Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-1030-RC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical characteristics of size resolved atmospheric aerosols in Iasi, north-eastern Romania. Nitrogen-containing inorganic compounds controlling aerosols chemistry in the area" *by* Alina Giorgiana Galon-Negru et al.

Anonymous Referee #1

Received and published: 18 December 2017

This study analyzed the ionic components of size-resolved atmospheric aerosols sampled in northeastern Romania including ion balance, aerosol acidity, formation of ammonium and nitrate, and influence of relative humidity and air mass origins. Measurements of aerosol chemical composition in Eastern Europe are scarce; however, similar measurements at this particular site have been previously reported. The paper is long and I feel the discussions in the paper need to be written more concisely, e.g. especially the discussion of the formation of ammonium and nitrate. The tendency for ammonium

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nitrate to form at lower temperatures and higher relative humidity and the strong association between ammonium, sulfate and nitrate has been previously reported. The presence of strongly acidic aerosols is an interesting and potentially important result that could use more discussion on the acid rain implications. This paper is strongly focused on the chemical and physical processes driving the formation of secondary pollutants, but more consideration needs to be given to the gaseous precursors and where these emissions are coming from.

Specific Comments

P4, lines 31-34: Chemical composition and seasonal variation of water soluble ions at this site has been previously reported. Could you emphasize the new work that will be presented in this paper?

P8, lines 16-17: For long range transport, which region is contributing to PM? Can you specify what the local contributions might be at this site, e.g. commercial/industrial or vehicular emissions contributing to PM?

P8, lines 22-32: Could the differences in PM concentrations between this study and the previous study be due to the different particle size cutoff? The previous study reported PM1.5 and PM(>1.5), whereas this study reported PM2.5 and PM10. Could it also be due to sampling in different years in which there may be changes in the emission sources (e.g. emissions reductions)? It seems the difference in altitude of the sampling site (25m vs. 35m) is too small to account for the difference in PM concentrations. What was the altitude of the sampling site in Alastuey et al. (2016)?

P9, lines 22-24: “increased frequency of dust and also due to more intense anthropogenic activities in the neighbourhood of the sampling site” This result seems inconsistent with earlier results, which stated that PM was likely from long range transport instead of local contributions.

P9, lines 24-26: Why would dry deposition during the warm seasons be enhanced for

PM2.5 but not PM10?

P9, lines 30-32: I'm not sure how you arrived at the conclusion that the results are not due to emission sources but are likely due to meteorology based on the seasonal trends in the PM size distribution. Please explain how changes in the meteorological conditions and which met parameters affect the seasonal PM size distribution.

P10, lines 6-9 and other places: I suggest moving the detailed statistics in parentheses to a table or the supplement because it makes the rest of the text difficult to follow.

P10, lines 22-28: It's not clear what the missing NH4+ is referring to because NH4+ measurements are available. Please briefly explain why there is another fraction of NH4+ that is not measured by the IC data.

P11, lines 4-23: This text could be condensed or moved to the methods section or the supplement. This section should present the results of the model runs.

P12, lines 20-23: What are the ecosystem or human health implications of strongly acidic aerosols? I suggest providing a brief explanation on the importance of aerosol acidity. What is contributing to the strongly acidic aerosols, pH 0-3? At this aerosol pH, does it contribute significantly to acid rain in this region? Are there any reports of acid rain impacts in this region?

P13 line 29 – P15 line 9: This section examined NH3/NH4+ partitioning as a function of relative humidity. Considering that ammonium tends to be associated with sulfate and nitrate, I think that the discussion on NH3/NH4+ partitioning needs to consider changes in the sulfate and nitrate concentrations as well as temperature which affect both sulfate and nitrate production. Also, temperature and relative humidity are strongly correlated. Can you confirm that the relationship with relative humidity is not actually due to temperature? Do you still see the strong relationship between NH3/NH4+ and relative humidity when you analyze cold and warm seasons separately?

P14, lines 29-31: I think that this conclusion needs to be corroborated by examining

the seasonal trend in the ammonia emissions since ammonia concentrations were not available. Ammonia is typically higher during the warm seasons, but here you suggest it is high enough all year round.

P15, lines 13-16: Are these results expected? For the ions associated with fine particles, are the percentage increases lower than those ions associated with coarse particles?

P15, lines 18-21: Sulfate is usually higher during warm seasons because of increased oxidation of SO2. I'm not sure about the explanation of lower mixing heights or temperature inversions because not all of the pollutants were higher during cold seasons.

P17, line 5: Ammonia is usually higher during warm seasons because of higher temperatures and increased agricultural emissions. What are the sources of ammonia during cold seasons?

P17, lines 16-19: The relative humidity explanation for the high sulfate concentrations is not consistent with the lack of correlation between sulfate and meteorological parameters mentioned earlier (lines 12-13).

P22, lines 13-20: The molar ratios in Fig. 7cdef between cold and warm seasons are not very different and all the R2 values were close to 1. It seems that a complete neutralization of H2SO4 and HNO3 also occurred in the warm seasons. Can you explain the lack of difference in the molar ratios?

P27-28: This section can be improved by showing the inorganic ion concentrations associated with the different air mass origins identified in Fig. 1. While physical and chemical processes are important in the formation of secondary pollutants, it is also important to examine where the gaseous precursors e.g. SO2, NOx and NH3 are coming from. There might be a change in the air masses during different times of the year leading to seasonal variability in the gaseous precursors, which in turn affects the secondary pollutants.

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