

## ***Interactive comment on “Simulation of nitrogen deposition in the North China Plain by the FRAME model” by Y. Zhang et al.***

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Received and published: 27 October 2011

We thank the referee for the comments. We would like give feedback both in general and in detail.

We accept the comments of Referee 2 that FRAME is a relatively simple modeling system by modern standards and our work could be regarded as a preliminary screening level study. We look forward to seeing the results of future application of complex models at a fine resolution in this region. We believe that the usefulness of the model should be assessed not purely on its degree of complexity but on its fitness for purpose and level of performance. Dry deposition is well known to vary on a diurnal and seasonal basis. However the aim of the study is to estimate annual nitrogen deposition. Use of appropriate average annual deposition velocities can achieve this with a

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reasonable degree of success.

The FRAME model has been compared with other Atmospheric Transport Models (including state of the art models such as CMAQ and EMEP4UK) over the UK. This included a detailed comparison of the models with measurement data on gas and aerosol concentrations in air and ion concentrations in precipitation from the national monitoring networks. There was no clear evidence of the complex models achieving an overall significantly better correlation with annually averaged measurements than a simple model. Different models gave better performance for the various chemical compounds. Notably, FRAME achieved one of the best correlations with measurements for gas concentrations (NO<sub>2</sub> and NH<sub>3</sub>) in this study (Carslaw, 2011). This may be due to the fine vertical resolution of 1 m near the ground for FRAME, which can be important for nitrogen compounds (such as NH<sub>3</sub> and NO<sub>x</sub> which are emitted at low level). The report from this study is in the public domain and we invite the reviewer to read the contents: [http://uk-air.defra.gov.uk/library/reports?report\\_id=652](http://uk-air.defra.gov.uk/library/reports?report_id=652). (Figures and Tables of the correlation and statistics were attached in appendix 1 in the supplement)

An optional representation of the bi-directional exchange of ammonia is included in FRAME and this has been reported by Vieno (2005) for the UK. The results showed that permitting a net emission of ammonia from the surface resulted in a small elevation of ammonia concentrations. However this only significantly influenced average annual ammonia concentrations in remote regions with very low ammonia concentrations. The difference between use and non-use of bi-directional exchange did not significantly alter the correlation of the model with annual average ammonia concentrations. As the concentrations of ammonia in the North China plains are several times higher than in the UK, we expect bi-directional exchange to be even less significant in this region.

There is very little reported in the published literature on nitrogen deposition modeling in this region. Our work is a preliminary study intended to focus interest on environmental issues in a global nitrogen hot spot. The data bases applied to support the model application are clearly not as highly developed as in North America or Europe.

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However, this study offers useful preliminary estimates of the scale of the issue of nitrogen deposition in the North China Plains. We need a model which works effectively and can be supported by the current database, to start this work, rather than wait for model input data and measurement data which is as detailed as an American or European database. We agree that the results of this work are limited by the shortage of other modeling results to compare. However, it is important to note that it is exactly the limited application of atmospheric transport models in this region to date which is the primary reason for us to conduct this work. This is the first time a model has been applied to fully calculate the emission, transportation and deposition processes and estimate the potential exceedance of N-compounds at a high resolution in this global hotspot region. We consider that the results of this study are important not only to inform policy makers on abatement of pollutant emissions and ecosystem protection but also to initiate further detailed studies.

Vieno, M. (2005) The use of an Atmospheric Chemistry-Transport Model (FRAME) over the UK and the development of its numerical and physical schemes, PhD thesis, University of Manchester

We have made revision lists to all the questions from the referee in detail. These are listed below:

Q: Abstract- "contributed nitrogen budgets" needs rewording

A: It was changed to "Contributions of N deposition budgets from the seven provinces in this region were proportional to their area ratios."

Q: Introduction- Note that Kim et al (2003) used a nested grid with a grid size of 8.9 km over the NCP. Unfortunately, their simulations did not cover the same year as this study making direct comparisons difficult.

A: Thanks for the suggestion. A sentence was added. "Kim et al. (2003) used a nested grid with a grid size of 8.9-km over parts of the NCP. Unfortunately, the simulation was

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conducted for the year 1996, which is difficult to apply to recent situations."

Q: Methods- Check p 8164, lines 5-7 for grammar

A: It was changed to "...Hubei, Liaoning, Inner Mongolia, as well as parts of the Bohai and Yellow Seas, bordering and within the model domain but may not belong to the NCP. The location, boundary and landuse of the NCP. ..."

Q: It was changed to "...Hubei, Liaoning, Inner Mongolia, as well as parts of the Bohai and Yellow Seas, bordering and within the model domain but may not belong to the NCP. The location, boundary and landuse of the NCP. ..."

A: We agree that other studies have shown closer correlation with measurements of concentrations of gases and nitrogen compounds in precipitation (i.e. see the Defra model inter-comparison for the UK referenced above). These studies benefit from more detailed inventories and mapping of emissions. We note that the conclusion includes a comment recommending further development of techniques for the spatial mapping of NO<sub>2</sub> and NH<sub>3</sub> emissions. We agree that regression coefficients do not give the full picture. A table of appropriate statistics, commonly used in such studies has been added (Table 3 in the text). The aim of the present study is to highlight the high levels of N deposition in a global hot spot, not to demonstrate close agreement of an atmospheric transport model with monitoring data. We do not have data available from a complex Eulerian model for this region. However we hope that such models will be applied to nitrogen deposition in the North China Plains in future. A comment to this effect has been added to the text

Q: - I'm not sure I really see the point of your comparisons of the NCP budget with those from other countries. The emissions, meteorology, land cover, etc in the NCP are quite different than those areas. The usefulness of this comparison is not brought out in your discussion.

A: Same to the question of Referee 1: Yes, the emissions, meteorology, land cover, etc

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in the NCP are quite different from those in the Europe. Here, we just conducted the comparisons to imply that the N deposition in the NCP is much higher than that in the Europe. N deposition in the NCP, UK and Poland were all simulated by the same model and for similar areas. As N deposition hotspot in the world, the averaged deposition rate was more than five times that of the EU27. That's why the comparison of averaged deposition values was carried out for the NCP and EU.

Q: - Page 8169, lines 14-18: "bi-exchanged" might be better expressed as bidirectionally exchanged. You acknowledge here the importance of the bidirectionality of the ammonia flux, but the model doesn't include this. I'm not sure how that then leads to some of your explanations for your modeling results. Also, all gases are modeled in the resistance paradigm with some connection to the canopy resistance, not just bidirectionally exchanged ones.

A: It was deleted. As we mentioned in the general reply to referees above, the bidirectional exchange of ammonia has previously been tested in FRAME. The results showed that whilst this can be important for seasonal variations in ammonia concentrations, there was not a significant change to mean annual ammonia concentration in source regions. This was found to be important only in remote areas with very low concentrations of ammonia. In the high ammonia concentration region of the North China Plains, we believe that this does not significantly influence the simulated annual average ammonia concentrations in this study. The annually averaged ammonia concentration and deposition can be represented effectively using the canopy resistance. We deleted "As a bi-directional exchange species".

Q: - Page 8169, line 20: I would think that HNO<sub>3</sub> deposition would play a very strong role in the oxidized nitrogen deposition and probably more so than NO<sub>2</sub> deposition. To only look at correlation with concentration and not consider the deposition velocity may lead to incorrect conclusions.

A: We agree with the comment that HNO<sub>3</sub> makes an important contribution to oxidized

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nitrogen dry deposition. Unfortunately flux measurements of dry deposition of gases are not available. Long term monitoring of dry deposition does not commonly occur in national air pollutant monitoring programmes. Deposition velocities are more typically measured during shorter term intensive campaigns at a limited number of sites. A co-TAG (Time Averaged Gradient) system has been installed at a site in typical farmland area in the NCP. Although data is not yet available, the instrument is intended to provide data on long term monitoring of deposition fluxes of gases, including HNO<sub>3</sub>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub>, NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup>. In practice gas concentrations are frequently used as a surrogate to compare with modeled concentrations. The FRAME model has been compared with concentrations of nitric acid in the UK (Carslaw, 2011) and found to give reasonable agreement. However we agree with the reviewer's comment that correlation with concentration does not fully validate estimation of dry deposition.

Q: - Page 8170, line 2: Shen et al (2009) did not measure deposition. They used monitored concentrations paired with a deposition velocity (inferential method) to obtain deposition estimates.

A: Yes, Shen et al.(2009) did not measure deposition velocity. We specially used "...cited much higher deposition velocities..." to explain that. Here, we point out Shen's work not for comparison but for dispelling readers' confusion. In the same region, the 'measured' results were much higher than the modeled results, which are mainly due to using different deposition velocity values. People will be confused with this. That's why the difference of deposition velocities was emphasized.

Q: - Page 8170, line17: "in spite of" – this sentence is unclear

A: It was changed to "...by acidification and eutrophication in north America and Europe in spite of few studies of critical levels available in this region

Q: - Page 8170, line 21: "this value" what are you referring to?

A: It was changed to "3 μg m<sup>-3</sup>"

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Q: - Page 8170, line 26: "upper scale" maybe should be "upper limit"

A: It was changed to "upper limit"

Q: - Section 3.4 in general: I'm not sure I understand why you chose the critical load of 30 kg/ha/yr when the cited studies for that number are for other ecosystems.

A: The critical load of N deposition is just used for evaluating natural and semi-natural ecosystems not for agricultural ecosystems. We select 30 kg N ha<sup>-1</sup>yr<sup>-1</sup> as the critical load and would like to show the potential risk of N deposition to water bodies (i.e. reservoirs, lakes, rivers, seas) and forests and grasslands surrounding North China Plain.

Please also note the supplement to this comment:

<http://www.biogeosciences-discuss.net/8/C3897/2011/bgd-8-C3897-2011-supplement.pdf>

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Interactive comment on Biogeosciences Discuss., 8, 8161, 2011.