Organic nitrates and other oxidized nitrogen compounds contribute significantly to the total nitrogen depositions in the United States

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It is well recognized that the NO_X ($NO + NO_2$) emission has been reduced steadily in the United States, but NH_3 emission is increasing strongly without regulation. Li et al. (1) estimate the NH_3 dry deposition by using the multilayer model (MLM) and bidirectional approaches, and conclude that the control policy for NO_X emission has now shifted the total (wet + dry) nitrogen deposition in the United States from the oxidized nitrogen-dominated to the reduced nitrogen-dominated pattern. The authors further conclude that the NH_3 dry deposition alone could contribute more than 50% to the total nitrogen deposition in some regions in the United States.

Although Li et al. (1) mention the effect of organic nitrogen compounds, they do not include it in their study. Previous studies (2–4) have indicated that organic nitrogen compounds could contribute 12–40% of the NO_y budget in the United States. Therefore, it is crucial to take these studies into account for the assessment of total nitrogen deposition. Although no observations are available for the organic nitrogen deposition, the atmospheric chemical modeling technique provides us an alternative way. Lamarque et al. (5) showed good agreement of NH_4^+ and NO_3^- wet depositions in the United States between the multimodel mean (MMM) results from global climate-chemistry models in the framework of the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) and observations. We thus use the MMM results from ACCMIP to calculate the total oxidized and reduced nitrogen depositions. The time-slice simulation starts at 2000, with 4- to 10-y data available, depending on the individual models. Fig. 1 indicates that, if the organic nitrates and other oxidized nitrogen compounds are taken into account, 86.64% of areas of the contiguous United States are dominated by the oxidized nitrogen deposition. When compared with Li et al.'s (1) work, the main difference occurs at the western and eastern coastal regions, where Li et al. suggest that NH_X deposition contributes more than 50% of total nitrogen deposition but MMM results suggest the fractional contributions of oxidized nitrogen in total nitrogen depositions are generally higher than 70%. On the other hand, Li et al. admit that the NH_3 dry deposition rates calculated by the MLM approach are a factor of 1.90 higher than those derived from the bidirectional model. Therefore, Li et al. could have overestimated the flux of NH_3 dry deposition over the United States but underestimated the contributions from oxidized nitrogen without considering the organic nitrates and other oxidized nitrogen compounds.

Although we strongly agree with Li et al. (1) that the reduced nitrogen deposition has become important in recent years in the United States, we do not think that it is appropriate to conclude that the reduced nitrogen deposition is already dominant without accounting for the organic nitrates and other oxidized nitrogen compounds. From the MMM results, we believe that oxidized nitrogen is still the dominant form of total nitrogen depositions in the United States so far. Further research for measuring organic nitrates is urgently needed to provide a more comprehensive insight into the total nitrogen deposition and its associated impact on the ecosystem in the United States.

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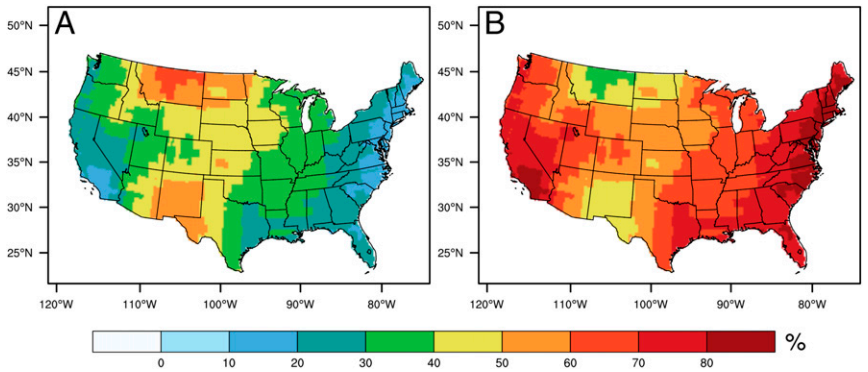

Fig. 1. The plots are made based on the MMM from ACCMIP by a time-slice simulation (4–10 y) starting at in 2000 (archived at badc.nerc.ac.uk/home/index.html). (*A*) The fractional total nitrogen from reduced nitrogen deposition ($NH_3 + NH_4^+$) to the total nitrogen deposition [$NH_3 + NH_4^+ +$ oxidized nitrogen ($NO + NO_2 + HNO_3 + HNO_4 + NO_3 + N_2O_5 +$ PAN + other organic nitrates)]. (*B*) The same as in *A*, but for the total nitrogen from oxidized nitrogen deposition. The unit is percentage.

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