

Interactive comment on “Regional N₂O fluxes in Amazonia derived from aircraft vertical profiles” by M. T. S. D’Amelio et al.

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We thank the reviewer for a detailed reading of the manuscript and for his/her comments which we have made every attempt to address in full. In the responses below, we have attempted to clear up some misunderstandings about our flux calculation and definitions of words we use. Where appropriate, we have added clarifying statements to the text. In general, we feel we have taken the issue of uncertainties at every level of our analysis very seriously: from analytical uncertainty associated with calibrations to a full propagation of uncertainty of all the terms in our flux calculation algorithm. Moreover we have conducted sensitivity tests to explore potential biases in our approach and been very open about potential sources of systematic and random error, both in the text and by the plotting of error bars wherever possible. The review has certainly

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helped to improve the quality of our paper.

1. We have changed page 17430 line 12 to read “The higher rainfall around the MAN site could explain the higher N₂O emissions, as a result of increased soil moisture accelerating microbial nitrification and denitrification processes”. 2. (page 17433, line 1-15) We agree that there are significant assumptions and uncertainties in the flux estimation and have tried be open about them and to quantify some of these via sensitivity tests in the later sections. The introduction, however, is probably not the best place to discuss them. With regard to ‘integrate’, we feel its usage is appropriate. What we mean by ‘integrate’ is that a top-down method like ours implicitly accounts for all source and sink processes between the upwind/background measurement and the downwind/observation site, i.e. what is seen in the atmospheric mixing ratios is the sum of all sources. This should answer the second question of defining ‘total flux’: total flux = sum of all source/sink process between the Atlantic coast and our observation site. The top-down calculation contains no specific source information – e.g. it can’t distinguish, per se, between agricultural and forest emissions – but it will help constrain the total of all sources, known and unknown. 3. Since our analytical method is very precise and reproducible, we chose to show analytical details to contribute this knowledge to the research community. We would have liked to reference this, but there are no peer reviewed descriptions of the method. The issue of balance of detail is of course one of opinion, and in our opinion, because the method cannot be referenced we need here to describe it in sufficient detail for experimentalists. Those not interested in the details could simply skip over that section. 4. (page 17435, line 1-18) We do show in Figure 3 and state in the text that the small differences between the ascending and descending profiles suggest that local (i.e. ~100 km) influences are small. This may indicate that in the local area near the measurements, source strengths are fairly homogeneous. However, moving from the local to regional, we don’t believe our measurements can say anything about the heterogeneity of fluxes. By making measurements not just near the surface but up to 4 or 5 km asl, we are taking advantage of the naturally averaging properties of the atmosphere to gain an ‘image’ of the regional

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fluxes while sacrificing information on local variations. 5. (p 17435, l 19-25) Yes, all the profiles sampled between 2000 and 2005 (ascending and descending profiles) are included in Figure 3. The predominant wind direction in this region is presented in Figure 7 (the back trajectories were calculated for each profile and every 500m). Almost all profiles were sampled between 12-14h LT (or near this time), because this is the time with better condition and stability, under clear sky conditions. The time variation between the ascending and descending profiles is about 30 minutes. So there is very small possibility of the wind direction change. We have modified the caption of Figure 3 to include some of this information. 6. (p 17436, l 16-28) Reviewer 1 is correct in stating that the choice of convective boundary layer height was defined arbitrarily. It is important to stress, though, that this choice is for illustration only and plays no role whatsoever in our flux calculation. Looking at equation 3, z_{top} , the upper limit of the integral is defined as the top of the profile, not the top of the boundary layer. This is now stated more clearly in the text. In fact, we use a column integration technique (as opposed to a boundary layer budgeting approach) precisely to avoid the concerns that the reviewer expresses regarding the variability in CBL height. To clarify this, we stated in the text (17436-17437) "Because of strong convection one cannot count on surface emissions to be trapped in the CBL, so we use a column integration technique (Miller et al., 2007) that does not distinguish the CBL and free troposphere." That said, we choose 1200m as a convenient reference point considering the behavior of other gases sampled (CO₂ see below, CH₄ and CO) that tend to be relatively homogeneous above this altitude and show strong surface influence below this altitude. F_{ans_01} 7. (p 17436, l 16-28) We agree that it is difficult to see differences between above and below our arbitrarily defined 'CBL'. However, we wish to stress again that even though the CBL- Free Troposphere gradient is related to the surface flux, it is not relevant to our calculation methods. Thanks for pointing out the mistake on the heights. We had a typographical error and the right number at Figure 4 is: "Fig. 4. N₂O Time Series, Marine boundary layer sites ASC, RPB (thin lines) and FTL, SAN and MAN vertical profile average (a) above 1500m and (b) below 1200 m". 8. (p 17436, l28; p 17437, l

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1-5) There is a misunderstanding of referee 1. In the paper we explain that is one advantage of our method... "Because of strong convection one cannot count on surface emissions to be trapped in the CBL, so we use a column integration technique (Miller et al., 2007) that does not distinguish the CBL and free troposphere". As far as the type of convection, we are considering convection generally, both shallow cumulus convection and deep convection. It is true that flights are not occurring during periods of deep convection, but vertical profiles of air samples can still be influenced by deep convection that occurred in the previous days upwind of the site, thus redistributing surface flux initially trapped in the CBL into the free troposphere. Shallow convection will additionally cause redistribution of flux from the CBL. 9. (p 17347, l 8-10) SF₆ has a positive trend, because it has been constantly emitted through anthropogenic sources. This gas has a lifetime of 3200 years, meaning that it as long as there are emissions, its atmospheric concentration will continue to increase. For references: Olivier et al., 1990, Burgess, et al., 2006, and NOAA (<http://www.esrl.noaa.gov/gmd/ccgg/iadv/> - accessed in September 2009). 10. (p 17438, l 3) Thanks for this question as the assumptions do need to be either met or tested to be insensitive to the flux results: a. While this would be true when looking just at a single height – for example a measurement atop a tall tower – flux homogeneity is not a required assumption when using a column integral. For example, in the column, if all the flux were emitted either a) completely evenly b) all within 100 km of the coast or c) centered in a point, the profiles would all give the same integral, even though the shapes of the vertical profiles would likely be very different. b. This is also not a required assumption. Again, the boundary layer height is not a factor in our calculation. However, we do assume that a) the vertical profile of N₂O at the Atlantic coast is uniform and b) that the weighted MBL average we calculated based on RPB and ASC data is not affected by oceanic fluxes between those sites and the Brazilian Atlantic coast. Assumptions (a) and (b) can be checked by analyzing a small set of coastal vertical profiles measured above Fortaleza (see Fig 1). And indeed we see little or no vertical gradient in these profiles and also see that they match well with the ASC and RPB background, which is expected because the

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oceanic N₂O flux should be very small. To clarify this point, we have added the following text to end of the first paragraph of the Results section. “Additionally, FTL data show minimal vertical gradients validating our assumption that marine boundary layer data can be used to represent the column of air entering the continent.” c. This is a required assumption. There is a small source of N₂O in the atmosphere that comes from oxidation of NH₃, which is estimated to be ~ 0.6 Tg N/yr, with most coming from agricultural and industrial sources. In comparison, tropical forest soils are typically estimated to be an order of magnitude larger. Neglecting this source could potentially give a bias to our flux estimates as large as 5-10% at most. This is now mentioned in the Results. d. Perhaps the biggest assumption of our method is that flying to 4 or 5 km is high enough to capture all convective redistribution of surface flux. It most probably is not high enough, and neglecting flux that has escaped beyond out the top of our ‘leaky flux chamber’ will result in a negative bias to our flux estimates. This is mentioned on p17443 l 16. Additional information about the difference between the background and the highest part of profile (>3000m) gives an idea about what we lose with convection process that is not considered in our flux calculation. The comparison showed that the highest part of profile is 0.5 ppb higher than background for the annual average. This shows that the real flux should be higher than calculated one. (This information will be added in the paper). Since our flux calculation is based on each profile, it is independent of the variability during the year. 11. (p 17438, l 13) In the paper we explain that when it was not possible to determine a t value we used the method described in Miller et al, 2007, which is reproduced here: “t is the time since the air has been over land, estimated using mean 850 mb windspeed of 10 m/s (www.cdc.noaa.gov/cdc/data.ncep.html) and a mean distance to the coast of 1700 km (it is much less to the northeast and more to the southeast); we derive a mean value for t of 2 days, to which we assign a 50% uncertainty.” 12. (p 17438, l 15-20) The HYSPLIT model is cited in the paper with the home page where all information can be found (<http://www.ready.noaa.gov/ready/open/hysplit4.html>). The meteorological winds used as input to HYSPLIT were FNL winds from the NCEP global model 13. (p 17438, l 26;

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p 17439, l 1-16) We also struggled with the placement of this discussion. Since this comparison shows how our results are in agreement with NOAA this can be considered methodology, but on the other hand, this discussion is part of the assessment of sensitivities to our calculated flux, and thus belongs in Results and Discussion. 14. (p 17439, l 19) We have changed ‘climatology’ to “monthly mean flux”. We had used climatology to emphasize that our results were more valid statistically (due to inherent errors in wind times, background subtraction, etc.) when each month’s data over all years were averaged, compared to fluxes calculated for individual profiles. 15. (p 17439, l 24-30) We want to explain that the variability of the fluxes over time presents a higher value than the estimated uncertainty of our method for a single profile. This suggests that the profile to profile variability is not just a result of the uncertainties and actually has a geophysical origin. The paper has been revised to clarify this. 16. Figure 8c. Thanks for your observation. I think some problem happened and the fig 8c was removed. It has been fixed. 17. (p 17440, l 6) forrest has been changed to forest, but ‘forest conversion’ is correct usage; “where significant forest converted has occurred” does not make sense. 18. (p 17440, l 10-15) The rainfall data was measured by INMET (National Institute of Meteorology) monitoring stations, who supplied us the daily rainfall data. This information and some discussion of the relationship between N₂O fluxes and precipitation occur in these pages. For clarity, we now write ‘...composed of station data...’ l 14 17440. Model output should not be considered ‘data’ and we thought its origin would be clear. 19. (p 17440, l 27) It will be changed to “In some years (as January of 2003 and 2005)...”. 20. (p 17441, l 1-5) We do not ‘discard’ any negative values we calculate. All values are used in all the means presented and in their standard deviations. We are only saying that from a biogeochemical point of view, negative fluxes are not reasonable, and thus we interpret them as deriving from errors in our flux calculation method. Further, we never mean to suggest that the positive values are free from errors, whether they are caused by background subtraction or something else. In the paper we clarify that “Our method for calculation of flux is good to represent the mean behavior and not isolated points”. The major SF₆ source comes from North

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Hemisphere and we can see very clear some times a plume in one part of the profile. So, isolated points some times are not very precise for this reason, but in the average this kind of difference is minimized. 21. (p 17441, l 6) Thanks for the suggestion. We will change “interpreted in the climatological sense” to “interpreted seasonally” 22. (p 17441, l 8-10) Thank you for pointing out this inconsistency. However, upon rethinking our results, we can fairly state (as mentioned above in response to comment 15) that the calculation uncertainty for a flux calculated from a single profile is substantially less than the variability over a year. The sentence “Like the mean values, the uncertainty in our method means that we can not interpret the variability shown in Fig. 8a and b as actual flux variability.” has been removed. 23. (p 17441, l 20-25) We do not say that the surface heterogeneity is important for the calculated flux, only that different wind directions bring influence from different types of landscape, which may help explain some of the observed profile to profile flux variability. Moreover, equation (3) integrates the difference between the sampled profiles and the background regardless of the degree of heterogeneity. The background was defined the concentration in the coast, so, this difference considers all contribution between the coast and the site of sample, account for the surface heterogeneity. 24. (p 17442, l 1-15) As mentioned in the answers to comments 23 and 10, despite heterogeneity of sources, the integration method is not sensitive to this. Moreover, the biomass burning signal we observe may be local, but not necessarily local in relation to our measurements: i.e., we observe influence from biomass burning that has been transported from hundreds to 2000 km away from our site. 25. (p. 17442, l 20) We added in the text the calculation of the p value calculated by a two-tailed t-test. For all 11 profiles that we obtained correlation (r^2) higher than 0.3, it was significant at $p < 0.05$; this has been added to the text. The evidence observed in this study is explained from p 17442, l 16 until l 26 showing the clear evidence that sometimes we see emission from biomass burning together with CO₂, CO and Methane emissions. Then we know that the enhanced dry season emission is not always due to biomass burning. We don’t know all the factors regulating this emission. But is very important to publish this observed evidence to stimulate future studies. 26.

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(p 17443, l 10-15) We are only considering the surface flux in this paper. We realize that the way this was originally written could cause some confusion and these words have now been re-written to read: “We integrated the fluxes from the surface to 4270 m, but systematically excluded observations below a given height (152 m, 300 m, 600 m and 1000 m), thereby assuming the mole fraction at that given height was constant to the surface.” The sensitivity tests described in these lines are meant to ask the question, what would the result of our flux calculation be if we had no data below x m asl and instead assumed the mixing ratio was uniform from that level to the surface? In other words, we are not attempting to calculate vertical flux at these various levels; we are always still calculating the surface flux. If we were trying to extract information from the vertical gradient itself (for example if we were using a 1-D boundary layer budget approach), then the flux divergence (in the case of a 1-D budget, accounting for the horizontal advection) would be important, but not in our case. 27 (p 17443, l 21) We do not agree. The word “regional flux” means the scale that our data (from vertical profiles) represents. While this is beyond the scope of this paper, recent work we have been doing using the Lagrangian Particle Dispersion Model FLEXPART shows that the surface influence is substantial between our sites and the coast and is not dominated by the nearest several hundred km, for example. As described in the text, our sensitivity study of calculating flux while ignoring the lowest levels and the results of Figure 3 are also consistent with the fact that we in fact are seeing a regional (order 105-106 km²) and not a local flux (order 102- 104 km²). 28 (p 17445, l 12-20) Yes, as mentioned above, the calculation implicitly considers all sources and sinks from coast to sampled site. The main points in the conclusions are: a) we infer forest soil fluxes that are consistent with previous bottom-up estimates and tropical forest is certainly wide-spread in the eastern Amazon basin; and b) we see hints of larger than expected biomass burning contributions of N₂O. As mentioned earlier, although individual fires may be local in scale, burning as a whole is certainly a regional phenomenon in this part of the world.

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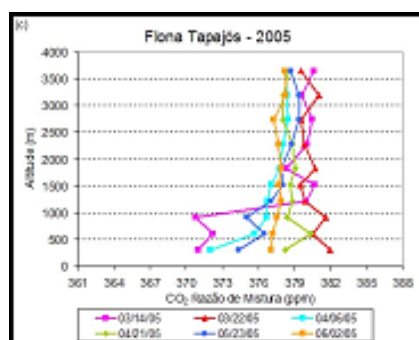


Fig. 1.

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