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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric nitrogen budget in Sahelian dry savannas" *by* C. Delon et al.

Anonymous Referee #1

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1 General comments

This paper presents the budget for reactive atmospheric nitrogen compounds in Sahelian dry savannas. This semi-arid region is unexplored in terms of atmospheric N emissions and inputs. The authors used a combination of measurements of NO_2, HNO_3 and NH_3 using passive samplers and wet deposition measurements at three sites in Niger and Mali as well as modeling approaches to derive N budgets. The inferred deposition fluxes are scaled up to the whole Sahelian region and dry savannahs worldwide. Biogenic NO soil emissions are calculated using an Artificial Neural Network approach. The organic fertilization rate is modeled based on livestock populations, the synthetic fertilization rate is neglected. The NH_3 volatilization rate is estimated from other stud-

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ies. The NO_x and NH_3 emission inventories from biomass burning were based on satellite data (vegetation map, data on biomass densities and burning efficiencies) and emission factors reported in literature. Methods to estimate NO_x and NH_3 emissions from domestic fires were deduced from the literature. This paper contains unique data from an unexplored environment and the reviewer does appreciate the efforts made in this work. Although the paper may be a valuable contribution to our knowledge of nitrogen cycling in Africa, the reviewer has serious doubts about the applied methods used to derive dry deposition fluxes.

(1) All nitrogen species considered here are reactive, e.g.,

$$NH_3 + HNO_3 \leftrightarrow NH_4NO_3 \quad (1)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (2)$$

$$NO_2 + h\nu(\lambda < 420\,\mathrm{nm}) \rightarrow NO + O(^3P) \quad (3)$$

and their reaction timescales may be faster than turbulent transport times, which will not allow the application of the inferential method used here to calculate fluxes. The authors have not taken this into account and they have not shown that vertical exchange fluxes between surface and measurement height were constant.

(2) Apparently, not a single meteorological or micrometeorological measurement was performed to estimate or validate the applied v_d values; instead v_d values were taken from other studies. These studies, are however, also based on resistance modeling approaches and not on measurements (e.g., aerodynamic gradient or eddy covariance method) in dry savannahs, such that the applied v_d values are highly uncertain. NH_3 typically features bi-directional fluxes and compensation point concepts are applied. The authors have not considered this, although the NH_3 (re-)emission may be partially captured by the applied NH_3 volatilization rate.

(3) The paper neither contains quantitative error estimations of the fluxes nor estimates on potential maximal an minimal fluxes that may be calculated by varying v_d within a

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certain range. The simple upscaling approach to the global N budget for dry savannahs is therefore not justified. There is no evidence that the three measurements using passive samplers are representative for the entire Sahelian region and dry savannahs worldwide.

Knowing that it is very difficult to perform additional sophisticated measurements in this region, the authors (or rather the IDAF/AMMA participants) should at least consider to strengthen their work with micrometeorological measurements, which allow the determination of aerodynamic resistance R_a and the quasi-laminar boundary layer resistance R_b. The surface resistance R_c could then be modeled or deduced from other studies and/or scenarios could be calculated. Furthermore, multi-layer exchange models could be used to check for chemical flux divergences. Wind speed and wind direction should also be measured (see below for details).

I do not recommend the paper to be published because it does not meet the quality standards of ACP.

2 Detailed comments

Page 14194, lines 175-176: Which definition of detection limit was used (1σ, 2σ or 3σ)?

Page 14195, line 5: The unit ppb is mixing ratio, not a concentration (c); units for c must be in μg m^{-3} and for v in ms^{-1}. For which pressure and temperature were the concentrations calculated? At which height were the concentrations measured? An equation for the flux calculation with units is missing. Additionally, the applied inferential method $F = v \cdot c$ to calculate fluxes presumes that vertical exchange fluxes are constant with height, i.e. no advection and no internal sources and sinks (chemistry) along the transfer path is present (e.g., De Arellano and Duynkerke, 1992). The authors have not taken into account these considerations in their study such that the application of the

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method is not justified.

Page 14195, line 10: The concept of using constant deposition velocities taken from other studies in the absence of micrometeorological measurements is critical. Both studies from Zang et al. are based on big-leaf resistance modeling approaches to calculate deposition velocities and no values are given for dry savannahs. The applied values of 0.2 cm/s for NO_2, 1 cm/s for HNO_3, and 0.35 cm/s for NH_3 as mean deposition velocities are therefore highly uncertain and no experimental proof exists to justify their application. Additionally, deposition velocities are height dependent. Potential variations of v_d between wet and dry season are not taken into account.

Page 14195, line 24: What exactly are the uncertainties and how large are they?

Page 14195, line 26: '. . ..and is consistent with temporal scale of gas measurements.' What do the authors mean with this statement?

Page 14198, line 5: What is the uncertainty of simulated soil surface moisture and soil temperatures? Were they compared to measurements?

Page 14201, line 20: "As a consequence, 50% of loss rate has been applied to the input of N by animal manure previously prescribed for the calculation of NO emissions." This sentence is unclear. I guess a NH_3 volatilization rate of 50% was used and therefore the other 50% were attributed to soil NO emissions?

Page 14203, lines 16-25: Talking about the interconversion of NO_x and O_3 the authors should rather think about fast chemical reactions occurring between measurement height and surface, potentially violating the constant flux layer assumption (see above).

Page 14204, line 9: PAN decomposes at high temperatures (Grosjean et al., 1994). Its life time in Sahel would probably be low. Was the temperature measured?

Page 14204, lines 16-24: "Emission and deposition fluxes are similar during the wet season." And so on . . . Can you really say that? How certain is the deposition estimate?

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Afterwards, the authors state that the emission module produces errors, is there any estimate on how large these errors are? Do you keep these 'wrong' emission values in your further calculations?

Page 14204, line 25: The "maximum deposition" always refers to maximum concentrations since constant v_d values were used. A physical and seasonal interpretation of NO_2 and HNO_3 deposition fluxes is absolutely not possible since the authors have no knowledge how the true seasonal cycle of v_d would look like. The seasonal cycle of v_d is dependent on stomata opening, surface moisture and temperature, atmospheric turbulence and so on. . .

Page 14205, line 1-3: "The similar orders of magnitude observed between emission and deposition both in wet and dry seasons leads to the conclusion that NO_2 and HNO_3 deposition velocities have been correctly estimated for the region." How can the authors come to that conclusion? This statement is only valid if the three measurement stations explored are located in a completely closed and well mixed reaction vessel with constant surface properties and all N emitted would be finally deposited back to the surface. Can the authors proof this? Are wind direction and wind speed measurements available?

Page 14208, line 7: How about NO emissions from traffic in and around larger cities during the wet season? If air masses are transported from these regions such emissions may contribute additionally to N deposition fluxes. Can the authors exclude the influence of traffic?

Page 14208, line 20: How high is the level of uncertainty?

Page 14209, line 20-24: Can you really show that the three stations are representative for the whole Sahel region?

Page 14209, line 28: What are two-way fluxes? Do you mean bi-directional?

Page 14210, line 12 onwards: For all the reasons mentioned above is certainly not

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adequate to scale up from the measurements and modeling exercises to the global scale.

Page 14211, line 17: “These average fluxes are considered to be representative of dry savanna ecosystems.” The reviewer does not agree. There is no evidence for this statement in the paper, particularly with regard to the drawbacks of the method used to calculate dry deposition fluxes.

Page 14211, line 26 onwards: How high are all these uncertainties?

3 References

De Arellano, J. V.-G., and Duynkerke, P. G.: Influence of Chemistry on the Flux-Gradient Relationships for the NO-O_3-NO_2 System, Boundary-Layer Meteorology, 61, 375-387, 1992.

Grosjean, D., Grosjean, E., and Williams, E. L.: Thermal-Decomposition of PAN, PPN and Vinyl-PAN, Journal of the Air Waste Management Association, 44, 391-396, 1994.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 14189, 2009.