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Geoscientific
Model Development
Discussions

Interactive comment on "Modeling atmospheric ammonia and ammonium using a backward-in-time stochastic Lagrangian air quality model (STILT-Chem v0.7)" *by* D. Wen et al.

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Response: We thank the reviewer for the comment. We have provided detailed, point-by-point responses to the comments and suggestion below.

The overall impression is that that the authors have done a very nice job, but also that the article lacks regarding two important parts: Description of the ammonia emissions is insufficient, and the other part is that this work needs to be compared and discussed with results obtained in work with similar models and methods.

Response: 1) the following description of NH_3 emission has been added to line 9 on

Page 2761:

One of the key objectives of NAESI was to improve the 2002 national inventory on NH_3 emissions, especially from agricultural sources, using updated, Canadian specific, agricultural activity data and emission factors. The updated inventory can therefore account for spatial variation due to regional differences in farming practices and climatic conditions for each livestock category, and temporal variation due to seasonally different agricultural practices or seasonally variable temperatures that have different effects on agricultural NH_3 emissions throughout the year (Ayres et al., 2009). Total Canadian NH_3 emission in 2006 is about 5 Mt, about 90% of which are from agricultural. More information about emissions from other source types can be found at http://www.ec.gc.ca/pdb/websol/emissions/2006/2006_canada_e.cfm.

and the following description was added to Line 21 on Page 2761:

Temporal allocations of emissions were performed by SMOKE using predefined temporal profiles, allowing SMOKE-processed emissions to represent diurnal, weekly, and monthly variations.

The following corresponding reference was added to the paper:

Ayres, J., Bittman, S., Sheppard, S., and Girdhar, S.: Sources of ammonia emissions. In: Lillyman, C., and K. Buset.: Canadian Atmospheric Assessment of Agricultural Ammonia, pp. 112–130. National Agri-Environmental Standards Technical Series Report No. 4-1, 2008.

2) The following text about comparison against a similar model's results was added in Line 23 on Page 2763:

In a previous study (Skjøth et al., 2004) using a similar model (ACDEP), correlation coefficients obtained for the years 1999-2001 for three sites in Denmark varied from 0.43 to 0.69 when measured and simulated diurnal mean NH_3 concentration were used. Correlation coefficients increased to a range of 0.83 to 0.93 when measured and simulated monthly NH_3 concentrations were used.

The following corresponding reference was added to the paper:

Skjøth, C. A., Hertel, O., Gyldenkœrne, S., and Ellermann, T.: Implementing a dynamical ammonia emission parameterization in the large-scale air pollution model ACDEP, J. Geophys. Res., D06306, 109, 1-13, 2004., doi:10.1029/2003JD003895

The authors have completely failed to recognize the rather similar work performed of a Danish research group more than 15 years ago. This Danish group used a similar type of model, the ACDEP model, and they even applied a near to identical chemical scheme (Hertel et al, 1995) including a parameterization of the formation of ammonium nitrate, other inorganic as well as organic nitrates. This model has been extensively used for studies to simulate ammonia concentrations (de Leeuw et al., 2003; Skjøth et al., 2002; Skjøth et al., 2004) and nitrogen depositions (Hertel et al., 2002; Hertel et al., 2003) and uses a recently open source model that dynamically handles ammonia emissions (Skjøth et al., 2004; Gyldenkœrne et al., 2005; Skjøth et al., 2011). The emission model is already successfully implemented in the DEHM (Brandt et al., 2012) and the Unified EMEP model (Berge, 2010) and underway in the EMEP4UK model (Reis et al, 2011). Furthermore, several reviews have highlighted the applied approach as a needed requirement for many model calculations with respect to ammonia and ammonium containing particles (Hertel et al., 2006; Hertel et al., 2012; Menut and Bessagnet, 2010). Other groups in Europe have also used similar but more simple methods as those that are based on the FRAME model (Fournier et al., 2004; Kryza et al., 2011; Zhang et al., 2011) that were originally developed for the English area (Fournier et al., 2004). Or the TREND model (Asman, 2011), applying a simple seasonal variation of the ammonia emission.

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nia and ammonium: an overview with special reference to Denmark, Atmos. Environ., 35(11), 1969–1983.

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Hertel, O., Christensen, J., Runge, E. H., Asman, W. A. H., Berkowicz, R., Hovmand, M. F., and Hov, O., Development and Testing of A New Variable Scale Air-Pollution Model - ACDEP, Atmos. Environ., 29, 1267-1290, 1995.

Please also note the supplement to this comment: http://www.geosci-model-dev-discuss.net/5/C1132/2012/gmdd-5-C1132-2012-supplement.pdf

Response: the following was added to Line 23 on Page 2747 to address the issue:

Extensive efforts have been made to modeling studies of atmospheric NH_3 using different models either in Eulerian framework (Brandt et al., 2012; Berge, 2010; Reis et al, 2011; Wu et al., 2008; Makar et al., 2009; Sakurai et al., 2005) or in Lagrangian framework. Although the Eulerian approach is powerful and widely used for elucidating the chemical and physical mechanism in the atmosphere, the Lagrangian approach demonstrates key advantages in presenting sub-grid scale process, minimizing numerical diffusion, artificial dilution and computing resources. The Lagrangian approach has been widely adopted in various models in atmospheric ammonia modeling such as the FRAME model (Singles et al., 1998; Kryza et al., 2011; Zhang et al., 2011), the TREND model (Asman and van Jaarsveld 1992; Asman 2001), the ACDEP model (Hertel et al, 1995; de Leeuw et al., 2003; Skjøth et al., 2002; Skjøth et al., 2004; Hertel et al., 2002; Hertel et al., 2003; Skjøth et al., 2011; Gyldenkœrne et al., 2005), The TERN model (ApSimon et al., 1994), and the NAME model (Redington and Derwent, 2002). Most existing Lagrangian models for atmospheric ammonia modeling are either box-based models or use simplified dry chemical scheme. In this study, we attempt to model atmospheric ammonia using a stochastic time-inverted Lagrangian particle model in which a comprehensive dry chemical scheme (CB4) and a back-trajectory method are used. Plumes in Lagrangian particle models are represented by a large number of fictitious particles, which move with random trajectories to represent atmospheric turbulence. Particle models are able to account in detail for three-dimensional variations in the wind field and the effects of turbulent dispersion. High resolution and the improved accuracy of the vertical dispersion parameterization make these particle models particularly useful for simulating highly variable emission rates in complex dispersion scenarios.

The following corresponding references were added to the paper:

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Wu, S. Y., Hu, J. L., Zhang, Y., Aneja, V. P.: Modeling atmospheric transport and fate of ammonia in North Carolina - Part II: Effect of ammonia emissions on fine particulate matter formation. Atmos. Environ., 42 (14), 3437-3451, 2008.

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