

Interactive comment on “Spatial and vertical extent of nucleation events in the Midwestern USA: insights from the Nucleation In Forests (NIFTy) experiment” by S. C. Pryor et al.

Anonymous Referee #1

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This manuscript aims to investigate the spatial and vertical extent of new particle formation events in the Midwestern USA, as well as the role of H₂SO₄, NH₃ and organics in new particle formation and growth. Although the study contains interesting results, additional work is required before the manuscript can be considered for publication in ACP.

Main Comments:

1.) The Information content of the paper does not justify the length of the article and attempts should be made to shorten the manuscript. There is a lot of unnecessary

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information that was already presented in a previous paper (Pryor et al., 2010). It seems that excerpts have been copied “word by word” from that manuscript (sections 3.32-3.34)! This is unnecessary! Please justify why the analysis of the back trajectories, nucleation parameter and condensation sink is presented again, though it was discussed in great detail previously (Pryor et al., 2010).

Further, the manuscript should be restructured. There is a lot of duplication/repetition in the manuscript which can be avoided. The discussion is scattered throughout the paper (parts in Chapter 2, 4 and 5). I recommend deleting chapter 2 and moving the relevant text to the introduction/method and discussion sections. Reorganize Chapter 4 and reconsider the number of figures (e.g. figures 12 and 13 needed?). Chapter 5 is simply an extended list of points already made in the main text. Delete and replace with conclusions! Any additional discussion of the results that is -at the moment- given in section 5 should be included in section 4. Table 1 and 2 could be moved to supplementary?

2.) The authors aim to assess the relative role of H₂SO₄, NH₃ and organic compounds in atmospheric nucleation. This would be a significant development but would require a systematic analysis of the correlation between the new particle formation rate and sulfuric acid (e.g. Kuang et al., 2008; Sihto et al., 2006) and organics (e.g. Kerminen et al., 2010; Paasonen et al., 2010). A more quantitative analysis of the data should be considered! Especially the use of the Fractional Aerosol Coefficients (FAC) to estimate the low volatile organic vapor concentration is questionable and needs further justification. As stated correctly, the approach neglects the availability of oxidants (OH/O₃) as well as the loss to the preexisting aerosol surface/mass. Why using it, especially since OH was measured? It is stated: “OH measurements for three nucleation days and two nonevent days show a substantial difference in OH distribution throughout the day”. Therefore I would expect that the true organic oxidation products vary accordingly (probably showing a diurnal cycle more similar to H₂SO₄). The lack of any correlation between FAC and particle number is not surprising and (most likely) mean-

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ingless for new particle formation! Since OH (and probably O₃) was measured, a more detailed analysis taking into account the oxidation capacity of the air mass and the loss to the preexisting aerosol could be conducted!

Specific/technical comments:

P23290, L19-24: "There are indirect indications that the growth in locales with high sulfur dioxide emissions is dominated by sulfuric acid (Sakurai et al., 2005; Stolzenburg et al., 2005; Petaja et al., 2007). Generally in less polluted regions the growth is dominated by other compounds (Boy et al., 2005; Wehner et al., 2005), but the role of organics in the growth can also be substantial in polluted environments (Smith et al., 2008).": Unclear, please rephrase!

P 23291 Lines 1-21: "but although regional new aerosol particle formation events are frequently observed at locations separated by up to 300 km, "they are rarely identical" (Hussein et al., 2009).": Unclear, please rephrase!

3-2.1: P 23296 line 20 – p 23297 line 9: The whole paragraph is confusing, please rephrase! The discussion on previous inter comparisons is irrelevant and misleading. P 23296,line 20 - p 23297,line 2 could be deleted.

Chapter 3.2.1: Why are Grimm and the CPC's not included in an inter comparison?

Chapters 3.3.2-3.3.4: Already presented in Pryor et al.,2010 (see comments above). If discussed, parameters could be introduced in the same way as the "condensation sink".

Chapter 4.1.1: Why no growth rates given for Indianapolis?

Chapter 4.1.2: The results presented in chapter 4.1.2 are closely related to chapter 4.3 (P23310-lines 1-26). Move to Chapter 4.3 to avoid unnecessary repetitions? Does the double event add any addition information?

Chapter 4.2: Back trajectories, reference to Pryor et al., 2010?

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Chapter 4.3: Subheadings would be very helpful. P23308 line 1- p23309 line 28 and Figure 12: The general relation between CS, NP was already discussed in Pryor et al., 2010 (see comments above).

P23309, lines 25: Correlation of those 5 data points seems to be questionable. What is the meaning of this correlation?

Figure 5a: Datapoints missing: H₂SO₄ before 13:00, NH₃ after 10am. Figure 5 lower panels: VOC data hard to read. Needs improvement.

Figure 14: needs improvement. Vertical bars indicating the variation in the composite data should be included. Figure 5 indicates some strong variation in ammonia throughout the day. This seems to average out in the composite data. Again vertical bars indicating the variations would be helpful.

In References/Citations: IPCC reference correct?, Plass-Dulmer>Plass-Dülmer, Vehkamäki >Vehkamäki, Petaja>Petäjä, Sipilä>Sipilä etc.

References Pryor, S. C., A. M. Spaulding, et al. (2010). "New particle formation in the Midwestern USA: Event characteristics, meteorological context and vertical profiles." *Atmospheric Environment* 44(35): 4413-4425. Kuang, C., McMurry, P. H., McCormick, A. V., and Eisele, F. L.: Dependence of nucleation rates on sulfuric acid vapor concentration in diverse atmospheric locations, *Journal of Geophysical Research-Atmospheres*, 113, doi:10.1029/2007JD009253, 2008. Sihto, S. L., Kulmala, M., Kerminen, V. M., Dal Maso, M., Petaja, T., Riipinen, I., Korhonen, H., Arnold, F., Janson, R., Boy, M., Laaksonen, A., and Lehtinen, K. E. J.: Atmospheric sulphuric acid and aerosol formation: Implications from atmospheric measurements for nucleation and early growth mechanisms, *Atmospheric Chemistry and Physics*, 6, 4079-4091, 2006. Kerminen, V.-M., et al. : Atmospheric nucleation: highlights of the EUCAARI project and future directions, *Atmos. Chem. Phys.*, 10, 10829-10848, doi:10.5194/acp-10-10829-2010, 2010 Paasonen, P., et al., : On the roles of sulphuric acid and low-volatility organic vapours in the initial steps of atmospheric new particle formation,

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