Geoscientific Model Development Discussions

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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.

D. Wen et al.

dwen@uwaterloo.ca

Response: We greatly appreciate the valuable comments provided by the reviewer, which have helped us to revise and improve the paper. Our point-by-point responses are detailed below.

GENERAL COMMENTS It is useful to model the concentrations of NH3 and NH4+. The paper is in general well documented, but does not pay enough attention to the description of some important aspects of the components it focuses on. This makes it difficult to judge whether the model is appropriate or not. At least some important information is missing.

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Response: Based on the comments provided by this reviewer, we added more description on NH_3 emissions in several places. We also added comparison of our results with existing studies which used similar approaches. Section 4.3.2 was also rewritten in a more logical and smooth way.

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.

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.

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3) the following was added to Line 23 on Page 2747:

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.

SPECIFIC COMMENTS 1. The title of the paper is confusing. The trajectories are backward trajectories, but the chemistry and loss processes are forward. So leave "backward-in-time" out.

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Response: the title was changed to: "Modeling atmospheric ammonia and ammonium using a stochastic Lagrangian air quality model (STILT-Chem v0.7)"

2. p. 2755. Emission/dry deposition. Emission of NH3 occurs mainly from (near) surface sources. Dry deposition of NH3 occurs at the surface. In agricultural areas the NH3 concentration near the surface is much larger than the average concentration of NH3 over e.g. the lowest 50 m of the atmosphere. The authors should make clear what the limitations are of their model with regard to correctly modelling the NH3 concentration and dry deposition in source areas. If their model is unable to take into account the vertical resolution in source areas, e.g. near receptor points, the model is not very well suited for NH3/NH4+. It is in that case, however, maybe possible to come up with some correction factors.

Response: The model does make an attempt to introduce vertical variations. First of all, the turbulence strength is a function of height, with weaker turbulence strengths near the ground. Since the sensitivity of receptor concentrations to upwind surface fluxes (i.e., "footprint") is dependent on the amount of time each particle spends in the lower part of the mixed layer, the weaker turbulence strength would translate into longer residence times, and thereby stronger footprints. While proper simulation of the micrometeorology near the surface is difficult, we argue that it is a general weakness amongst all atmospheric chemistry models that attempt to cover the regional scale (as opposed to, localized models that rely on, e.g., large eddy simulations) and not limited to our approach.

3. p. 2761. It is referred to emission inventories. No information, however, is presented on the diurnal and seasonal variation in the emission rate, which is very important for NH3. I would suggest that this information is added. If no diurnal and especially seasonal variation is taken into account, then the article should be refused.

Response: the NH$_3$ emission inventory used in this study includes both diurnal and

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seasonal variations in the emission rates. Seasonal variability arises from accounting for different agricultural practices and meteorological conditions that differ between seasons. In addition, temporal allocation of NH_3 emission was also performed by the SMOKE modeling system, so the SMOKE-processed hourly gridded emissions input have included detailed information about temporal variations of NH_3 emissions at diurnal, weekly and monthly time scales.

To clarify this point, we have added the following description of NH_3 emission 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 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.

4. p. 2763. It is difficult to compare modelled and measured NH3 concentrations when no information is available of the emissions/spatial variation in the emission density near the stations. So it would be very useful if the authors could provide information on that. Figs. 6 and 7. It is difficult for me to understand what Figs. 6 and 7 actually

represent and what we could learn from that. It would be nice if the authors could explain that more clearly or otherwise the figures could be left out. What could be nice to look at is the contribution of emission areas as a function of the distance to the measuring site, e.g. sources from 0-1 km contribute for x1 %, 1-10 km x2 % etc.

Response: information about NH_3 emissions spatial variation in the emission density near the stations can be found in Fig. 1 even if the horizontal resolution of emission is somehow limited. Fig. 6 and 7 are not for comparing modeled and measured concentrations. Instead, they are to show the upstream influences from various locations and atmospheric processes to a specific receptor site. We completely regenerated Fig.6 and 7 to further clarify the spatial distribution of different processes on the receptor concentrations. In the original version of Figs. 6 and 7 the emission contribution from a particular gridcell was the gross contribution, but did not account for the loss along the transport pathway to the receptor. The new version of the figures shows the net contribution from each upwind gridcell on the receptor. In addition, as requested by the reviewer, we added a figure (Fig. 8) to show the contribution of emission areas as a function of the distance to the measuring sites. Section 4.3.1 was rewritten according to the updated Fig. 6 and 7 and new Fig. 8. Figure numbers after Section 4.31 has been adjusted.

Section 4.3.1 has been rewritten as follows:

Since the STILT-Chem model simulates an ensemble of air parcel back-trajectories that arrive at the receptor, the evolution of concentrations of modeled species can be calculated along each trajectory during every time step, for each process involved. This allows us to investigate upstream processes affecting concentrations at specific receptors. Figure 6, for example, shows calculated contributions and losses from different processes in upstream areas. Those values were obtained by averaging within each grid cell the values associated with different trajectories. The different processes either enhanced or reduced the concentration of NH_3 in air parcels that arrived at Egbert on 2 July 2006, at 18:00UTC.

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The calculated footprint (Fig. 6a; cf. Sect. 2.2) shows the main air flows that affected the level of NH_3 simulated at Egbert at that time. Fig. 6b shows source contributions of every upstream location to the NH_3 concentration arriving at the receptor at that time. Losses of the NH_3 concentrations at the receptor at that time due to dry removal, chemical conversion, and wet removal in each upstream location are presented in Fig. 6c, d, e, respectively. The NH_3 concentration was mainly enhanced by local sources. Only dry deposition occurred in a small part of upstream locations significantly reduced the NH_3 concentration because dry deposition can only take place within the surface layer. Compared against dry deposition, loss from chemical conversion took place over a more widespread upstream region due to the fact that chemical conversion can take place at higher altitudes rather than being restricted to the surface layer. Loss of NH_3 from wet removal is highly localized and dependent on precipitation rates in the upstream regions.

The results presented in Fig. 6 are only useful for investigating upstream sources or sinks influencing the receptor at one time. However, the same analyses can be averaged over a long time period to identify upstream sources and sinks that impact receptors significantly. As an example, the upstream contributions and losses caused by different processes as shown in Fig. 6– such as emissions, dry deposition, wet deposition and chemical conversion –were repeated for each simulation hour, and were then averaged over the entire six-month simulation period. The resulting averages are displayed in Fig. 7 for two sites, Longwoods and Chalk River, with very different characteristics (Fig. 1).

Figure 7 clearly shows that, averaged over 6 months, NH_3 concentrations at receptors were mainly influenced by air flows from west and southwest. Only sources and atmospheric processes in western and southwestern regions to the receptors greatly affected them. NH_3 concentrations at receptors were enhanced (represented in red color) by emissions from the upstream areas. Contributions from different upstream locations also differ significantly. The main source regions that impact Longwoods and

Chalk River are located in southwestern Ontario in Canada, Minnesota, Iowa, Wisconsin, Michigan, Indiana, and the northern part of Ohio, in the US. As noted earlier, Longwoods is representative of sites associated with extensive local agricultural operations and near strong NH_3 sources while Chalk River is a forested site surrounded by low emissions strengths. The strengths of the emission contributions of these areas to Longwoods were much higher than to Chalk River. This helps explain why the simulated and measured NH_3 concentrations at Longwoods were much higher than those at Chalk River. Dry deposition and chemical transformation are the major depletion processes (represented in blue color) of NH_3 in the upstream areas while wet deposition is less important. Southwestern Ontario, Lake Michigan, Lake Huron and northern Ohio were identified as important upstream areas for the loss of NH_3 concentrations due to dry deposition process. Higher loss from dry deposition near Chalk River was probably due to higher dry deposition velocity of forest in the vicinity of the site. Chemical conversion occurred at Southwestern Ontario, Michigan and Wisconsin was important for two receptors. The influence of wet deposition is mainly dependent on the precipitation amount and NH_3 concentrations in the upstream areas. Wet deposition in southwestern Ontario, Michigan, and areas in the vicinity of the two sites affects NH_3 at both sites; however, its influence is the smallest mainly because of the sporadic nature of precipitation.

In order to illustrate the relationship between sources contribution strengths and distances to a receptor, we calculated percentages of sources contributions and corresponding distances to the sites for each particle at each time step for the entire simulation period. The time-averaged contribution percentages were shown in Fig. 8 as functions of distances to two sites – Longwoods and Chalk River. Clearly, strengths of sources contributions to NH_3 concentrations greatly depend on their distances to the sites. Source contributions declined nearly exponentially with distances from a receptor. E-folding distances for the two sites were also presented in Fig. 8 (black dashed lines) to show the distances at which source contributions declined to 1/e of the values averaged in less than 10 km. The e-folding distance was about 90 km for Longwoods,

two times longer than that Chalk River, indicating that source contribution to Chalk River decayed more rapidly than Longwoods mainly because Chalk River is located in a relatively clean region where NH_3 emission contribution could be more easily consumed by other processes such as chemical conversion, dry and wet depositions.

5. p. 2768. "However, contributions from dry deposition are all negative, with smaller absolute magnitudes". This is not a relevant remark. It is always so that dry and wet deposition are sinks by definition. The only situation where this could be different is if the exchange with crops is modelled taking a compensation point into account and such a parameterisation is not chosen here. Chemical conversion is of course a loss for NH3 and a gain for NH4+. This is also very logical: this is put into the model and will therefore become visible in the results.

Response: the following text in Line 1 on Page 2768:

"However, contributions from dry deposition are all negative, with smaller absolute magnitudes. Contributions from dry deposition are within the range from -20 to 0 μgm^{-3}"

has been changed to:

"However, dry deposition losses are smaller in absolute magnitude varying from -20 to 0 μgm^{-3},"

6. It is also stated that the production of NH4+ is almost equal to the loss in NH3. This is again an irrelevant remark. They should be exactly equal as otherwise the model does not have mass conservation.

Response: the following text was removed from Line 8 on Page 2768:

"almost equivalent in magnitude to chemical loss of NH_3".

7. I feel that the wording is also a bit strange: "negative contribution". I guess it is more

normal to speak about loss.

Response: we modified all corresponding "negative contribution" to "loss".

8. So section 4.3.2 (and the figures belonging to that) should be rewritten.

Response: section 4.3.2 has been rewritten as follows:

Key atmospheric processes such as diffusion, deposition, and chemical conversion depend on meteorological conditions such as wind direction, wind speed, temperature, and precipitation. Thus, these processes may vary in upstream areas at different times, dynamically affecting the concentrations of a species measured or simulated at a receptor. In order to understand the relative importance of each process, total upstream contributions of each process to the simulated NH_3 and p-NH_4^+ concentrations at the Egbert site were calculated for each simulation hour.

The time series of different upstream processes are presented in Fig. 9, along with the simulated NH_3 and p-NH_4^+ concentrations (the net contribution of all those processes) for the purpose of comparison. Note that negative values here refer to concentration loss whereas positive values denote enhancement. We can see that the simulated concentrations and contributions from each process vary considerably with simulation time, due to changing upstream areas and different behaviors of atmospheric processes in those areas. With the exception of emissions, all other processes reduced NH_3 concentrations. Emission contributions to NH_3 at Egbert vary from 0 to more than 30 μgm^{-3}, with an average of 8 μgm^{-3}. The time series for dry deposition, another surface process, varies in almost the same pattern as from emission contributions. However, dry deposition losses are smaller in absolute magnitude varying from -20 to 0 μgm^{-3}, with an average around -4 μgm^{-3}, which is greater than from chemical conversion that varies between -15 to 0 μgm^{-3} with an average of -2 μgm^{-3}. Contributions from wet deposition depend on the amount of precipitation and are generally the smallest, with an average less than -1 μgm^{-3}.

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Unlike NH_3, the sole contributor to p-NH_4^+ is chemical conversion, with an average around 2 μgm^{-3}. p-NH_4^+ losses from both dry deposition and wet deposition are approximately within a range from -4 to 0 μgm^{-3}; however, the average loss from wet deposition is around -0.4 μgm^{-3}, about half of the average loss from dry deposition.

Figure 10 shows the average contribution or loss from each process over the entire simulation period at all six sites, derived from time-averaging the time series of different process contributions or losses (an example of such time series is Fig. 9 for Egbert). We can see that dry deposition and chemical conversion are the dominant processes in loss of NH_3, whereas p-NH_4^+ is depleted primarily by dry and wet deposition. Wet deposition plays a more significant role in loss of p-NH_4^+ than NH_3. The simulation results indicate that the level of NH_3 could be more than two times higher than the values seen in Fig. 3 if removal processes were absent.

The differences of upstream process contributions or losses to NH_3 are significant between the agricultural sites (Egbert, Longwoods, and St. Mary's) and the forest sites (Chalk River, Sprucedale, and Haliburton). On average, emission contribution, dry removal, wet removal, and chemical conversion to the agricultural sites are 2.5, 2.2, 1.6 and 1.7 times the values to forest sites, respectively. For p-NH_4^+, as a secondary pollutant with weaker spatial variability and a longer atmospheric lifetime, results indicate much smaller difference between the two groups of sites. Dry deposition, wet deposition and chemical conversion to the agricultural sites are 1.3, 1.1 and 1.7 times the values to forest sites, respectively.

We calculated the ratio of each sink (loss) to total sources (contribution) for each site using results displayed in Fig. 10, and the resulting values are shown in Table 4. Ratios of total sinks to total sources are calculated as well. Between the two groups of sites, the difference in ratios of total sinks to total sources is significant. The forest sites are on average 0.20 and 0.22 larger than the agricultural sites for NH_3 and p-NH_4^+, respectively. Out of the 0.20, the difference for NH_3, 65% is attributed to the difference (0.13) in ratios of chemical conversion to total sources. We suspect that

such a large difference resulted from significant difference in air temperatures due to different latitudes of the two groups sites (estimated from NARR dataset of the first layer (975–1000 mb), air temperature mean over the entire simulation period for the forest sites is about 2°C lower than the agriculture sites). In the chemical processes of NH_3, the $NH_3/HNO_3/NH_4NO_3$ equilibrium is very sensitive to the temperature (Stelson et al, 1979; Seinfeld and Pandis, 2006). An increase in temperature from 20°C to 30°C could increase the equilibrium gas-phase concentration of NH_3 and HNO_3 (equimolar) from 11.0 to 38.4 μgm^{-3} (Stelson et al, 1979), indicating significantly reducing the gas-to-aerosol chemical conversion of NH_3. For p-NH_4^+, the largest contributor to the difference in ratios of total sinks to total sources between the two groups of sites is dry deposition, accounting for about 64% (0.14) of total difference (0.22), mainly because the dry deposition velocities of p-NH_4^+ are generally larger for forest surface than crop surface (Zhang et al., 2001).

The following corresponding references were added to the paper:

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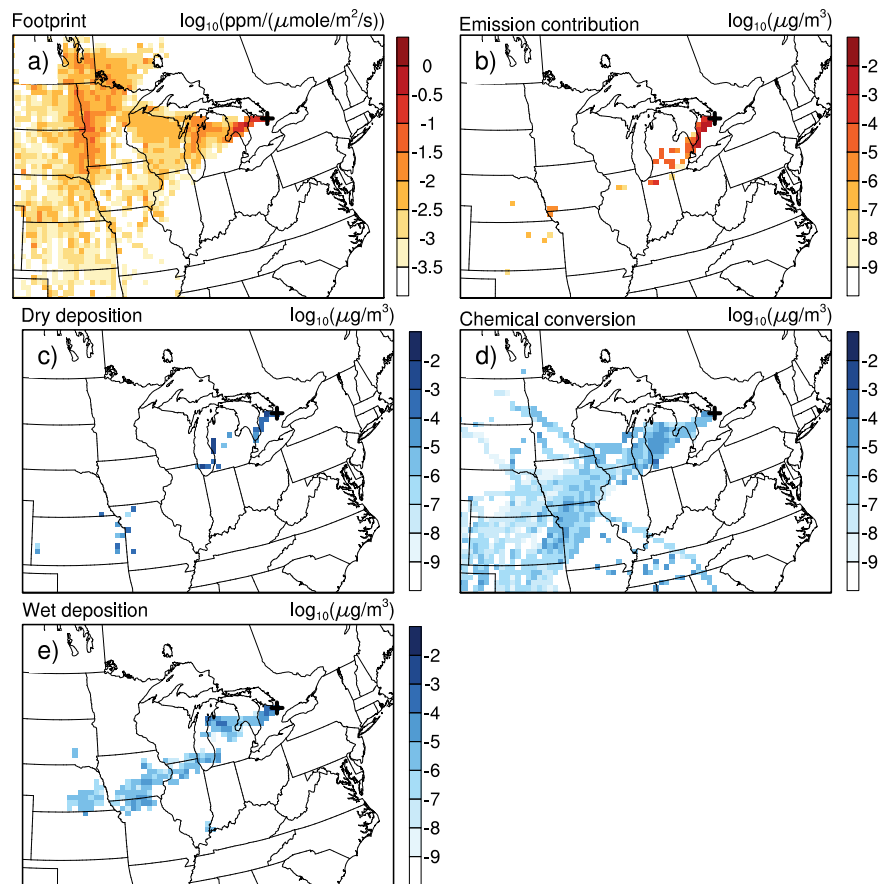Interactive comment on Geosci. Model Dev. Discuss., 5, 2745, 2012.
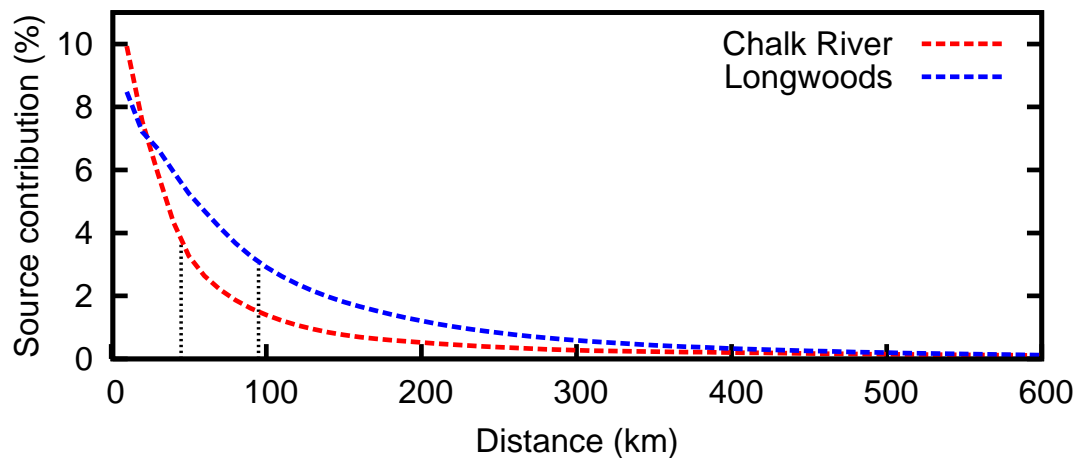
Fig. 1. Fig. 6 Modeled upstream parameters and processes impacting NH3 concentration simulated at Egbert (location indicated by "+") at 18:00 UTC on 2 July 2006.
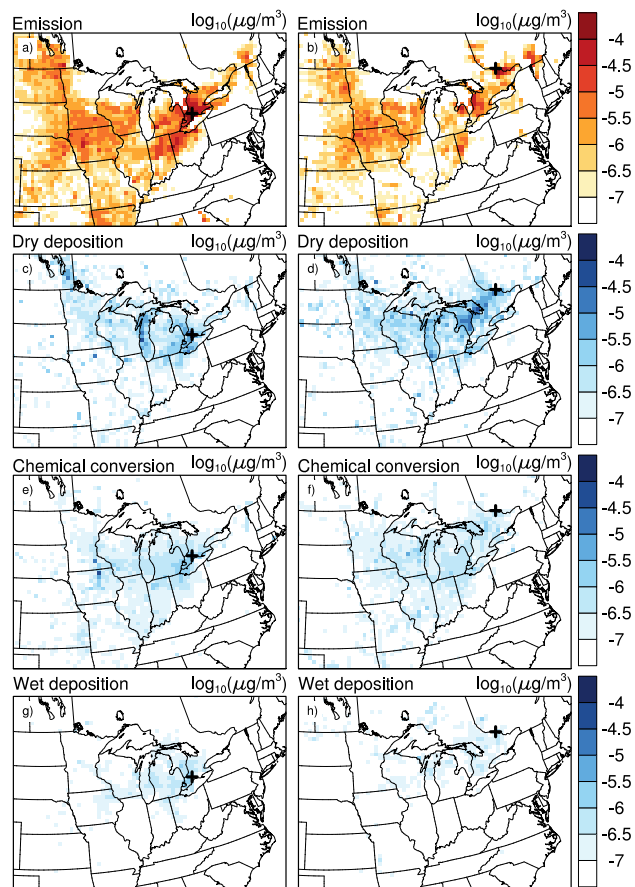
Fig. 2. Fig. 7 Modeled upstream contributions and losses to simulated NH3 concentrations from emission, dry deposition, wet deposition and chemical conversion for Longwoods (left) and Chalk River (right).

Fig. 3. Fig. 8 Source contribution percentages as a function of source distances to two receptors