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Interactive comment on "Dry and wet deposition of inorganic nitrogen compounds to a tropical pasture site (Rondônia, Brazil)" *by* I. Trebs et al.

I. Trebs et al.

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The authors tackled a difficult task - the measurement of all major components of dry and wet deposition of inorganic reactive nitrogen species. They did so under rustic field conditions in the Amazon region of Brazil. Their results demonstrate that the deposition of reactive nitrogen under these conditions may be substantial.

They show that wet deposition is more important than dry deposition although the vast majority of the work treats dry deposition. This imbalance is awkward but reflects the author's concerns with the technology of measurement and the modeling of dry deposition. Overall, this is a valuable paper but the authors should pay closer attention to error analysis. Additionally, the authors should not extrapolate from three months when air in Rondônia is relatively polluted to a full year. The comparisons to global models are inappropriate.

Response: The authors do not agree with the last statement. The only way N deposition has been estimated in tropical regions up to now is by the use of global chemistry and transport models. Our study presents firsts results of N deposition based on field measurements in the tropics. From this standpoint, the authors find it worthwhile to compare their data to results derived in the past by global models. It is not true that the air was polluted at the measurement site over the three month. Moderate polluted to clean conditions were present at least half of the measurement period, such that extrapolation to the whole year seemed to be a reasonable approximation (more explanations, see below).

Reply to specific comments:

The analytical work follows precedents established in other work and appears in all aspects to be strong and appropriately described and defined. The inclusion of analytical precision estimates in Table 1 is very useful. I commend the authors for putting all of the analytically information in one convenient and informative table. Inclusion of references to the methods in the table would make this an even more valuable resource.

Response: The authors do not see a reason for this. Detection limit and/or precision of all instruments employed are listed in the last column of Table 1.

The discussion of the models for inference of dry deposition flux is well done. Analysis of errors beyond the analytical errors is not as complete as might be hoped. The authors chose to illustrate errors by selection of bounding cases with high and low flux estimates. This is commonly done where errors are difficult to define but I find the authors high and low bounds to be extremely conservative.

Response: All our estimates rely on surface-atmosphere exchange fluxes obtained in the temperate latitudes. This is well justified in case of HNO_3 and HONO. However, it is not known which factors mainly control bi-directional exchange of NH_3 in the tropics

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and which role surface water films and stomata opening plays in these processes. Therefore, we have chosen our input parameters (Γ, pH) such that the diel pattern of NH_3 exchange found in temperate latitudes may be reproduced. This estimate might appear conservative, but is the only reasonable approach the authors can think of.

One example of conservative high and low estimates can be given for the case of NO_2 the most important species for dry deposition. The surface resistance for NO_2 is taken between 200 and 300 s m^{-1} based on Kirkman et al. 2002. Kirkman reported this resistance as having median daytime and nighttime values of 209 and 229 with interquartile ranges of 182 and 149 for day and night respectively. Even if we accept the interquartile range as an adequate definition of error, the surface resistance of NO_2 might easily range from only slightly above 100 to somewhat more than 300. Interquartile range is not a usual definition of error and it is not conservative. One might guess that given the loose treatment of errors, the estimates of dry deposition could easily vary by more than the factor of about 2 given in the conclusion. Without a more detailed analysis, it is hard to know whether the error might not be closer to a factor of 10. The ranges are plausible but they are not carefully justified based on rigorous grounds for error analysis. The authors should attempt to be more conservative in their error analysis. They should justify the selection of ranges based on the distributions of concentrations and resistances used in their models and the probabilities of these ranges. Where distributions are irregular, boot-strapping analysis may be an option for estimation of probabilities.

Response: As the referee already mentioned, errors are difficult to define in our study and therefore we estimated high and low boundary cases (scenarios). The present dataset would not allow a reliable error analysis for the estimated fluxes. The authors agree with the referee that the dry deposition estimate of NO_2 is too conservative. The authors have decided to perform high and low flux analyses also for NO_2 by using the minimal and maximal values of R_c as determined by Kirkman et al. 2002. These R_c values will be used to give an estimate of the minimal and maximal NO_2 flux and a

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discussion of the consequences for the total annual deposition estimate will be added to the manuscript.

There are errors that are barely quantified but deserved more attention. For example, the flux of ammonia from excreta appears to be an afterthought in this work. The single estimate for the flux of ammonia from excreta (8 % of N) is based on a personal communication from L. Bouwman (Is this A.F. Bouwman as Lex is known in the scientific literature?). There is a considerable literature on ammonia volatilization that has been completely ignored. For example see the introductory paragraph from Frank and Zhang (1997) quoted below. "Ammonia volatilization from ungulate urine can be a major pathway of nitrogen (N) loss in grazed grassland (Woodmansee et al. 1978; Schimel et al. 1986). Measured losses of ammonia-N mostly range 10% - 40% of the urea-N added to plots; although amounts of 2-90% have been reported (e.g., Musa 1968; Stewart 1970; Denmead et al. 1974; Vallis et al. 1982; Bouwmeester et al. 1985; Schimel et al. 1986; Ruess McNaughton 1988). High variation in ammonia loss is a complex function of several interrelated factors that include soil texture (Schimel et al. 1986), organic matter (O'Toole et al. 1982), pH (Ernst Massey 1960), cation exchange-capacity (CEC; Campbell et al. 1984), soil micrometeorology (Sherlock Goh 1984), and vegetation (Ruess McNaughton 1988)." Ammonia emission from excreta given the authors estimates would total 3 to 4 kg N ha^{-1} yr^{-1}. This is as large as all estimated dry deposition. The net flux of ammonia depends on the correct estimation of the compensation point. This in turn requires a correct estimation of the ammonia flux from excreta. Might the loose treatment of the ammonia flux from excreta inject considerable uncertainty into the final estimate of total ammonia exchange?

Response: L. Bouwman is A.F. Bouwman, this will be changed. The authors have not used literature references for ammonia volatilization since, as mentioned by the referee, these estimates can vary by several factors. Instead, the authors decided to trust the estimate from an expert and use it for the calculations. Moreover, there is quite some evidence that pasture sites are Nitrogen limited (see introduction of the paper).

Also, Kirkman et al. 2002 found a low N status of the soil. Therefore, we assume that the Nitrogen recycled through animals and the subsequent ammonia volatilization from excreta is much lower than e.g., for European cattle, and that 8% is a representative value for the pasture site. Certainly, this flux component introduces considerable uncertainty in the overall ammonia flux, but is up to date “the best we can do”.

The extrapolation from three months of measurement to an annual flux is not justifiable. The month of September is the smokiest month of the year and is not representative of the full dry season. October is a transition but it is still quite smoky and even November taken as representative of the wet season is not nearly as clean as other wet season months. Data taken from MODIS on Terra show monthly average optical depth for a 5 degree by 5 degree region that approximates the location of Rondônia state (Review Figure 1). These data were retrieved through NASA’s Giovanni system <http://daac.gsfc.nasa.gov/techlab/giovanni/index.shtml>. Annual deposition flux may be substantially overestimated based on these three months where the air is quite dirty. Even if the annual estimate were justifiable, the comparison of this area of Rondônia to regionally averaged values for the Amazon is not appropriate. The area studied in the center of the state of Rondônia is one of the most heavily perturbed areas in the entire Amazon. The high density of cattle pastures in this area, leads to a very high frequency of vegetation fires in the dry season making this one of the more polluted areas of the Amazon region. The authors write (p. 3162, line 1), "Not surprisingly, our N deposition estimate for the Amazon pasture site is a factor of two to eight higher than model predictions for the Amazon region (Fig.13). Because this is not an especially useful comparison, Figure 13 should be eliminated from the paper. Furthermore, the conclusion that (p3163, line 8) "the contemporary net N deposition to tropical ecosystems may be underestimated by at least a factor of two" should be moderated. The tropical forest region should not be compared directly to the most perturbed section of the Brazilian Amazon.

Response: The authors do not agree with this. Figure 13 will not be eliminated from the

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manuscript. In section 4.8 the authors wrote: "Wet deposition of nitrate and ammonium in Central Amazonia was previously estimated to 1.7 kgN ha^{-1} yr^{-1}, 2.1 kgN ha^{-1} yr^{-1} and 2.8 kgN ha^{-1} yr^{-1} by Stallard Edmond (1981), Andreae et al. (1990) and Williams et al. (1997), respectively. Since these measurement sites were situated in remote areas with less fire density, these estimates are about a factor of two lower than a value of 4.7 kgN ha^{-1} yr^{-1} (ammonium and nitrate only; nitrite excluded) obtained in this study." This statement implies, that the measured wet N deposition under more pristine Amazonian conditions is already as high as most of the estimated by global models (see Fig. 13). Adding dry deposition to these pristine wet deposition estimates, the total deposition would be higher than predicted by global models. This shows that our findings for polluted conditions in Rondônia are reasonable. In this context, we would like to note that our argumentation is quite strong and will be changed to a more moderate discussion, mentioning the FNS site might be more polluted during the dry season than other Amazonian pastures which will increase our N deposition estimates. It should be noted that averaging the pristine Amazonian estimates with estimates from polluted Rondônia (based on field measurements) the average N deposition in Amazonia is clearly underestimated by global models.

Technical corrections:

The region in question is not rainforest but rather it is moist forest based on the commonly used Holdridge Life Zone system. The term "primary" forest is inappropriate. It may be more appropriate to refer to old growth forests. See Clark (1996) for a discussion of term related to tropical forests.

Response: This will be changed.

It is unnecessary to refer to "(micro)-meteorological data." These are simply meteorological data. (See p 3146, line 17).

Response: This will be changed.

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It would be useful to learn what portion of the data was excluded because of the turbulence considerations discussed on p. 3149. What bias may this have on the dry deposition estimates?

Response: about 10 % of the data were rejected; a comment will be added to this paragraph.

For Figure 3 it would be very useful to see a graph of D_r for the various species.

Response: We have calculated D_r as mentioned in the text. However, since the Damköhler ratio is not familiar to many scientists we have chosen the more common way of presenting characteristic time scales.

Figure 12 should be converted to a Table giving the absolute values of dry and wet deposition rather than simply percentages. Error estimates associated with the values would also be helpful. These should be given only for the study period and not as annual values as discussed above.

Response: This way of presenting the data was adopted from Hesterberg, R., Blatter, A., Fahrni, M., Rosset, M., Neftel, A., Eugster, W., and Wanner, H.: Deposition of nitrogen-containing compounds to an extensively managed grassland in central Switzerland, Environ. Pollut., 91, 21-34, 1996 and was found quite useful to evaluate the contribution of the different N species / processes to the total N deposition. The values represent averages of high and low flux scenarios and are not for the whole year, but only for the study period comprising of September, October and November (see caption of Figure 12a-c).

Figure 13 should be deleted because the comparison is inappropriate for reasons discussed above.

Response: We will not delete Figure 13 for the reason explained above.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3131, 2005.