

Interactive comment on “Characterization of the size-segregated water-soluble inorganic ions at eight Canadian rural sites” by L. Zhang et al.

Anonymous Referee #3

Received and published: 11 September 2008

Review: Manuscript # 2008-270-1-01 “Characterization of the size-segregated water-soluble inorganic ions at eight Canadian rural sites”, Leiming Zhang et al.

Summary:

This paper describes the measurement of size-segregated water-soluble inorganic ions contained in airborne particulate matter at rural sites in Canada. Samples were collected using a cascade impactor and analyzed using ion chromatography. Eight to thirty-seven samples were collected at each location in 1-2 separate campaigns typically carried out in different seasons. The major conclusions in this study are that (1) the size distributions of SO₄²⁻, and NH₄⁻ were generally unimodal, peaking at 0.3-0.6

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μm during most of the campaigns, (2) size distributions of NO_3^- were generally unimodal during warm seasons, peaking at 4–7 μm , and bimodal during cold seasons with an additional peak at 0.3–0.6 μm , (3) Cl^- , Na^+ , Mg^{2+} , and Ca^{2+} had unimodal distributions during ~half the sampling events with a peak at 4–6 μm , and bimodal during the other events with one peak at 2 μm and the other peak at 6 μm , (4) K^+ was bimodal during most of the events, with one peak at 0.3 μm , and the other at 4 μm , (5) the measured ion concentrations varied by one order of magnitude across the sites, and (6) the emissions source and meteorological conditions played important roles in determining the particle size distributions.

This paper summarizes a careful study that measured the characteristics of the particles using rigorous quality control procedures. The authors are to be commended on the immense amount of work that they have performed measuring particle size distributions. However, it is not clear to me how the results can be used in future studies. The sampling patterns do not characterize "typical" conditions at any of the sites. By themselves, these measurements could not be used to estimate the annual average flux of acids to the ecosystem, nor could they be used to estimate annual average contributions to scattering and absorption for climate calculations. The results also do not seem to be characteristic of typical pollution events at any of the sites, or at least the reader was not informed how the measurements relate to episodic concentrations. The best use of the results would seem to be as model validation assuming an effort is underway to reproduce the measured concentrations using the Canadian modeling tools. In this case, the dataset would indeed be useful for future studies, and should be published. Detailed comments follow below.

Detailed comments:

1. Page 3 line 8. "Particle size distributions vary greatly with season, location, and air mass origin." This statement is true, but the introduction does not go far enough in describing how the current measurements are useful in light of this seasonal and emissions source uncertainty. Can the results be taken as typical for an annual

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cycle? The case has not been made for this in the analysis presented by the paper. Are the results typical of conditions during a particularly important episode? The most useful feature of the results would seem to be as a model validation dataset. If that is indeed the case, then a few details of the planned modeling exercise should be included in the introduction to explain to the reader why these measurements are useful for anything other than a snapshot on the day they were collected.

2. Page 2 line 23 the statement "Nitrate (NO₃⁻) was found to be in the fine particle model in some cases and in the coarse particle model in many other cases" is vague. Please avoid the use of qualitative descriptions. What cases have fine vs. coarse-particle NO₃⁻ in this previous study?

3. Page 6 Sample and Analysis section. Were the filters coated with anything to prevent particle bounce? How did the authors verify that bounce artifacts did not skew the size distribution and possibly lower the measured total concentrations?

4. Page 6 Sample and Analysis section. Were the Teflon filters wetted with any substance before extraction in water? Teflon is hydrophobic, which may reduce extraction efficiency.

5. Results section. Several sentences are presented for each water-soluble ion describing the source origin and general atmospheric processing. This information is generally well known to most environmental scientists. The authors may wish to consider shortening this section of the paper and making reference to suitable texts or previous papers.

6. Results section. My overall recommendation is that the results section should be compressed in size so that it emphasizes the most interesting results efficiently and skips some of the details associated with the more routine findings. I would recommend that Figure 3 be removed from the manuscript entirely since it doesn't present useful information. Likewise, I had a difficult time interpreting the importance of Figure 6. If the authors have an important point to make with this figure, then they should

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rewrite that section of the paper to emphasize the point. Figure 7 should be revised to only show the ratio of NH_4^+ to the molar equivalence sum of sulfate and nitrate. The comparison to sulfate alone is not very useful. Figure 8 needs significant revision to convey the information in a more compact format.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 13801, 2008.

ACPD

8, S6899–S6902, 2008

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