Dear editor, colleagues,

Below we address the reviewer comments and questions raised during the open discussion of the paper "Response of secondary inorganic aerosol concentrations and deposition fluxes of S and N across Germany to emission changes during high PM10 episodes in spring 2009". We would like to thank the reviewers for their comments. We think that their comments have improved the paper considerably.
We have listed all reviewer comments below. Answers are provided in Italics.

Anonymous Referee #1:
This manuscript explores the response of SIA concentrations to changes in precursor emissions during high PM10 episodes in central Europe. This work is significant in that it considers these responses during spring time episode conditions rather than over long timescales previously reported by others.
The scientific quality and presentation of the manuscript is very good. There are only a few minor points to raise.

1) It would be benefical to understand how the model treats the dayly and seasonal profile of the ammonia emissions presented as an annual average in Fig1 c).
- *We have expanded the description of the emissions in section 2.1:*
"The temporal variation of the emissions is represented by monthly, day-of-the-week and hourly time factors for each source category. These factors were taken from the EURODELTA data base (Thunis et al., 2008). The applied monthly and hourly time factors for ammonia are shown in Figure 2. The seasonal variation in ammonia emissions is uncertain and may differ regionally as function of farming procedures and climatic conditions (Geels et al., 2012). The seasonal variation for Germany shows a distinct maximum in March/April due to the application of manure. The diurnal cycle in the emission follows Asman (2001) with half the average value at midnight and twice the average at noon."

Added to the reference list:
Asman, W.A.H. (2001), Modelling the atmospheric transport and deposition of ammonia and ammonium: an overview with special reference to Denmark, Atmos. Environ., 35, 1969-1983

Thunis,P. , Cuvelier, C., Roberts, P., White, L., Post, L., Tarrason, L., Tsyro, S., Stern, R., Kerschbaumer, A., Rouil, L., Bessagnet, B., Builtjes, P., Schaap, M., Boersen, G., Bergstroem, R.: Evaluation of a Sectoral Approach to Integrated Assessment Modelling including the Mediterranean Sea, Eurodelta II report., Joint Research Center, Ispra, Italy, 2008.

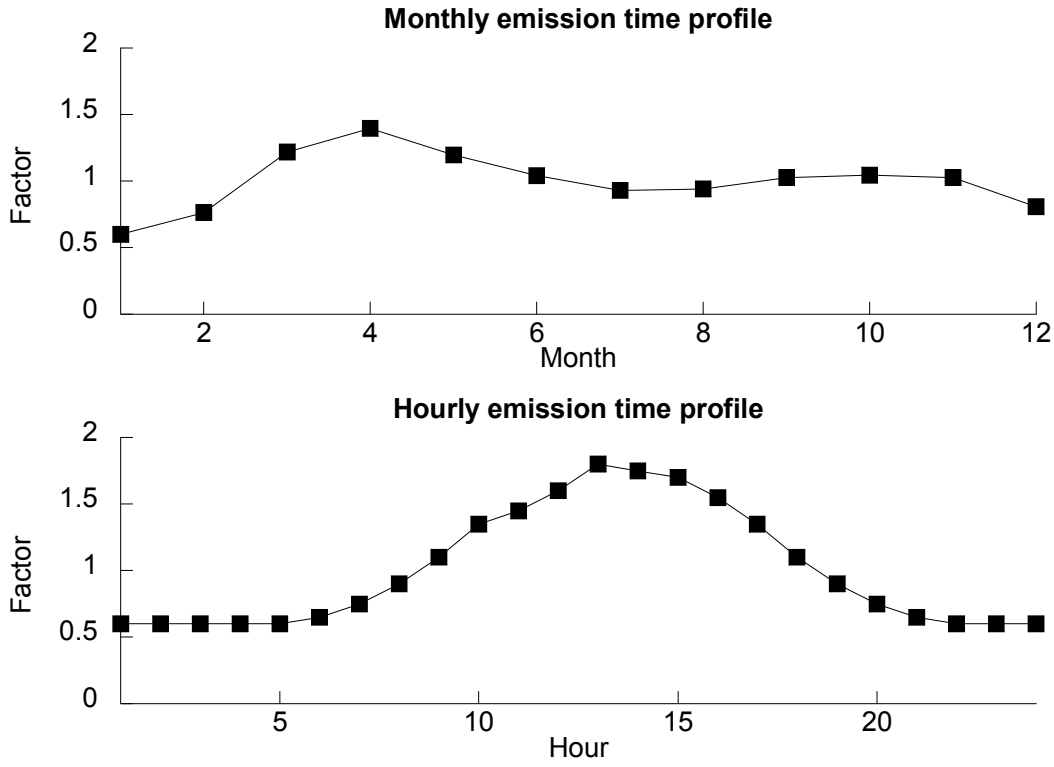

Fig. 2: (a) Monthly and (b) hourly emission time profiles used in RCG

2) It is assumed that the reduction scenarios and analysis in section 4.3.2 refer to the combination of the two episodes described in sections 4.2.1 and 4.2.2? It would be helpful to clarify this and indicate the modelled period the Figure captions and text.
- *As suggested by Anonymous Referee #1 the modelled period has been added to the text in section 4.3.2 (page 15796; line 23):*
"Concentrations and deposition fluxes are averaged over the investigation period from March 24th to April 28th."

and to the figure caption for Figure 9:

"Figure 9: Mean modelled (a) SIA concentration and (b) total deposition flux of the base run to the left of the dashed line and mean change in (a) SIA concentration and (b) total deposition flux for different emission scenarios to the right of the dashed line. Panels (a,b) show results for scenarios on the German domain excluding the boundary conditions and panels (c,d) those on the German domain including the boundary conditions. The mean refers to the average over the investigation period from 24th March to 28th April."

3) Given the non-linear nature of the model responses, some discussion of whether the authors feel that a similar result would be obtained by studying a period without an episode would be interesting, as would more discussion regarding comparison with the findings of annual average studies of this type referred to in the introduction. If the authors felt they had sufficent data the modelling period could be split into 'non-episode days', 'episode1' and 'episode2' and figure 9 could be calculated for the 3 cases. This would allow the effect of

the emission scenarios on the different cases to be evaluated and possibly some conclusion to be drawn on the effectiveness of different reduction scenarios on different types of episode. Alternatively this could be considered as an extension in future modeling where a number of suitable episodes could be selected for modelling.

- *We agree with Anonymous Referee #1 that the investigation of several episodes compared to non-episode periods would be very useful to be able to assess the effectiveness of different reduction scenarios on different types of episodes. However, we do not feel we have a sufficient amount of data to investigate "episode" and "non-episode" periods separately with our data set of only 5 weeks. The investigation of a number of selected episodes would indeed be a very interesting future task. We have added the latter to the text in section 5: "Therefore, a future study should expand the investigation period to several years/springs to better differentiate between episodes and non-episodes."*

In section 5 we have also added a more detailed comparison of our 5 weeks period investigation to 2 former annual average investigations:

"The response of SIA concentrations on NH_3 emission changes was found to be non-linear with an increasing SIA reduction effectiveness with increasing reduction of NH_3 emissions. The latter was also found in the German PAREST project in which scenarios were performed on an annual basis for the year 2005 (Builtjes et al., 2010). Tarrason et al. (2003) performed emission scenarios reducing German emissions of NO_x (-25%) and NH_3 (-25%) separately for the year 2000 using the EMEP model. The maximum reduction in annual mean SIA concentration following the -25% NH_3 emission reduction was about 1 µg m^{-3}. Considering that SIA concentrations peak in springtime, this is in good agreement with the maximum reduction in mean SIA concentrations of 1.3 µg m^{-3} for the -20% NH_3 emission scenario in the investigation presented here. In addition to the SIA reduction stated by former studies…"

Anonymous Referee #2:

General comments:

In this paper by Banzhaf et al., a CTM model is used to study the origin and characteristics of high PM events in Germany during spring 2009. By applying a number of emissions scenarios the formation of secondary inorganic aerosols (SIA) is studied in detail with the model. Especially the non-linear response in the chemistry and subsequent deposition fluxes of important air quality species is discussed. The impact of cloud chemistry in the model is also discusses. The issue of non-linearity in the response to emissions changes is very important in relation to e.g. planning of future national and international emission targets.

It is all in all a very interesting paper focusing on an important and complex scientific issue related to modelling of air chemistry. The paper gives a detailed description of the applied methods and analysis. The only negative point is the language, which seems to be somewhat influenced by the German language structure. The topic of the paper and the methods as well as conclusions is of relevance for the readers of ACP. I recommend the paper for publication after some revisions, see suggestions below.

Specific comments:

1) The title reflects the content of the study – but it is not very easy to read. I advise to find a shorter and more "fancy" title.

- *We agree with Anonymous Referee #2 and changed the title to:*

"Impact of emission changes on secondary inorganic aerosol episodes across Germany"

2) I am not native English speaking myself, but to me the language in the paper is a bit hard to read – the sentences are very long and "German". You should go through the paper and divide several of the sentences and/or include more commas. An example of such a sentence can be found on page 15802: "In these regions SIA formation is limited by the availability of NH_3 while in ammonia-rich areas SIA formation is limited by HNO_3 as even following a reduction of NH_3 a sufficient amount of NH_3 remains to neutralize the available HNO_3."

- *To improve the readability of the paper we have shortened several sentences throughout the manuscript. E.g. the sentence mentioned above by Anonymous Referee #2:*
" In these regions SIA formation is limited by the availability of NH_3 while in ammonia-rich areas SIA formation is limited by HNO_3. Following a reduction of NH_3 in these HNO_3-limited regimes a sufficient amount of NH_3 remains to neutralize the available HNO_3."

3) In section 4.3.2 you discuss and compare a lot of numbers – it would help the reader if these numbers were included in a table. It is e.g. hard to keep track on which numbers to compare in order to find the impact of the non-linearity.

- *As suggested by the Anonymous Referee #2 we have added the tables below to the manuscript in section 4.3.2:*

Table 5: Relative change in concentration compared to the base run for total SIA, SO_4^{2-}, NO_3^- and NH_4^+ for the German domain scenarios and the European domain scenarios

Scenario	Change in concentration on GD (%)				Change in concentration on ED (%)			
	SIA	SO4	NO3	NH4	SIA	SO4	NO3	NH4
-NOX-SO2	-4.6	-5.7	-3.8	-4.5	-23.1	-36.4	-10.2	-22.5
-NH3	-12.6	-5.9	-18.3	-13.2	-21.9	-9.4	-32.7	-22.9
-NH3-NOX-SO2	-15.8	-10.5	-20.2	-16.1	-39.1	-42.2	-35.3	-40.0

Table 6: Relative change in deposition of S and N compared to the base run for the German domain scenarios and the European domain scenarios

Scenario	Change in deposition on GD (%)		Change in deposition on ED (%)	
	S	N	S	N
-NOX-SO2	-13.9	-4.4	-42.6	-8.8
-NH3	-6.5	-15.2	-8.7	-19.8
-NH3-NOX-SO2	-18.9	-20.3	-47.5	-29.9

4) Fig. 9. Why not include the base case directly in the plot (and not only the numbers). It might help the reader to "visualize" the changes.

- *As suggested we have included the base case in the plot as shown below. By means of the new plots the reader can relate the impact of the different scenarios to the average base case SIA concentrations and S and N deposition fluxes.*

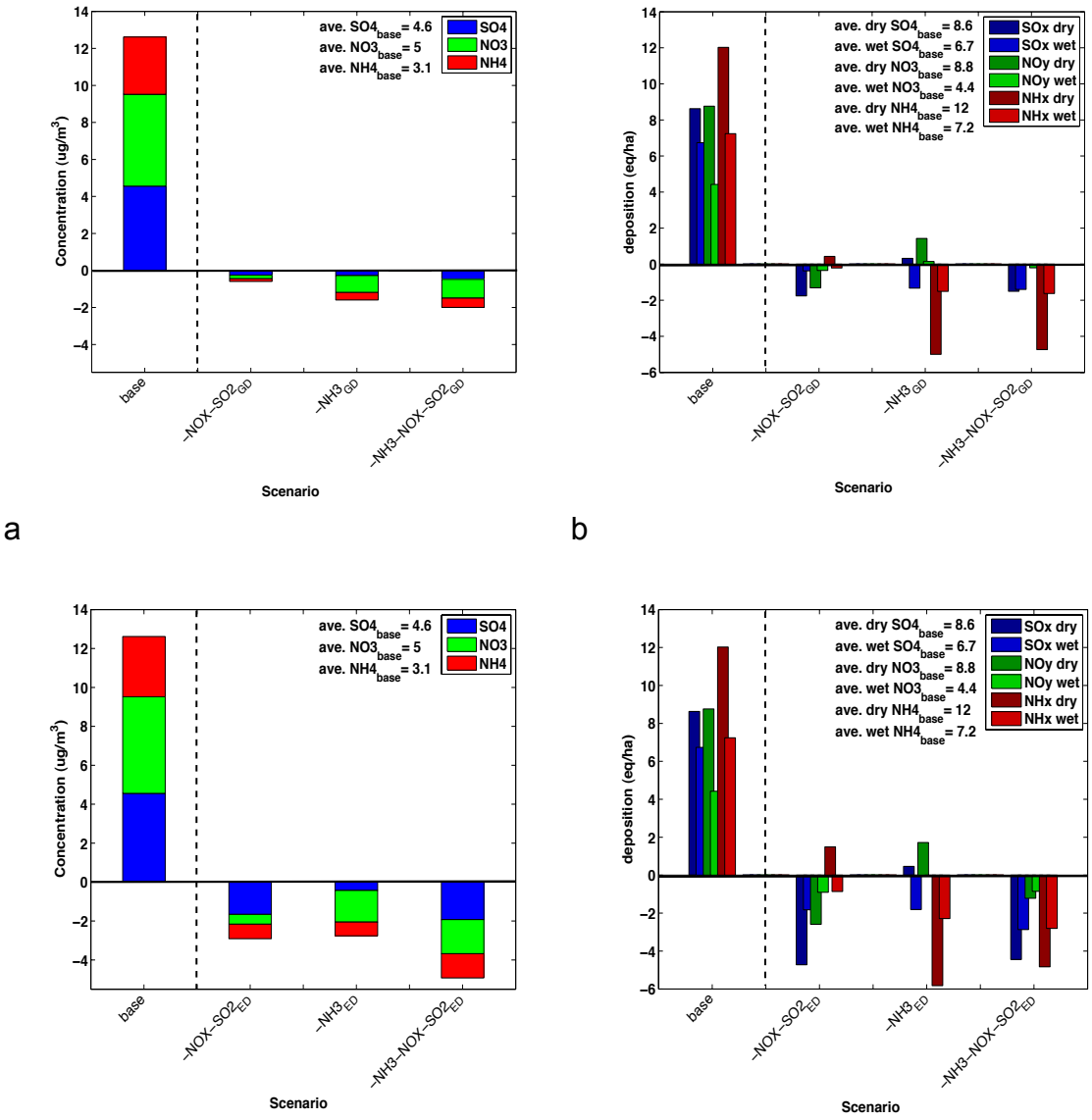

Figure 9: Mean modelled (a) SIA concentration and (b) total deposition flux of the base run to the left of the dashed line and mean change in (a) SIA concentration and (b) total deposition flux for different emission scenarios to the right of the dashed line. Panels (a,b) show results for scenarios on the German domain excluding the boundary conditions and panels (c,d) those on the German domain including the boundary conditions. The mean refers to the average over the investigation period from 24th March to 28th April.

5) As also mentioned by the other reviewer, a few more details on the applied emissions setup (e.g. temporal variation) would be appropriate.

- *See answer to comment #1 of Anonymous Referee #1.*

Technical corrections:

5) p. 15789. Lin. 16 "..the German and: : :" change to "..the German domain and : : :"
6) p. 15802. Lin. 6. "Fagerli et al., 2008" should be "Fagerli and Aas : : :."
7) p. 15806. Lin 28. "region" -> "regional"
8) p. 15807. Lin. 1. "The sensitivity 1" remove "1"?
10) p. 15811. Lin 12. Remove "1"?
- *All technical corrections above have been adopted.*

9) p. 15810. Lin 9. Delete "ten"?
- *The co-author is called Harry ten Brink.*

S.L. Napelenok (Referee):
This manuscript details a study of model based, brute-force emissions reductions of inorganic PM precursors and subsequent impacts on PM concentrations over the "German zoom domain…. [that] inevitably…. comprises parts of neighboring countries and seas" (this phrasing made me smile for some reason). The most interesting part of this manuscript to me was the analysis of the dependency of PM on cloud droplet pH, and I was disappointed that more discussion and model simulations was not provided on this topic. The manuscript is well written and generally clear, but could use some minor editing for punctuation, some awkward phrasing, and casual language. I believe it warrants publication in ACP, but I have a few suggestions for the authors that may be used to improve it.

1+3) The period chosen for the study was reportedly very dry with "precipitation amounts….far below average," and very high sunshine duration. These conditions seem very conducive to PM formation, but not for analysis of aqueous chemistry, since, presumably, there were not very much cloud cover. Again, I believe the pH sensitivity is an important topic for research, so how robust is your analysis from this dry period? On the same note, what fraction of sulfate was oxidized in cloud water versus dry gas chemistry during this period?
What was the average droplet pH in the modeled pH case, or how far away was it from the constant of 5.5 and was there significant variability?
- *We agree with Mr Napelenok that the pH dependency is an issue that deserves further attention. The sensitivity to pH should be investigated for different seasons, and preferably longer time periods to explain the observed trend. In our case we look at a particular month with two episodes. In the text we do not provide any information on the cloud cover during this period. We consider this to be an omission, as we do not like to imply that there were no clouds during this study period. Although the study period was very dry, cloudy periods were present. In fact, during the first episode the cloud cover across the SIA source region northern France (3rd of April) and Germany (4th of April) was largely between 70-100%. During the second episode the cloud cover was lower over Germany with large parts of central and north eastern Germany being cloud free. However in south western Germany as well as Belgium and northern France the cloud cover was 40-100%. We added this information to the*

description of the episodes in section 4.2.1 and 4.2.2 to clarify, that clouds were playing a role.

Mr Napelenok regrets that we did not provide more information on the pH dependency within this study. We have used the comparison to a constant pH simulation to add a feature with noticeable information to the paper. It was motivated by our earlier work to incorporate the pH dependent cloud chemistry instead of a constant pH scheme. This work has been published in Banzhaf et al. (2012) and incorporates the discussion and data the referee asks here. Hence, we like to refer to this paper for a more detailed analysis of the pH values and impacts. Unfortunately, we have limited computing resources at FU-Berlin and for these high resolution scenarios we did not output pH values or budget calculations. Our previous works showed that the droplet pH is quite variable within the model simulations and that providing an average is not useful. The modelled cloud water pH values range between 3 and 8. The range shows why it is not sensible to use an average, constant pH in model simulations as the formation efficiency of SO_4^{2-} is highly sensitive to pH.

2) The emissions reductions scenarios were examined entirely in the context of the model, which was validated using reasonable methodology for the base case. Is it possible to use the speciated observational data without regard to their spatial and temporal aspects to check if the responses simulated by the model are even possible, particularly for the extreme reduction cases? In other words, if you are modelling specific amounts of precursors at some site (including low ammonia for instance) that lead to specific speciated inorganic PM concentrations, is there some other place in Germany that has the same ratios of precursors and PM that was observed to compare? Seems like there needs to be additional grounding of the results in reality if at all possible.

- *Within Europe quite contrasting regimes are present with respect to SIA formation. European evaluation studies, e.g. EURODELTA (Vautard et al., 2009), has shown that RCG is capable to reflect the major gradients in Europe with respect to SIA, though best model performance is found for central Europe. Even within Germany relatively large gradients in ammonia and nitrate levels exist. RCG is able to capture those gradients as evidenced by evaluation studies such as PAREST (Builtjes et al., 2010) and Mues et al. (2012). These regions reflect areas with very different ammonia emissions, as already the difference between north-western Germany and other parts of Germany can easily reach a factor 5. Still we agree that a dynamic evaluation is needed which is now stated in the text in section 5. We are currently investigating the ability to model the 1990-2010 time period in which similar or larger emission reductions or changes in ammonia to SO_2 ratios occurred as described in this paper. This analysis will provide additional information on the model performance:*

"In addition, a dynamic evaluation should be performed to investigate the ability of the model to correctly respond to emission changes. We are currently investigating the model performance for the 20 year period from 1990 to 2009 for this purpose."

4) I disagree that this work is more significant because it was performed over a shorter period (spring) compared to previous studies (year-long), because, logically, the yearlong

studies also analyzed the spring. Instead, it is probably significant because the shorter episode was studied in more detailed with several emissions scenarios.
- *As stated in the first sentence of the last paragraph of the paper: this study confirms the important role of ammonia….. Hence, we believe this study is complementary with earlier work. As requested by reviewer 1 we have added some additional lines on the comparison to earlier studies. Of course, we have highlighted the difference between our study and earlier work. Note that only few studies exist that cover a full year and that except two, these do not cover Germany. Hence, we emphasize the detailed evaluation of the episodes as SIA concentrations, most notably ammonium nitrate, contribute more to PM in a relative during episodes than at clean conditions.*

5) I would like to see a couple of sentences on how this particular model (RCG) is used in regulatory settings if at all. This would be helpful to connect your findings to actual policy process. Similarly, what are the current strategies for controlling this type of PM in Germany and are they on the right track according to what you found?
- *RCG is used within Germany to evaluate emission reduction strategies for the German government (PAREST; Builtjes et al., 2010). Moreover, RCG is one of the models that is used to benchmark the EMEP model against within the TFMM EURODELTA (Vautard et al., 2009) studies. This has been included in the text in section 2.1.*
Current strategies for PM-reduction so not focus on ammonia as the reduction targets are basically 0%. Hence, we feel that the German government (and the EU) should focus more on mitigating ammonia emissions as described in the final paragraph. We have highlighted the German connection in this paragraph in section 5 as requested. E.g.:
"EU ammonia emission targets for 2010 and 2020 given by the NEC Directive and the Gothenburg protocol are not stringent enough and do not force the European member states policy to act on ammonia emission reduction"