers should be higher than the parameterizations implemented in current chemical mechanisms (Fuchs et al., 2013; Horowitz et al., 2007). Thus, model OH concentrations in pristine forested areas are significantly underestimated (Taraborrelli et al., 2012). We acknowledge that the modeled ORGNTR yield will not be perfect, but feel the inaccuracies introduced in background NO_2 concentrations will be small in comparison to the large fluxes of pollution from biomass burning.

Similarly, in the discussion of the study by van Noije et al. (2006) (page 22768) and comparison of year 2000 GOME observations with TM4 simulations using 1997–2002 average GFED emissions, it is not clear why this demonstrates that the TM4 and TM5 chemistry and transport of NOx over South America is reasonable. The inference appears to be that a 2x NOx emission produces a 2x NO2 column? This says nothing about the validity of the chemical scheme and model transport?

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 NO_2. In Williams et al. (2012) the authors implemented an on-line photolysis algorithm in TM5 v3 and found that a revised JNO_2 value increased NO_2 concentrations above 800 hpa, adding approximately 0.2×10^{15} molecules/cm^2 to the column over South America. Williams et al. (2012) also estimated that OH concentrations near the surface should increased by 15%, implying that the conversion of NO_2 to HNO_3 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 NO_2 to HNO_3, the predominant chemical sink of NO_2, and therefore a high bias in β.

Because HO_2 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 effected by biomass burning is likely greater than 5-10%. We therefore make an estimate based on the discussion above and the sensitivity simulations by Stavrakou et al. (2013) of increasing the reaction probability of HO_2 uptake on aerosols to 1, that β is overestimated by 25% and that NO_2 tropospheric columns are biased low in TM5 v3 by 0.2×10^{15} molecules/cm^2 + 0-20%. This estimate is conservative as HO_2 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 NO_2 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 NO_2 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 NO_2 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 NO_2 concentrations are low. Thus, an increase in the NO_x concentration will decrease the NO_2 lifetime. The instant dilution of the emissions will dampen this effect leading to an overestimation of the NO_2 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 NO_2 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. NO_2 Tropospheric Column Retrievals:

Incorrect assumptions about the NO_2 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 NO_2 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 NO_2 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 NO_2 tropospheric columns retrieved by DOMINO (Leitão et al., 2010). If aerosols are well mixed with the NO_2 plume, enhanced light scattering will increase the sensitivity to NO_2 in this layer. AMFs would be underestimated and NO_2 tropospheric columns would be overestimated if this effect were not taken into account. If aerosols exist as a layer above the NO_2 plume, they effectively shield the lower atmosphere, reducing the sensitivity to NO_2. Ignoring this effect would overestimate the AMFs and underestimate NO_2 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 NO_2 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. 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 HO_2 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 HO_2 loss to aerosols (Stavrakou et al., 2013)
    iii. 25% to account for resolution effects (Valin et al, 2011)
  c. TM5 NO_2 columns should increase by 0.2×10^{15} molecules/cm^2 + 20%
    i. 0.2×10^{15} molecules/cm^2 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 NO_2 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.
(5) Background Fraction Constant: The fraction of fire emissions to background emissions is accurate.
(6) Background Fraction Constant + Bias: The fraction of fire emissions to

background emissions is accurate, and the bias assumptions from (2) are included.

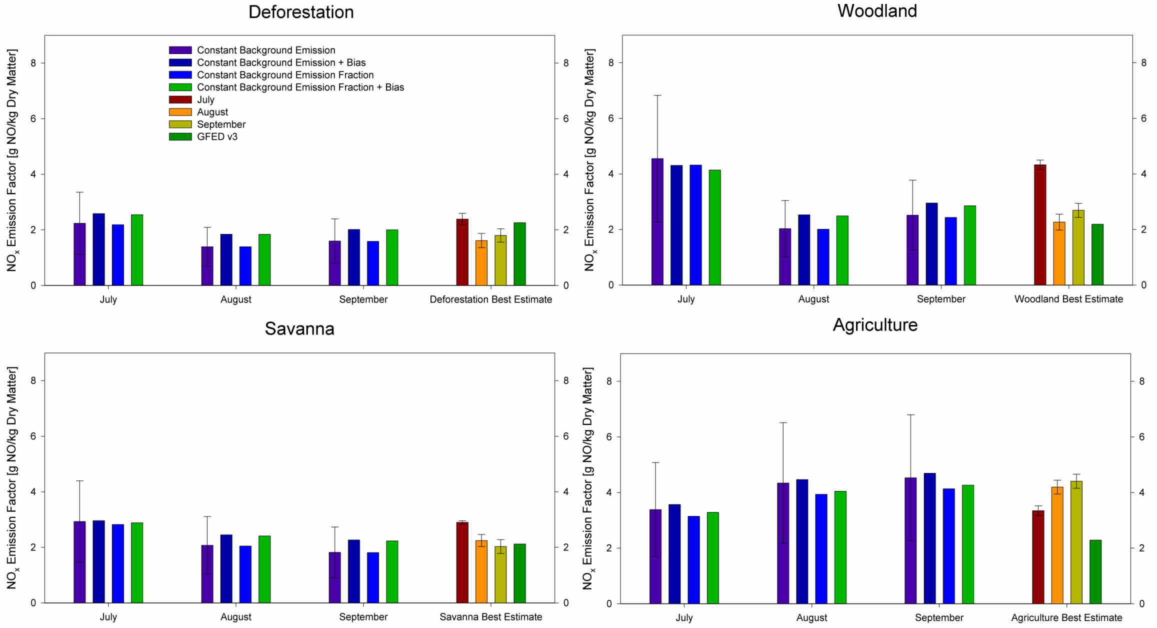

Figure 1. NO_x emission factor estimates considering two different assumptions for (a) the background NO_x emission contribution to the NO_2 tropospheric column, and (b) biases in model and observed NO_2 column. Error bars for the ‘Constant Background Emission’ case represent the 50% combined uncertainties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

The model chemistry scheme given in Huijnen et al., (2010) for TM5 appears not to include acetone. Is this correct? How might this omission lead to biases in NOy partitioning for this region?

> Although acetone does oxidize to C2O3, its lifetime is relatively long in the boundary layer due to rather weak photolytic destruction (Arnold et al., 2005). Therefore, we do not expect that this missing source of C2O3 in the CBM4 chemical scheme will significantly alter the partitioning of NO_2 into PAN for the lower troposphere over South America.

Specific comments Page 22771, line 12-15: Discussion of sources of uncertainty in deriving OMI-constrained emission factors. Huijnen et al., (2010b) appear to show model biases in NO2 somewhat larger than 30% over South America, the region of interest here.

> The reviewer is correct that over South America, outside of the burning season, the model bias is somewhat larger than 30%. However, over polluted North America and the European Union, where bottom-up emissions inventories are better constrained, the simulated NO_2 columns are on average within 30% of OMI observations. We acknowledge that NO_2 concentrations in pristine regions will be more uncertain (see the discussion on Page 1), but believe that for the polluted boundary layer during the peak of the burning season (the time frame for this analysis) an uncertainty of 30% for the TM5 simulation is appropriate.

Page 22777, line 9/10: Given that conclusions regarding increased emission factors from, agricultural burning is based on only a few pixels, is there any other evidence / literature to support this?

> Because of a bug in the code to calculate the OMI constrained NO_x emission factors, we revise the agricultural burning estimates to: 3.4, 4.3, and 4.5 g NO/kg dry matter for July, August, and September, respectively. Our original values were 4.0, 5.3, and 5.5 g NO/kg dry matter for July, August, and September, respectively. For the other biomes, the revised values differ from our original estimates by at most 7%, except for the estimate for woodland burning in July, which changes by 10% - 5.1 g NO/kg DM to 4.6 g NO/kg DM.
>
> Although lower than our initial estimates, on average the OMI derived NO_x emission factors for agricultural burning are still a factor of 1.8 higher than the 2.3 g NO/kg DM currently implemented in GFED v3. Yokelson et al. (2011) report 3.64 g NO/kg DM as the average from air borne sampling of 14 agricultural fires. However, the range of the observations was 2.2-5.7 g/kg DM, which overlaps with our estimates.

Fig. 5: It would be useful to also see spatial maps of the model-observation bias and how this changes spatially when using the new OMI-derived emissions versus GFED3.

> Change made. See Figure 2 below.

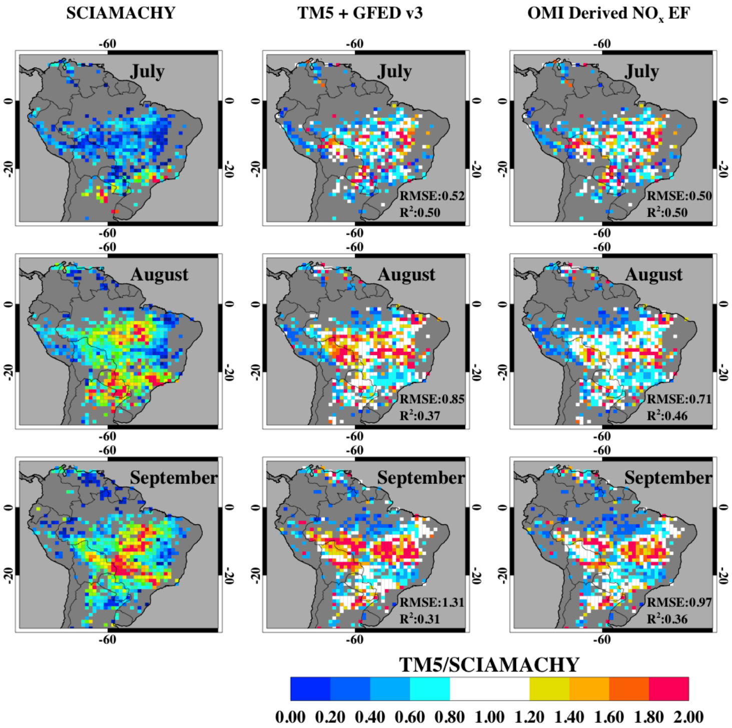

Figure 2. Monthly average SCIAMACHY observed (left column) and the ratio of modeled to observed NO_2 tropospheric columns (center and right columns). Only grid cells that have fire emissions as indicated by GFED v3 are considered in the monthly average. Satellite observations were re-gridded to 1◦ × 1◦ on a daily basis, where grid cell averages were taken only when the satellite had enough valid observations to fill 30 % of the grid cell. Satellite observations with cloud radiance fraction greater than 50 % (cloud fraction roughly 20 %) were excluded. In the center column are monthly average TM5 NO_2 tropospheric columns using GFED v3 emissions divided by the SCIAMACHY monthly NO_2 columns and in the right column are the TM5 NO_2 columns using the fire NOx emissions calculated with OMI derived monthly NO_x emission factors divided by the SCIAMACHY monthly NO_2 columns. The modeled columns have been transformed with the SCIAMACHY averaging kernels.

REFERENCES:

Arnold, S. R., Chipperfield, M. P. and Blitz, M. A.: A three-dimensional model study of the effect of new temperature-dependent quantum yields for acetone photolysis, Journal of Geophysical Research, 110(D22), D22305, doi:10.1029/2005JD005998, 2005.

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 NO_2 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.

Fuchs, H., Hofzumahaus, A., Rohrer, F., Bohn, B., Brauers, T., Dorn, H.-P., Häseler, R., Holland, F., Kaminski, M., Li, X., Lu, K., Nehr, S., Tillmann, R., Wegener, R. and Wahner, A.: Experimental evidence for efficient hydroxyl radical regeneration in isoprene oxidation, Nature Geosci, 6(12), 1023–1026, doi:10.1038/ngeo1964, 2013.

Gery, M. W., Whitten, G. Z., Killus, J. P. and Dodge, M. C.: A photochemical kinetics mechanism for urban and regional scale computer modeling, Journal of Geophysical Research, 94(D10), 12925–12956, doi:10.1029/JD094iD10p12925, 1989.

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 $HO^{\cdot} + NO_2$ reaction rate, Atmospheric Chemistry and Physics, 12(2), 653–667, doi:10.5194/acp-12-653-2012, 2012.

Horowitz, L. W., Fiore, A. M., Milly, G. P., Cohen, R. C., Perring, A., Wooldridge, P. J., Hess, P. G., Emmons, L. K. and Lamarque, J.-F.: Observational constraints on the chemistry of isoprene nitrates over the eastern United States, Journal of Geophysical Research, 112(D12), D12S08, doi:10.1029/2006JD007747, 2007.

Leitão, J., Richter, A. and Vrekoussis, M.: On the improvement of NO 2 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 Roozendael, 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.

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.

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 NOx 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.

Taraborrelli, D., Lawrence, M. G., Crowley, J. N., Dillon, T. J., Gromov, S., Groß, C. B. M., Vereecken, L. and Lelieveld, J.: Hydroxyl radical buffered by isoprene oxidation over tropical forests, Nature Geosci, 5(3), 190–193, doi:10.1038/ngeo1405, 2012.

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 NO_2 observations, Atmospheric Chemistry and Physics, 11(22), 11647–11655, doi:10.5194/acp-11-11647-2011, 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., Burling, I. R., Urbanski, S. P., Atlas, E. L., Adachi, K., Buseck, P. R., C, W., Akagi, S. K., Toohey, D. and Wold, C. E.: Trace gas and particle emissions from open biomass burning in Mexico, Atmospheric Chemistry and Physics, 11, 6787–6808, doi:10.5194/acp-11-6787-2011, 2011.