

Interactive comment on “Organic nitrogen in PM_{2.5} aerosol at a forest site in the Southeast US” by M. Lin et al.

Anonymous Referee #2

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General comments

There is increasing evidence that organic particulate nitrogen makes a significant contribution to the organic aerosol mass and that organic nitrogen represents an important, yet mostly ignored, fraction of the total N deposition. There are few measurements of the organic N fraction, especially with automated measurement techniques. The coupling of a steam jet aerosol collector to a total N analyser introduced in this paper therefore makes a useful contribution both in terms of instrument development and application. I therefore support the publication of the manuscript, but only after several serious concerns have been addressed.

Major scientific comments

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I agree with Referee 1 that the quantification of LODs should be improved and that the results should be analysed and interpreted more fully, with less speculation.

The authors seem to suggest that HULIS dominates the C/N ratio of the organic aerosol at this site. This is both highly speculative and very unlikely, especially for PM_{2.5}. AMS factor analysis, for example, indicates that the aerosol is dominated by oxygenated organic aerosol, usually attribute to SOA production (see Zhang et al. 2007 reference above). Indeed, the authors use the C/N to argue that it is consistent with HULIS and later argue that it is consistent with organo-nitrates. Elsewhere they point out that the C/N ratio is dependent on wind direction. How does the C/N ratio compare with recent measurements by AMS (e.g. Aiken et al., 2007; Anal. Chem. 79, 8350-8358)?

In addition, I have another major concern that should be addressed: in general, the authors should discuss their results more fully in the context with other previous aerosol measurements at this site, e.g. by Aerosol Mass Spectrometry (Stroud et al., 2007; J. Atmos. Sci., 64, 441-459; Zhang et al. 2007, GRL, 34, L13801, including auxiliary material). In particular, I am concerned that these previous measurements identified significant amounts of nitrate during warm periods in July 2003 (0.38 $\mu\text{g m}^{-3}$) and September 2004 (0.1 $\mu\text{g m}^{-3}$), while no nitrate was identified in the present work. This makes me wonder whether the nitrate detection was working properly. Maybe NH_4NO_3 volatilised within the cyclone or NO_3^- was de-nitrified to NH_4^+ during sampling? Because ON is derived as the difference between total N and inorganic N, this would obviously feed through to the quantification of ON. In the light of these earlier measurements, temperature can clearly not be used as a reason why no nitrate was seen.

Minor scientific comments

P17159, L13. The authors may want to add non-US evidence, e.g. from Europe (Cape et al. 2005, Water Air Soil Poll. Focus 4(6), 1573-2940.

P17163, L6. The authors should discuss potential artefacts. For example, cation IC

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for NH_4^+ may be cross sensitive to amines and amino acids (cf. Husted et al. 2001, *Physiologia Plantarum*, 109(2), 167-179).

P17164, L7. Was the TN instrument also calibrated with organic N. This referee has found that organic N components can absorb to tubing, providing losses for ON which would not be seen for IN.

P17164, L22. It is counter-intuitive that the error on a difference measurements (ON) is smaller than the error on the component from which it is derived (TN, IN).

P17164, L24. What is the definition of a blank here? Is this a blank of the entire sampling system, e.g. operation without air pump and steamer?

P17165, L1. Presumably the charge balance should be tested including nitrate. Is it only because no nitrate was detected, that nitrate was ignored?

P17168, L11. The anticorrelation between ON and temperature could also be due to effects of boundary layer height dynamics which would also correlate with temperature. Similarly, the fact that in January NO, OC and EC show similar patterns could just be caused by boundary layer dynamics.

Technical comments

P17163, L13. 'in detail by Geron (2009)'

Table 1. Add units to caption. State averaging time. Why is the minimum concentration of ON not negative? The diurnal cycles indicate that negative values were derived (which is to be expected since it is a difference measurement).

P17167, L12. 'While elevated concentrations of ON ...'

P17169, L25. 'demonstrated that at this site'

Reference list: this needs careful formatting with sub- and superscripts where appropriate.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 9, 17157, 2009.

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