

## Review of Fu et al. for Atmospheric Chemistry and Physics

### *General Comments*

In “Estimating NH<sub>3</sub> emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model”, Fu et al. present a recently-developed modeling framework in which agricultural activity can be parameterized by meteorological, crop, and soil data to provide more detailed ammonia emissions from fertilizer to an air quality model, which is applied to China. To drive the models, they aggregate important national datasets, such as fertilizer application rate per crop and region, which are important contributions to the scientific literature. They estimate total ammonia emissions from fertilizer use as being much lower than other studies that employed emissions factors from the US or UK. The ammonia emissions estimates from this work are similar to bottom-up estimates from the national statistical database in spatially and temporally. Additionally, the emissions are consistent with meteorological fields that drive the air quality model, which calculates the ammonia emissions rate as it progresses through time. The bi-directional flux of ammonia is included in CMAQ. The comparison of nitrate observations collected at three urban locations is used to demonstrate the utility of employing the revised modeling system.

Given the importance and challenge of estimating atmospheric ammonia concentrations, particularly in a country seeking to reduce concentrations of aerosol, this advancement in integrated modeling of the tightly-linked agricultural and atmospheric processes is an important contribution to the scientific literature. Nevertheless, some clarifications and additional information are necessary to increase the utility of the work to the reader. Thus, I would support publication in Atmospheric Chemistry and Physics after minor changes and responses to the comments included below.

#### **1. Application of EPIC model to China**

The authors explain the lower emissions total from this study as being explained by US and UK emissions factors being applied to China (Sect. 3.2.2). In this study, EPIC is described as simulating “a wide range of vegetative systems, tillage systems, and other crop management practices” (p.750, l.25-6). To what extent are these methods specifically reflective of practices in China (versus the US) since the model was developed by US researchers for application in the US (originally)? Are there sensitivity tests that could be conducted to examine how much parameters that are known to differ between cultures influence the fertilizer applied? What extensions might be added in the future to more accurately represent farming practices in China?

#### **2. Soil characteristics**

The pH of the soil will have a significant impact on the partitioning of ammonium to ammonia. Since the cited website and associated data manual are in Chinese, the reader will be helped by an explanation in English in the paper of the method of estimating the pH of the soil across the country.

It is mentioned that some soil data are from the US soil profile. Which soil parameters are from this database? Why is it reasonable to use the US soil characteristics in these cases? How might these gaps in the Chinese database motivate future research in China?

Also, does the 25-year spin up period in EPIC alter soil pH and other soil characteristics from the input parameters?

### **3. Evaluation of model coupling**

The authors state that two simulations were conducted “to evaluate the performance of this NH<sub>3</sub> emission, fate and transport model”, but the description of the distinctions of these two modeling scenarios is incomplete, which leaves confusion about the intention of the comparison as well as the utility of it.

The base case is indicated to use the Zhao et al. (2013) emissions inventory. Does it include the bi-directional flux algorithm in CMAQ? If not, the authors would ammonia emissions to influence atmospheric concentrations differently from the second model run simply because the ammonia can be re-emitted once deposited.

The bi-directional case is described as using ammonia emissions from fertilizer that were calculated online CMAQ. Given the name of the case, it is assumed that this includes the bidirectional treatment, but clarification would be helpful for the reader. If the distinction between the two scenarios is not whether the bi-directional algorithm is included but rather the method of estimating agricultural ammonia emissions, this case should be renamed to indicate that distinction.

In addition to clarifying the distinctions, it would be helpful to explain the purpose behind the choice of model configurations in the two cases. Is the base case designed to reflect what others might model without the capabilities that these authors have added to the CMAQ framework?

The locations at which aerosol were collected are, presumably, urban. Were both anions and cations observed by ion chromatograph? If so, were their relative abundances indicative of the sulfate being fully neutralized by ammonium such that the authors would expect ammonium nitrate to be the primary component controlling nitrate presence? Was sodium or another cation present in the samples sufficiently to suggest that nitrate may partition apart from the contribution of ammonium?

If it is not possible to evaluate whether sulfate would be fully neutralized in these locations through observations, this information should be available in the CMAQ grid cells representative of the observation locations, which would provide some indication of the relevance of these measurements to evaluating ammonia emissions.

### **4. Comparison with other emissions estimates**

The other studies to which the ammonia emissions estimates of this work are compared do not include the bi-directional flux of ammonia. Could the authors include an estimate (perhaps based on the two studies conducted in this work) how different they might anticipate the estimates of ammonia emissions in the other studies to be if they were calculated in accordance with the method used in this work

(i.e., bi-directional flux of ammonia)? Perhaps the change would be negligible, but even this information would be worth including in Section 3.2.2.

## 5. Uncertainty analysis

The authors note that previous studies (e.g., the national statistical database) likely has uncertainties. Had those authors provided confidence intervals on their estimates, error bars in the ammonia emissions estimates might be included in Figures 5 and 7. Similarly, when future studies cite this work, they would be helped by having estimates of the uncertainty due to select parameters (e.g., parameters in the bi-directional flux model mentioned in Section 3.4). If it is feasible for the authors to provide quantification of uncertainty in ammonia emissions by propagating uncertainty in some parameters, future research would certainly benefit from such an estimate.

### *Specific Comments*

#### **A. Abstract**

*Lines*      *Comment*

20          Add space before “Compared”; “researches” to “research”

#### **B. Text**

*Page | Lines*

*Comment*

748   5	“aerosol and nitric acid (HNO <sub>3</sub> ) to generate” to “and nitrate (NO <sub>3</sub> <sup>-</sup> ) aerosol, adding to the concentration of”
750   9,14	“agriculture” to “agricultural”
750   21	“modeled 36 km CMAQ” to “CMAQ”
750   24	“agriculture” to “agricultural”
751   2	“it’s” to “it is” (also on p.759 at line 16)
751   5	“next” to “next section”
753   3	Please provide a citation of personal communication.
755   2	“fraction of the crop” to “fraction of cell used for crop”
756   8	“kg grid <sup>-1</sup> cell” to “kg grid cell <sup>-1</sup> ”
758   15	“alkaline gas in the atmosphere, NH <sub>3</sub> ” to “positive ion in the atmosphere, NH <sub>4</sub> <sup>+</sup> ”

- 759 | 1            Why were July 1-19 not included in the observations? Is November selected to evaluate the performance at lower temperatures?
- 760 | 5            “researches” to “research”
- 760 | 19           “human activity has on food production with air-quality” to “human activity has on air quality through food production”
- 760 | 19           “with climate model” to “with climate models”
- 764 | 18           The Williams et al. (2008) citation is for APEX, not EPIC, even though in the text EPIC is the model mentioned. Please correct the reference.

### **C. Figures**

*Figure 2.* Please add the locations of the nitrate observations to the map.

*Figure 5.* It is nice that the authors mention uncertainty in the statistical database on p. 759, l. 17. Does the statistical database include any confidence interval estimates that could be included as error bars?

*Figure 7.* Given the importance of temperature and precipitation to the emissions rate as noted in the text, could an indicator of these variables be provided alongside the current results? One option would be to produce a single box-and-whisker plot as Figure 7a with temperature on the left y-axis and precipitation on the right y-axis against the months of the year on the x-axis so that the median, quartiles, and extremes of these important driving parameters would be evident as readers evaluate the ammonia emissions (perhaps as Figure 7b).

*Figure 7.* In addition to the suggested addition above, making the units on the y-axis Tg (consistent with Table 3) would assist the reader in reading this absolute scale.

*Figure 9.* Please consider replacing this column chart with five pie charts that show the fraction of NH<sub>3</sub> emissions from each province for each of the five studies being evaluated. These could be ordered as the province contributing the most to the least for each study (i.e., for each pie). As it is, the results are very hard to compare from each study. If the authors have a special purpose behind using the bar chart, please at least order the provinces according to the most to least fractional contribution according to this study.