We would first like to thank Anonymous Referee #1 for its very helpful comments, which we hope have helped to improve the paper considerably. The comments from Anonymous Referee #1 are in normal font, while the answers are in *italic.*

**Reply to Anonymous Referee #1 comments bgd-9-c2123-2012**

This manuscript describes a simple parameterization of NH3 bidirectional exchange in the LOTOS-EUROS model and was reasonably well written. The model descriptions and results are likely to be of interest to many of the readers of Biogeosciences. I do have a couple of general reservations regarding the methods:

**General comments**

1.The assumption that the stomatal compensation point for NH3 was equivalent to the Henry's equilibrium dissociation assumes that the previous month's ambient concentrations were dependent on the stomatal compensation point alone. Any additional contribution to atmospheric ammonia from soil, animal or industrial sources would be inadvertently attributed to stomatal sources. Are the range of Γs used in this modeling study comparable to (Please present the range of Γs and indicate if these emissions are being double counted?)

*We believe that the stomatal gamma values are dependent on the previous month's ambient concentrations, not the other way around. Some research groups couple the value for Gamma_s to total N deposition, but we believe that a direct link with ambient ammonia concentrations is more realistic, as a high deposition of NOy would also lead to high Gamma_s values (NH4+ in the apoplast fluid), which is questionable. In the applied method, the stomatal compensation point adapts to its long-term environmental conditions. We acknowledge that the NH4+ concentrations in the soil also influence the available NH4+ in plants, but NH3 emissions due to enhanced NH4+ concentrations in the soil within 5 days after manure spreading are accounted for in the emission inventory. The stomatal emissions outside of this 5-day period are not accounted for in the emission inventory, so, there is no double counting due the stomatal emissions. In general, the stomatal gammas range from 100 to 10000 in background and agricultural areas respectively. For an explanation how the empirical relation for Γs was derived, see Eq. 16 and Figure 9 and the accompanying text in Wichink Kruit et al. (2010).*

2.The authors state that the compensation point accounts for some seasonality but then do not show any monthly or seasonal results to support this. Monthly observations apparently have a higher uncertainty, particularly in areas with a "low concentration range". However, the shift in the modeled seasonality with and without bidirectional exchange would be useful and the model could be compared to observations in "high concentration ranges" where there observational uncertainty is lower.

*The sentence mentioning the seasonality of the external leaf surface compensation point was wrong/not clear and is changed in: "The ratio between [$NH_4^+$] and [$H^+$] in the external leaf surface water, $\Gamma_w$, is further weakly dependent on temperature, which accounts for some seasonality in $\Gamma_w$." See also the comment on this point by the other referee (and the answer on this point) below:*

*p4883, l27: "weakly dependent": eq. 9 of 2010 AE paper by Wichink Kruit et al indicates an exponential function of temperature, and thus a strong (not weak) temperature dependence related to the NH3 Henry solubility and protonation constants.*

*Correct. There indeed is a strong temperature dependence in eq. 9. The sentence was intended to refer to eq. 13, but was formulated incorrect. We changed the sentence in: "The ratio between [$NH_4^+$] and [$H^+$] in the external leaf surface water, $\Gamma_w$, is further weakly dependent on temperature, which accounts for some seasonality."*

3.The manuscript describes the results in a generally qualitative manner and should be discussed more quantitatively, see the specific comments for details.

*We will try to add more quantative data to the manuscript.*

4.Both Figure and Fig are used to refer to the figures. Please follow the BG format.

*Only 'Figure' was used in the manuscript. BG format will be followed.*

**Specific Comments**

1.Abstract line 5: the acronyms DEPAC and LOTOS-EUROS should be defined

*The acronym DEPAC is explained, but the name LOTOS-EUROS is not. This is the name of the model that we use and is not an acronym.*

2.Abstract lines 13-14: Should "to a better extend" be "to a better extent"?

*Correct.*

3.Page 4879 lines 2 -3: There have been a number of measurements of dry deposition including several in this issue. I think that this statement needs to be qualified as regional, e.g. country to continental scale, estimates of dry deposition are generally made using chemical transport models

*Correct. We added: "regional, e.g., country and continental scale"*

4.Page 4880 line 23: What is intermediate complexity? I am sure that there are more descriptive ways of describing this model.

*We removed the first part of the sentence as it is vague.*

5.Page 4881 line 1: What exactly is "acceptable CPU time"? From my experience this could be anything from a few minutes in a windows environment to several hours of simulation time utilizing hundreds of CPU cores on a high performance computer cluster.

*Added: "of several days"*

6.Page 4884 Lines 3-4: References should be provided for "... based on many different studies of many different land use types".

*We added a reference to Wichink Kruit et al. (2010) in which all these studies are mentioned.*

7.Page 4884 line 5: "long-term concentrations" should be replaced with previous months concentration.

*Corrected.*

8.Page 4884 first paragraph: This parameterization appears to be at odds with micrometeorological and bioassay measurements of Γs have shown a rapid decay in Γs following fertilizer applications see Milford et al. 2001b. It seems like the parameterization presented here would miss these events and using monthly averages to parameterize the compensation point may smooth the temporal variability in the model concentrations.

*Correct. The increase in Γs after fertilizer applications is accounted for in the emission inventory. Accounting for increases in Γs after fertilizer application would cause a double counting of the emissions as the emission factors (in the emission inventory) are derived from net emission fluxes in the field. The parameterization presented/developed is only representing non-fertilized conditions, while the fertilizer applications are accounted for in the emission inventory.*

9.Page 4885 Lines 7-9: How the "multiplication factor ..." was estimated needs to be described better.

*As mentioned in the text, the multiplication factor is a first guess to obtain more realistic Γwater values than the constant Γwater value of 430 in the original DEPAC3.11 module. This is to account for the gradient that exists in the measurements of the Γwater values, which are rather well represented by this factor of 250 and the dry deposition pattern over sea. I believe this is clearly described in the text: "*The multiplication factor by which the dry deposition of $NH_x$ [in kg $ha^{-1}$ $yr^{-1}$] is multiplied to obtain the Γ values over water, is 250."

10.Page 4885 Line 24: The grid spacing of the high resolution domain should be presented. I am assuming that the grid resolution of the high resolution domain was 0.25o longitude by 0.125o latitude.

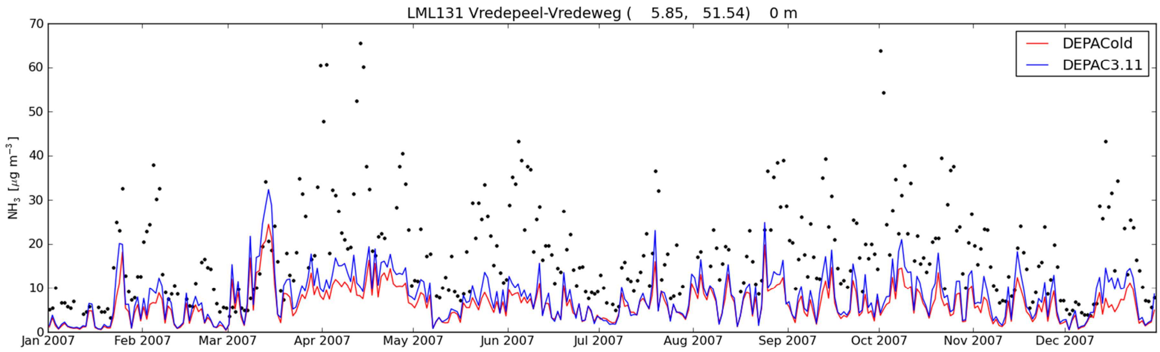
*This is a good point. Actually, the high-resolution run is on 0.125 x 0.0625 degrees, so we added the grid resolution wherever necessary/unclear.*

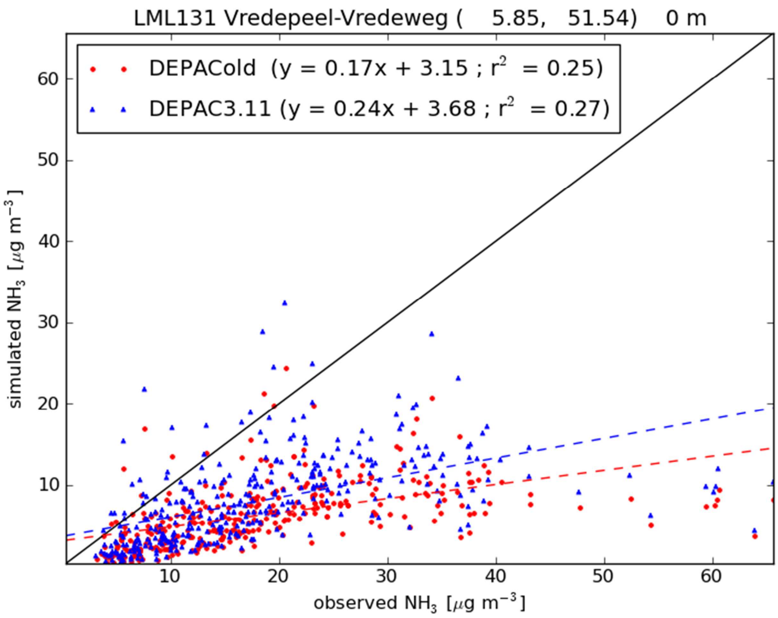
11.Second paragraph of section 2.3: Were the emissions the same in both model simulations? If so, were NH3 emissions from natural areas and agricultural crops estimated twice, once in the inventory and once in DEPAC3.11, in the bi-directional simulation?

*There are no NH3 emissions from natural areas and agricultural crops in the emission inventory. The emission inventory only accounts for fertilizer/manure applications and emissions from stables, while the compensation point approach only accounts for non-fertilized conditions of the vegetation.*

12.Page 4887 lines 18-19: One of the strengths in the modeling of bidirectional NH3 exchange would be a better representation of the seasonality by capturing temperature influences on the compensation point. Monthly time series figures should be shown to demonstrate how this model has changed the seasonality in the NH3 concentrations even if the uncertainty in the observations is greater in the monthly values.

*As can be seen in the Figure 10 of the manuscript, the seasonality is indeed better represented by the DEPAC3.11 version for station Westerland in Germany. For other stations in the higher concentration range, this effect is less pronounced. This is because concentrations generally peak around typical emission episodes (spring and autumn) and some of the model parameters in this parameterization are coupled to the previous hour or month concentration. Below, two figures are shown (a timeseries plot and a scatter plot) for a measurement station in the Netherlands located in an agricultural intensive area. Especially, the external leaf surface compensation point is low in summertime due to the temperature function in the Γw function and relatively low concentrations, i.e., the two most important factors in this exchange parameter. Low external leaf surface compensation points, which result in favorable uptake conditions, reduce the effect of relatively large stomatal compensation points, which would result in stomatal emissions and higher concentrations. It appears that there are no large differences in the net exchange during this summer period. The figure below shows that the largest differences between the two modules are found in spring and autumn and therefore also results in a better seasonality, which is reflected by the slightly higher correlation coefficient.*




13.Page 4887 line 23: This equation as it is written would make the observed concentration a function of the modeled concentration. I think something like Cobs(Zobs)/Cobs(Zmodel)=(Zobs/Zmodel)1/7 is what the authors meant. Additionally the wind power law assumes a zero slip boundary condition (U(Z=0)=0). If this is applied to ammonia concentrations, e.g. C(Z=0)=0, it is clearly at odds with the parameterization of a compensation point where the concentration at the surface is non-zero. How much did this interpolation change the results in the model evaluation? The authors may want to interpolate the modeled concentration gradient using the concentrations from layers aloft or the modeled compensation point to the observation height in the comparison.

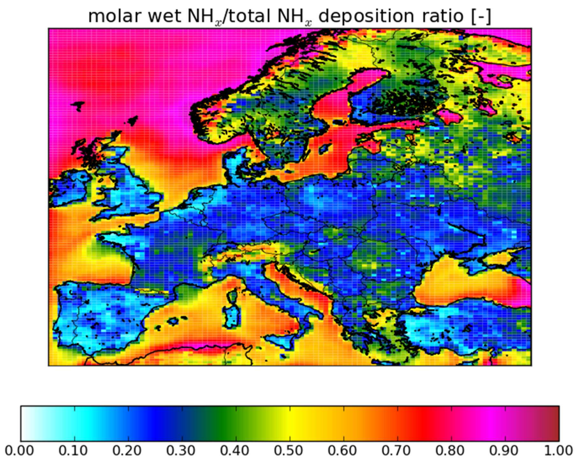
*The referee is right about this point. It is, however, not possible to correct the modeled concentrations to measurement height for each individual station. Therefore, the measurements need to be converted to standard model output height (2.5m). In general, the correction of the measured concentrations is relatively small (between 20% reduction (observation height 1.5 meter below model level) and 5% increase (observation height above model level)). Note that we use annual values here and that it concerns nature areas, which likely have very low compensation points, such that the assumption for a zero surface concentration might not be so wrong.*

14.Page 4889 Line 9: Please quantify "extremely low", e.g. on the order of 0.01 ppbv.

*Changed 'extremely' into 'very' and added 'in the order of 0.1 µg $m^{-3}$'*

15. Page 4890 Lines 1-2: It is not clear from Figures 4 and 5 that "This (presumably bidirectional exchange) makes nitrogen input by wet deposition more important than the input by dry deposition" and this claim should be quantified in the text.

*We added a plot with the relative contribution of wet to the total NHx deposition, which illustrates statement above.*




16.Page 4890 line 20: Is a few g m-3 approximately 2 g m-3?

*Changed 'few' into '*approximately 2'

17.Page 4891 line 10: What qualifies DEPAC3.11 as "better"? A lower bias or error?

*Reformulated paragraph.*

18.Page 4891 line 27: How much is "the bias in the regression" reduced?

*Added: 'with approximately 1 μg $m^{-3}$'*

19.Page 4891 line 29: Is a 0.06 drop in the r2 for 8 observations significant? Given the small number of observations, more robust metrics describing the model performance should be used.

*Due to the low number of stations, this figure (figure 8) will be combined with Figure 6, which lacks stations in the medium and high concentration range. The new figure will contain a better distribution of stations over a larger concentration range, which makes the statistics more reliable.*

20.Page 4892 line 1: Why is the modeled annual mean ammonia concentration at Eibergen not considered interpretable? Assuming that it is the point on the scatter plot with the largest increase, it appears from figure 8 that the modeled results at Eibergen were increased to be much closer to the observed value in DEPAC3.11.

*We didn't mean that the station is not considered interpretable, but the deterioration of the correlation is not considered to be interpretable. As figure 8 will be combined with figure 6, this comment is not applicable on the new data analysis anymore due to the larger number of stations considered and we will change the text accordingly.*

21.Page 4892 line 10: I agree with the authors that the grid cell size does introduce error when comparing to observations taken at a point, but the biases presented here appear that on the same order of magnitude as the precision in which ammonia can be measured using passive samplers. How does this compare with the uncertainty in the NH3 observations using passive samplers?

*We agree with the referee that the precision of the measurements is not mentioned in the text yet and should be considered when analyzing the results. Therefore, we added a sentence about the precision of the measurements of the passive samplers: "*These biases are in the same order of magnitude as the uncertainty in the measurements, which is estimated to be 10% for the annual average values (Stolk et al., 2009)."

22.Page 4892 lines 27-28: How many observations are in this "cluster of coastal measurements"?

*There are 26 yearly averages (of 90 in total) in the* "cluster of coastal measurements". We added this information to the text.

23.Page 4894 Lines 21-22: Was sulfate over the Mediterranean Sea in the DEPAC3.11 case fully neutralized and did this agree with measurements if they are available?

*We didn't check this so far; it is just a possible explanation for the observed changes. It would be beyond the scope of this paper to find this out.*

24.Section 4 would generally benefit from a better organization of the discussion and the conclusions should be discussed in a more quantitative manner when possible, e.g. "Altogether, the first order approximation ... to be quite successful". What is the definition of success in this case; a lower bias, the model runs with these changes, etc.?

*We will try to reorganize the discussion and conclusion section and try to add more quantative information instead of the relative quantifications.*

25.Page 4897 lines 6-7: Is this "not feasible" or computationally expensive?

*Replaced "not feasible" by "*computationally expensive"

26.Page 4897 line 15: Please quantify "only slightly affected".

*Added: "over land (less than 10% difference)"*

27.Page 4897 lines 25-27: You may want to expand on why you are comparing several CTMs in the ECLAIRE project and how that relates to this study.

*We included a model evaluation with NH3 concentrations from the "Nitroeurope" project in this study and removed the sentence here.*

28. Figures general: Specify the grid resolution in the figures

*Grid resolutions of the model runs are added in the captions of the figures.*

29.Figures 3-8, 10 &13: The font used for the titles and axes labels in these figures are too small to be legible.

*We will try to make the titles and axes labels more legible, but we will also describe the figures more extensive in the figure captions.*

Interactive comment on Biogeosciences Discuss., 9, 4877, 2012.