Dear Editor and reviewer 1 and reviewer 2

Concerning the article “Spatial and temporal variations in ammonia emissions – a freely accessible model code for Europe” under review for publication in ACP

This document concerns the comments both reviewers had to the manuscript as well as the suggested changes to the original manuscript

On behalf of all co-authors
Carsten Ambelas Skjøth

In general we are very pleased with the positive review from both reviewers and we would to thank the referees for suggesting improvements to the manuscript.

Due to the suggestions from the reviewers we have suggested extra background material into the discussion of the results about what has been done in other countries concerning temporal ammonia emission modelling.

This concerns temporal resolution of ammonia in the UK (Hellsten et al., 2008), model results with the FRAME and EMEPUK model including spatial and temporal variations of the emissions in the United Kingdom (Singles et al., 1998; Vieno et al., 2010a) and Poland (Kryza et al., 2011) and methods for handling ammonia emissions in the Netherlands on either national scale or specific landscape experiments (e.g. van Pul et al., 2008).

Additionally we have added more information to the statistics according the suggestions by reviewer 2.

Due to these general comments we therefore suggest following general amendments to the manuscript:

<u>On page 2137, line 28 to page 2138, line 7</u>
Change from
“Only a limited number of countries include emissions from crops in national inventories like those available from EMEP. A much better estimate than the default numbers in Table 2 is likely to be specific country based activity data as it is shown in this paper for the Danish area. A third factor is the information about the exact location of the agricultural fields and more detailed information about the location, amount and type of animals. Again this is an area where very detailed and high quality data are available for Denmark. Data of similar

detail cannot currently be obtained for other countries, but there may be some possibilities in analyses of satellite images and using international or national statistical data concerning agricultural holdings."

To following much more extensive discussion

"Only a limited number of countries include emissions from crops in their national inventories. A better estimate than the default numbers in Table 2 is likely to be specific country based activity data with a higher spatial resolution as it is shown in this paper for the Danish area. A third factor is the information about the exact location of the agricultural fields and more detailed information about the location, amount and type of animals. As ammonia has a high deposition rate, knowledge on the location of large ammonia emitters in the vicinity of the monitoring stations and their temporal emission stations is essential. This would probably alter the emission strength and the emission pattern for e.g. the German stations Zingst and Westerland where a large part of the grid is water. This kind of data was available for the Danish area. Data of similar detail cannot currently be obtained for other countries. Data of similar detail are likely to exist in other countries, such as the Netherlands (e.g. van Pul et al., 2008) and the UK (e.g. Hellsten et al., 2008), but are publicly not available due to confidentiality issues. However, there may be some possibilities in analyses of satellite images and in use of international or national statistical data concerning agricultural holdings like for the UK (Hellsten et al., 2008) or Poland (Kryza et al., 2011).

The dynamic temporal distribution model used in this paper showed a good performance for the Danish area and a smaller improvement for the German stations. This can be attributed to the spatial and temporal resolution of the input data. The ammonia emission is known to be dependent on incident global radiation, temperature, precipitation and humidity (Sommer, 1997; Sommer et al., 2003). The two major challenges in a temporal parameterization are therefore firstly how well the annual emission reflects the real emission. As many national inventories are only using one single emission factor to represent different activities like manure application, which take place under very different climatic conditions the emission estimates may be biased. Secondly, how well do the climatic co-variables included in the temporal model describe the real emission strength? The model used here (Gyldenkærne et al., 2005) is based on simple temperature relations and no dependence on solar radiation, precipitation and humidity is implemented. Hellsten et al. (2008) distributed the annual emissions in the United Kingdom in months according to farming activities to estimate "Emission potentials" and compared the monthly emission (kg N ha-1) with measured ammonia concentrations in the air (µg N m3). The UK data has a high spatial resolution of 5*5 km^2 which feeds directly into high resolution CTM models like the FRAME and EMEP4UK models (Singles et al., 1998; Vieno et al., 2010b; Vieno et al., 2010a). However, no climatic co-drivers were included in the temporal distribution of the ammonia emissions that feed into these models. The results from Hellsteen et al (2008) as well as more detailed process based studies (e.g. Sommer, 1997; Sommer et al., 2003) suggest that temperature effects should be taken into account when the emission estimates are used in chemical atmospheric transport models. "

In addition to this comment concerning the general contents of the manuscript, then the reviewers have specific questions which we address below:

Reviewer 1 writes:
"Why have these 9 particular measurement sites been chosen? And why only Denmark and Germany?"

Reply:
The 9 particular stations were chosen as they were the sites from the EMEP network that were within the model domain and that had measured ammonia as well as ammonium concentrations for the entire 2007. One additional site: Vreedepeel from the Netherlands does in fact also provide such data. However as written in the manuscript, then this station has been disregarded as this site is known to be influenced by very local sources which generally makes it difficult to reproduce the observed levels by regional scale CTMs. To the knowledge of the authors, then detailed hourly observations of ammonia have been made within the EMEP network within a dedicated campaign. These data were not available for this study.

Reviewer 1 writes:
Page 2133, row 9: Why have these particular dates (15 February and 15 April) been chosen?

Reply:
These two particular dates were chosen as two typical examples of ammonia emissions: The 15th of February then near all emissions are due to buildings and storage as application of manure during spring is usually in March and April and emissions from the 15th of February can to a good approximation be considered not to be present. Additionally, the temperatures are low which also reduces the emissions from non-heated buildings and storage facilities. The 15th of April is one of the peak emission periods in Northern Europe, where application of manure and mineral fertilizer takes place.

Reviewer 1 writes:
Page 2138, row 4-7: These types of data may exist also in other countries, but may not be publicly available due to confidentiality issues.

Reply:
This section is suggested to be expanded considerably and takes exact this comment into account, as such data exists at least for the Netherlands and the United Kingdom.

Reviewer 1 suggests following technical corrections:
"Consider changing the use of "animals" to "livestock", which is more appropriate in this context. In the paper, "modeling" / "modelling" is sometimes spelled with one "l" (American English), and sometimes with two "l:s" (Brittish English). Please make sure to choose one of these spellings, to have consistency in the paper. Some references are missing in the reference list, e.g. Dragosits et al. (1998) and Sutton et al. (2000). More references may be missing, as I have not looked through all of them. It is therefore recommended that the author check to make sure that all references are included. Page 2134, row 23: add an "s" to "concentration". Table 3 & Table 4, table text, row 3: Add a comma "," to the sentence: "In the first step"," data (new): : :.." to improve readability. Table 3 & Table 4, table text, last row: Change "unusual" to "unusually"."

Reply:
All this comments of technical nature to improve the manuscript has been highly appreciated and they are all taken into account. This includes following two references:
1. (Dragosits et al., 1998)
2. (Sutton et al., 2000)

If all the suggested changes are accepted, then the manuscript will be extended totally with 9 references. They are marked with bold in the reference list

Reviewer 2 writes:
I do believe that the paper could benefit from a more balanced presentation of the results. I suggest the authors address these issues before publication in ACP.

Reply:
We have added a substantial increase to the discussion of the results. Additionally we have also modified the tables 3 and 4 and we suggest further additions to the manuscript according to the specific comments below:

Reviewer 2 writes:
Page 2135 describes that the correlation between the simulated NH3 and measurements improves at all sites. It is also true that the bias increases at most sites. Please discuss why the correlation is improved, but the bias is not.

Reply:
The reviewer is right that this feature of the model results must be highlighted. We therefore suggest following change to the manuscript, line 2-3 on page 2137:
from
"especially for the spring peak. This overestimation"
To
"especially for the spring peak. This means that the model results has a larger seasonal variation compared to the old method. This variation do to some degree fits the overall seasonal measured values. Such changes can be seen by an increase in correlation coefficient and at the same time an increase in bias at some stations. This overestimation"

Reviewer 2 writes:
Because there is large seasonal variability, it would be best to report the bias metrics normalized by the mean, as a percentage rather than an absolute amount.

Reply:
This is in fact a good suggestion. But to the original numbers will be lost if we normalize the data. Additionally the absolute concentration level can be important instead of a relative fraction. One example is that the uncertainty of measurement obtained by the filter pack method is related to the absolute ammonia concentration.(Andersen and Hovmand, 1994). We therefore suggest changes that include the original data as well as the absolute measured concentrations, which will be an update of Table 3 and Table 4. The new Table 3 and Table 4 are found below.

Reviewer 2 writes:
On page 2136, the results of Figure 11 are discussed and compared to the results in Table 3. I find it very difficult to reconcile these data. In Table 3, Langenbrügge with the new emissions has a more negative bias and an improved correlation. In Figure 11, it seems the ammonia concentration increases considerably at this site, causing large divergence from the 1:1 line. How can these both be correct? Are these points to the right outliers? It would be possible to interpret if the
authors used a box plot rather than a scatter plot with so many overlapping points.

Reply:
The reviewer is right. Table 3 and Figure 11 do not match with respect to the statistics concerning bias. This is unfortunately a technical error that was introduced when modifying Table 3 and 4 for the ACPD production. All negative biases are supposed to be positive and all positive are supposed to be negative. Absolute values are the same and the discussion is related to the correct values. The updated Table 3 and Table 4 including the additional numbers concerning the absolute measured values will then look like the two tables seen below. Concerning the scatter plot, the reviewer is right there are a number of points concerning the station Langenbrugge with large divergence from the 1:1 line. This is due to a large overestimation of the spring concentrations which can also be seen in the new Table 3 that shows a considerably bias in the spring concentrations with the new parameterization. This bias was not present with the old method. Box plots are often used in the interpretation of measured concentrations while model comparisons usually use scatter plots or time series. We have used the later and after the additions to Table 3, we believe that the need for a box plot to complement the scatter plot has been considerably reduced.

Additional comments:
Please find the suggested new Tables 3 and 4 as well as a reference list from this document, where the references marked in bold are supposed to be added the paper.

Table 3. Correlation coefficients, observed mean and modelled bias between measured and modelled NH_3 before and after implementation of the new dynamic NH_3 emission inventory for northern Europe. Two steps have been used in the implementation of the new dynamic emission model. In the first step data (new) from the RAINS model have been used to distribute the national emissions in each grid cell of the high resolution domain. In the second step (dk), the previously presented detailed Danish emission distribution on the 15 emission functions presented in Table 1 in (Skjøth et al., 2004h; Skjøth et al., 2008a) has been implemented as a supplement to the RAINS data. Calculations performed with the DEHM model for the year 2007.
* At Keldsnor measured values from three days in October 2007 have been excluded here as they were unusual high and probably represented very local emissions.

	Correlation coefficient						Mean meas. concentration [µg/m^3], Bias [µg/m^3]				
	type	year	winter	spring	summer	autumn	year	winter	spring	summer	autumn
Zingst	old	0.61	0.18	0.61	0.73	0.61	1.06, -0.45	0.48, -0.10	1.60, -0.87	1.11, -0.53	1.00, -0.29
	new	0.68	0.43	0.65	0.71	0.56	-0.40	-0.35	-0.46	-0.51	-0.26
	Dk	0.68	0.43	0.66	0.71	0.56	-0.39	-0.35	-0.44	-0.49	-0.30
Langen-brugge	old	0.37	-0.03	0.38	0.54	0.15	1.29, 0.24	0.66, 0.44	1.89, 0.03	1.59, -0.24	1.04, 0.73
	new	0.48	0.18	0.21	0.49	0.59	0.47	-0.25	1.56	-0.15	0.67
	Dk	0.48	0.18	0.22	0.50	0.59	0.47	-0.25	1.56	-0.15	0.67
Wester-land	old	0.59	0.25	0.77	0.74	0.51	1.26, -0.82	0.57, -0.27	1.52, -0.93	1.82, -1.41	1.05, -0.60
	new	0.62	0.32	0.79	0.71	0.41	-0.75	-0.49	-0.51	-1.39	-0.58
	Dk	0.69	0.34	0.81	0.74	0.41	-0.74	-0.49	-0.43	-1.30	-0.70
Neuglob-sow	old	0.27	-0.16	0.04	0.29	0.23	0.84, 0.14	0.32, 0.36	1.46, -0.23	0.88, 0.01	0.67, 0.45
	new	0.61	0.44	0.39	0.17	0.66	0.37	-0.07	1.00	0.09	0.46
	Dk	0.60	0.38	0.37	0.17	0.62	0.31	-0.11	0.85	0.07	0.44
Anholt	old	0.47	0.41	0.58	0.59	0.44	0.18, 0.04	0.04, 0.07	0.28, -0.02	0.24, -0.01	0.13, 0.15
	new	0.59	0.55	0.58	0.48	0.45	0.05	-0.00	0.10	-0.01	0.14
	Dk	0.74	0.56	0.75	0.52	0.45	0.05	-0.00	0.11	0.04	0.06
Keldsnor*	old	0.41	0.31	0.47	0.52	0.26	0.54, 0.12	0.20, 0.21	0.83, -0.08	0.47, 0.15	0.61, 0.20
	new	0.44	0.47	0.57	0.41	0.14	0.14	-0.03	0.17	0.16	0.22
	Dk	0.51	0.51	0.67	0.41	0.19	0.14	-0.02	0.20	0.28	0.08
Lindet	old	0.39	0.49	0.34	0.55	0.60	1.14, 0.02	0.68, 0.19	1.67, -0.38	1.47, -0.42	0.97, 0.45
	new	0.55	0.33	0.63	0.56	0.26	0.36	-0.14	0.92	-0.07	0.69
	Dk	0.71	0.38	0.76	0.59	0.42	0.28	-0.13	0.78	0.41	0.19
Tange	old	0.46	0.55	0.57	0.56	0.10	1.20, -0.14	0.55, 0.16	2.14, -0.84	1.32, -0.37	0.81, 0.46
	new	0.55	0.45	0.58	0.38	-0.10	0.32	-0.13	0.55	-0.01	0.85
	Dk	0.77	0.50	0.76	0.51	0.05	0.33	-0.11	0.78	0.42	0.26
Ulborg	old	0.60	0.61	0.72	0.83	0.56	0.65, 0.69	0.18, 0.82	1.27, 0.36	0.79, 0.40	0.44, 1.15
	new	0.66	0.42	0.73	0.65	0.33	0.77	0.25	1.45	0.42	1.07
	Dk	0.78	0.50	0.77	0.77	0.35	0.79	0.26	1.70	0.82	0.51

Table 4. Correlation coefficients, observed mean and modelled bias between measured and modelled sum of NH_3 and NH_4^+ (NH_x) before and after implementation of the new dynamic NH_3 emission inventory for northern Europe. Two steps have been used in the implementation of the new dynamic emission model. In the first step data (new) from the RAINS model have been used to distribute the national emissions in each grid cell of the high resolution domain. In the second step (dk), the previously presented detailed Danish emission distribution on the 15 emission functions presented in Table 1 in (Skjøth et al., 2004h; Skjøth et al., 2008a) has been implemented as a supplement to the RAINS data. Calculations performed with the DEHM model for the year 2007.
* At Keldsnor measured values from three days in October 2007 have been excluded here as they were unusual high and probably represented very local emissions.

	Correlation coefficient						Mean meas. concentration [µg/m^3], Bias [µg/m^3]				
	type	year	winter	spring	summer	autumn	year	winter	spring	summer	autumn
Zingst	old	0.65	0.60	0.57	0.65	0.76	1.76, -0.78	1.33, -0.49	2.41, -1.13	1.54, -0.87	1.74, -0.60
	new	0.59	0.53	0.67	0.61	0.42	-0.79	-1.03	-0.68	-0.84	-0.64
	Dk	0.60	0.53	0.67	0.63	0.43	-0.79	-1.03	-0.64	-0.81	-0.69
Langen-brugge	old	0.46	0.56	0.44	0.36	0.43	2.04, 0.08	1.69, 0.05	2.61, 0.08	2.10, -0.52	1.74, 0.69
	new	0.41	0.45	0.43	0.34	0.22	0.19	-0.98	1.59	-0.42	0.53
	Dk	0.41	0.45	0.44	0.34	0.23	0.19	-0.98	1.60	-0.43	0.52
Wester-land	old	0.71	0.68	0.78	0.71	0.74	2.02, -1.24	1.49, -0.84	2.50, -1.41	2.31, -1.75	1.74, -0.94
	new	0.66	0.63	0.81	0.68	0.42	-1.23	-1.24	-1.00	-1.69	-1.00
	Dk	0.70	0.64	0.81	0.70	0.50	-1.21	-1.23	-0.90	-1.58	-1.14
Neuglob-sow	old	0.39	0.63	0.13	0.20	0.33	1.55, -0.07	1.29, 0.04	2.06, -0.18	1.40, -0.37	1.44, 0.25
	new	0.42	0.45	0.50	0.12	0.11	0.06	-0.76	1.08	-0.28	0.18
	Dk	0.42	0.48	0.48	0.14	0.12	0.00	-0.80	0.94	-0.29	0.15
Anholt	old	0.66	0.62	0.64	0.56	0.73	1.09, -0.69	0.89, -0.61	1.55, -1.00	0.90, -0.64	0.99, -0.49
	new	0.53	0.47	0.57	0.58	0.44	-0.71	-0.80	-0.86	-0.62	-0.55
	Dk	0.61	0.53	0.68	0.58	0.49	-0.60	-0.76	-0.56	-0.51	-0.54
Keldsnor*	old	0.65	0.61	0.64	0.57	0.64	1.88, -0.82	1.44, -0.69	2.71, -1.30	1.60, -0.78	1.78, -0.53
	new	0.56	0.45	0.70	0.50	0.31	-0.86	-1.10	-1.08	-0.75	-0.56
	Dk	0.65	0.61	0.64	0.57	0.64	-0.82	-0.69	-1.30	-0.77	-0.53
Lindet	old	0.43	0.45	0.48	0.59	0.49	2.46, -1.08	2.06, -0.99	3.29, -1.70	2.72, -1.56	2.05, -0.38
	new	0.49	0.23	0.65	0.67	0.27	-0.77	-1.42	-0.35	-1.18	-0.19
	Dk	0.61	0.26	0.75	0.69	0.39	-0.85	-1.40	-0.49	-0.66	-0.70
Tange	old	0.54	0.52	0.67	0.52	0.48	2.24, -0.95	1.64, -0.74	3.53, -1.86	2.14, -1.10	1.69, -0.14
	new	0.52	0.25	0.60	0.44	0.07	-0.50	-1.09	-0.42	-0.71	0.21
	Dk	0.67	0.26	0.70	0.51	0.24	-0.48	-1.07	-0.17	-0.24	-0.41
Ulborg	old	0.66	0.57	0.77	0.84	0.52	1.65, -0.10	1.14, 0.05	2.71, -0.76	1.57, -0.30	1.35, 0.49
	new	0.59	0.42	0.67	0.73	0.26	-0.04	-0.62	0.41	-0.26	0.37
	Dk	0.67	0.42	0.69	0.81	0.38	-0.01	-0.60	0.67	0.17	-0.21

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