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***Interactive comment on “Modeling the distribution of ammonia across Europe including bi-directional surface-atmosphere exchange” by R. J. Wichink Kruit et al.***

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General Comments

This paper addresses the issues of ammonia (NH<sub>3</sub>) exchange modelling between the Earth's surface (including soils, vegetation, water bodies) and the atmosphere, and of how to deal with the bi-directional nature of the exchange, using the compensation point concept, within an atmospheric chemistry and transport model (CTM, here the LOTOS-EUROS model) at the regional (European) scale. The emphasis is placed on the implementation of a new surface/atmosphere scheme within the CTM, on the comparison of modelled concentrations with ground-based observations, and touching

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upon on sub-grid modelling issues. As such the paper certainly addresses relevant scientific questions within the scope of BG, even if some aspects might be more typically found in atmospheric journals. The regional-scale implementation of the compensation point approach for NH<sub>3</sub> is fairly novel in the CTM community, although by no means unique at the time of writing, and the comparison of the new model outputs with the older, deposition-only DEPAC version is well worthwhile. The paper is straightforward, well written and clearly structured.

The authors argue that accounting for the bi-directionality of NH<sub>3</sub> substantially improves the comparison with ground-based observations of NH<sub>3</sub>, although concentrations in receptor (semi-natural) areas are slightly over-estimated while concentrations in emission (agricultural) areas are still underestimated. The inclusion of the compensation point approach in the CTM is undeniably mechanistically more realistic, but I am not entirely convinced that the measurement datasets used to validate the new approach are adequate or comprehensive enough to provide unequivocal evidence of a substantial improvement in modelled NH<sub>3</sub> at the European scale. The authors only use EMEP data (17 sites for NH<sub>3</sub>), which are mostly located in North-Central Europe (with 12 sites in Germany, Denmark and Norway alone), which are expected to be biased upwards for NH<sub>3</sub> concentrations (cf section 2.4), and which mostly cover low- to medium-range concentration situations (16 sites out of 17 are under 2.5  $\mu\text{g}/\text{m}^3$ ). The large emission areas (such as The Netherlands, Brittany or the Po Valley, see Fig. 3) are thus under-represented in the EMEP validation dataset, and the lone point above 8  $\mu\text{g}/\text{m}^3$  in Fig. 6 drives the whole regression.

It is true that validation datasets are rare, but there exist additional, alternative NH<sub>3</sub> data at the European scale ( $\sim 55$  sites) from the NitroEurope project (e.g. Flechard et al., ACP, 2011), which, being based on denuder techniques, do not suffer the potential filter ammonium nitrate evaporation bias of EMEP data, and which cover a wider range of concentrations, especially at the higher end of the concentration spectrum. There are also other data from national monitoring initiatives. The paper would greatly benefit

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from including such data for a more thorough and comprehensive testing of the model, not only for NH<sub>3</sub> but also for NH<sub>4</sub><sup>+</sup>.

### Specific comments

p4879, l25: “. . .bi-directional ammonia exchange with the surface can be simulated rather well nowadays. . .”: this may be true at the field scale provided that model parameters are fitted to local flux observations, but more often than not these parameters are not suitable for other sites, as underlined by the authors. The empirical tuning that is required when dealing with new flux measurements shows that our mechanistic understanding is still far from complete. Perhaps the “rather well nowadays” is still a little too optimistic.

p4880, l1-2: “. . . Especially the representation of the dependency on the pollution regime seems to be important. . .”: this is true for non-stomatal uptake and also to some extent for the apoplastic ammonium content of semi-natural vegetation, but for NH<sub>3</sub> exchange over agricultural crops and (fertilised/grazed) grasslands it should be pointed out that the pollution regime is (much) less relevant than, say, the type of synthetic fertiliser (eg urea vs NH<sub>4</sub>NO<sub>3</sub>) applied, the type of organic manure, grazing intensity, soil microbial processes, litter decomposition. It is slightly misleading to suggest that, once generic relationships have been derived for stomatal compensation points and non-stomatal resistance (as in Wichink Kruit et al, AE, 2010), “. . .the new scheme is supposed to be widely applicable because it accounts for the local pollution climate. . .”. There is also a great deal of variation and uncertainty in the soil and leaf litter source (or sink) strength in unfertilised as well as fertilised vegetation, not to mention the strong dynamics of NH<sub>3</sub> volatilisation from applied fertiliser or slurry.

p4882, l15-16: “DEPAC3.11 assumes that ammonia is present in the vegetation, water surfaces and soils. . .” : in Wichink Kruit et al, AE, 2010, p947, there is “. . .no pathway for NH<sub>3</sub> exchange with soil in this model approach, because the canopy is assumed to cover the soil surface completely.”, and the RIVM report by Van Zanten et al. (2010) in-

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dicates (p15) that “. . . For the soil compensation point not enough information is known to implement a parameterization, so this variable is currently set to zero. “ The soil contribution to the source strength is therefore not accounted for in DEPAC 3.11, and the above sentence should be modified to reflect this, and it should be emphasized in “2.2.2 Description of the “DEPAC3.11” module”, lest the text mislead the reader. Further, the argument of a complete soil cover by vegetation is not universally valid, and certainly not throughout the year.

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 NH<sub>3</sub> Henry solubility and protonation constants.

p4884, l7-8: “. . . the mean ammonia concentration of the previous month is used as the long-term ammonia concentration. . .”: how critical is the operational definition of the “long term” concentration? Could (should?) the mean (inter-) annual concentration used as a more stable, longer term concentration? Could this be tested in alternative sensitivity runs (requiring perhaps iterative runs to account for circularity between Gamma and NH<sub>3</sub> concentration)? Importantly, were the parameterisations given in the 2010 AE paper by Wichink Kruit et al derived using the previous month’s concentration relative to the fluxes reported, or using an (inter-) annual mean NH<sub>3</sub> concentration? A discussion of the model sensitivity to this term would be helpful.

p4885, l7-8: Why is only the dry deposition term used to scale Gamma<sub>w</sub>, and not the total wet+dry NH<sub>x</sub> deposition, since the NH<sub>4</sub><sup>+</sup> concentration in water is directly linked to total deposition?

p4885, l22-24: it is not clear further on in the paper where the one-way zoom option and increased resolution were used. For example, p4892, l25, does “. . . the 7×7 km<sup>2</sup> grid size resolution of the model. . .” refers to both Figure 9 and Figure 8 ? It could be useful to indicate in each figure caption the grid resolution used.

p4886, l18: please say why you consider the evaluation of the model performance to

be a challenge: is it because there are too few ground-based monitoring networks and data at the European scale, or because of scale/sub-grid issues, or a combination of both?

p4886, I25: Please explain why you exclude mountain stations in the comparison.

p4887, I1-3: The authors make a good point about the representativeness issue, ie that single measured point concentrations in the landscape may not necessarily represent the average concentration of the grid square, so that even if the model is correct, the comparison is doomed to failure. It could be added that this is a problem because the model grid square is much larger than, say,  $1 \text{ km}^2$ , and because of the heterogeneity of landscapes. However, the authors appear to argue that the EMEP datapoints shown in Fig. 6 are likely higher than their regional/local background/average, and therefore biased upwards when compared with a modelled grid-square average. However, with 16 data points out of 17 being  $< 2.5 \mu\text{g}/\text{m}^3$ , the EMEP concentrations can hardly be considered “high” concentrations when compared with those measured in areas of intensive agricultural (for example, the Dutch case in Fig. 8-9, where most concentrations are above  $2.5 \mu\text{g}/\text{m}^3$ , and often  $>5$  or  $>10 \mu\text{g}/\text{m}^3$ ). I thus do not find the argument that EMEP stations are overly influenced by local agricultural sources (and thus “over-estimated” from a grid square perspective) entirely convincing.

p4887, I25: the logic for selecting large nature areas ( $>500 \text{ ha}$ ) is sound, but it is not clear to me what grid resolution was used here for the comparison. Is it the zoomed-in option mentioned in Section 2.3 and on p4892, I25 (“... $7 \times 7 \text{ km}^2$ ...”)?

p4888, I25: “Including the compensation point approach causes the modeled ammonia concentrations to increase...”: it is clear from the Wichink Kruit et al AE paper (2010) and from the Van Zanten et al RIVM report that the change from DEPACold to DEPAC3.11 is not just about including stomatal and leaf surface compensation points, but that there were very significant changes in non-stomatal resistance Rns. What can one say about the respective individual impacts of introducing compensation points

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and changing resistances? You mention earlier that some of the uncertainty in  $R_{ns}$  is transferred onto  $\Gamma_{ns}$ , and this is an important point.

p4889, l11 onwards, and Fig.4 : Since this paper concerns itself with surface-atmosphere  $\text{NH}_3$  exchange, there should be at least one figure showing the European-scale  $\text{NH}_3$  exchange by both models. Figure 4 with  $\text{NH}_x$  ( $\text{NH}_3 + \text{NH}_4^+$ ) exchange is useful, but an equivalent figure for  $\text{NH}_3$  only is needed, or perhaps just two additional panels in Fig. 4, above  $\text{NH}_x$  exchange.

p4889, l16-17: “. . .while the largest increases are found in the larger nature areas. . .”: here one could add also in more extensive rural areas, which are not necessarily nature areas.

p4990, l3-11: since there are substantial changes to spatial patterns of wet deposition, can you say whether the new spatial patterns and their magnitude are more consistent with measured wet deposition from monitoring networks across Europe? The validation should be easier for wet deposition than for  $\text{NH}_3$ , for reasons mentioned above (quantity of available data, spatial variations and representativeness).

p4890, l21-23: “The slope of the regression between the simulated and measured concentrations is improved from 0.68 to 0.92. . .”: this is largely driven by one single point at ca 8-9  $\mu\text{g}/\text{m}^3$  (Eibergen station in the Netherlands). What is the change in slope without this single point? Further, why is this Eibergen data point not shown in Fig. 7? Since  $N=17$  is a subset of  $N=33$ , all data points of Fig.6 should also feature in Fig. 7, although clearly other data points are also missing in Fig.7 (compared with Fig.6), such as those in Italy, Hungary and the Czech Republic.

p4891, l5-6: “. . .in Fig. 7. Again, the largest increases occur in the higher observed concentration range. . .”: Unfortunately, the data in Figure 7 are medium range and do not extend very far into the high concentration range (only up to 4  $\mu\text{g}/\text{m}^3$  for  $\text{NH}_x$ ), so the model cannot be said to be validated for areas of intensive agriculture and emissions. Why not show the data from the Dutch monitoring network  $\text{NH}_3$  data (Fig. 8) alongside

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the EMEP data in Fig.6 (using e.g. different symbols), even if there are grid resolution and representativeness issues, which are discussed in detail anyway ?

p4891, I17-18: “. . .we now zoom in on the area with intensive agricultural activity in the Netherlands and compare the modeled concentrations with measurements from the NAQMN in Fig. 8.” : please say explicitly whether this model run was done with a different grid resolution ( $7 \times 7 \text{ km}^2$  ?), or whether you are merely comparing the default model runs (European scale , at  $25 \times 25 \text{ km}^2$ ) specifically with a focus on the Dutch monitoring network. In fact it would be very informative to show on the same graph a comparison of measured concentrations from the Dutch network and of modelled concentrations using the high and the low resolution modes of LOTOS-EUROS/DEPAC 3.11. Is there a significant improvement when using a higher resolution, and if not, this points to a parameterisation issue rather than a representativeness issue for measured concentrations.

p4891, I27: it seems to me that the bias (offset) in the regression in Figure 8 is not reduced, but increased from 1.39 to 1.92 from DEPAC old to DEPAC 3.11.

p4894, I13: “The surface resistance of  $\text{SO}_2$  was reduced. . .”: was this harmonization necessary because the  $\text{NH}_3/\text{SO}_2$  molar ratio increased (cf co-deposition issue) ?

p4894, I26-27: “. . .The impact on the model performance of LOTOS-EUROS for the SIA components in comparison to measurements is very small. . .”: this statement is not very clear. Do you mean that the improvement of model performance versus measurements is marginal? Please rephrase.

p4895, I9: “. . .require detailed meteorological and plant physiological parameters. . .”: it should also (again) be mentioned that soil sink and source strength is also a major component, although largely unknown and under-studied, and the soil source aspect is absent in this study.

p4895, I11-13: “The generalization step is often a shortcoming as parameterizations

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are derived for just one certain land use class, which is located in one certain pollution climate.” This may be so, but this paper cannot ignore that there have been other attempts beside the author’s to derive and implement generalised parameterisations, e.g. Massad et al, ACP, 2010; Zhang et al., JGR, 2010; CMAQ model, USEPA, cf [http://www.cmascenter.org/conference/2010/slides/bash\\_evaluation\\_coupling\\_2010.pdf](http://www.cmascenter.org/conference/2010/slides/bash_evaluation_coupling_2010.pdf)

p4895, l23-27: the co-deposition issue is still a reality, but SO<sub>2</sub> is no longer, or not necessarily, the dominant nor the only source of leaf surface acidity. Rather than take the NH<sub>3</sub> to SO<sub>2</sub> ratio, one should take the NH<sub>3</sub> to the sum of acids (SO<sub>2</sub>, HNO<sub>3</sub>, HCl, etc) as a scaling factor for the non-stomatal resistance. See Fowler et al, At. Env. Volume 43, Issue 33, 2009, Pages 5193–5267, Fig. 4.5 and comments.

p4897, l7: “. . .is not feasible.”: add “for CPU time reasons” ?

p4897, l25-27: I do not think this sentence is relevant nor needed here. As mentioned above, the "Nitroeurope" data alluded to (see Table 2 of Flechard et al, ACP, 2011) could easily be added to Figures 6 and 7 without much additional work, adding more weight to this comparison exercise.

#### Technical corrections

p 4879, l9: “. . .It HAS long been known. . .”

p4879, l13: “. . .over all KINDS of vegetated surfaces. . .”

p4879, l28: “. . .which ARE generally not available TO regional. . .”

p4882, l25: for clarity, suggest change to : “. . . an external resistance that represents deposition to non-stomatal leaf and stem surfaces, this sink being especially active when these surfaces are wet.” Similarly, on p4885, l19, change “. . .updated dry deposition. . .” to “. . .updated surface exchange. . .”

p4883, l1: insert “default” between “contains” and “values”

p4883, l15: change “dry deposition” to “surface exchange”, since this (the bi-

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directionality of fluxes) is precisely what distinguishes DEPAC 3.11 from DEPACold. Also, change “removal” to “exchange” on l16.

p4883, l21: “. . .as it WAS derived. . .”

p4883, l22: Add “In the present study,. . .” before “The external leaf surface. . .”

p4888, l27: “. . .number OF measurements. . .”

p4889, l3: “. . .amounts TO 1-2  $\mu\text{g}/\text{m}^3$ . . .”

p4892, l6 and l9: change “with” to “by”

p4892, l14: change “would be” to “was”

p4894, l11: change “. . .in this figure. . .” to “. . .in Fig. 13. . .”

p4894, l13: change “synchronized” to “harmonized”

p4894, l19: change to: “. . .OVER the Mediterranean Sea the ATMOSPHERIC  $\text{NH}_4^+$  concentrations. . .” to dispel any ambiguity ( $\text{NH}_4^+$  “in” the sea might otherwise refer to the aqueous concentration).

Figure 1: add distance unit (km) to the x-axis

Figure 12: the two panels should be separated (insert a space in the middle, or display maps on two different lines). Also: add scale (km), add a larger situation map of Europe to make it clear where this modelling domain is located. For measured concentrations, if possible : add color-scaled symbols, with black outline and with same colour scale as the modelled map.

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