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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of ions at Alpine waterfalls" *by* P. Kolarž et al.

Anonymous Referee #1

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The authors have studied the formation of waterfall-generated ions, or so called balloelectric ions, during three years of various field campaigns in the Austrian Alps. They have monitored the concentrations of waterfall-generated ions at five different waterfalls applying self-made Gerdien-type integral air ion counters and a modified Grimm SMPS aerosol spectrometer to cover the size range of 0.9–350 nm, from cluster ions up to large charged aerosol particles. The measurements showed high negative ion and moderate positive ion concentration gradients near the waterfalls, as expected of the generation of ions by balloelectric mechanism. The most important outcome of the study is the spatial distribution of the waterfall-generated ions, influenced by the local orography and meteorological conditions.

This work can be considered as one in the line of investigations of balloelectric ions started by Philipp Lenard (Lenard, 1982; 1915), continued by Chapman (1938a, b), Blanchard (1955), Gathman and Hoppel (1970), Levin (1971), Reiter (1994), Hõrrak et al. (2006), Hirsikko et al (2007), Laakso et al. (2006, 2007) and Tammet et al. (2009). The subject of the paper is certainly relevant to the aerosol researchers' community and also to the larger audience of the journal of Atmospheric Chemistry and Physics, giving an impulse to the further investigations of balloelectric ions. The authors have done a good and interesting job, which is worth to be published in the ACP. However, the present paper has also some shortages and can be accepted for the publication in the ACP if these problems are satisfyingly resolved. The authors are asked to consider the following general and specific comments, questions and recommendations.

General comments

We noted that the introductory part of the paper is quite short, the motivation and main objectives of the present work are missing. Also, too little attention is paid to the earlier investigations in this field. At least one important recent work on balloelectric ions by Tammet et al. (2009) is missing, but the contributions of some others, listed above, are also worth to be mentioned or introduced in some more detail. Thus, the authors are asked to consider giving their work a broader perspective with respect to other works in this field. Regarding an overview of atmospheric ions, we recommend including two recent review papers by Arnold (2008) and Hirsikko et al. (2011). The paper should also be structured in a more proper and reader-friendly way, concentrating passages of similar matters in appropriate sections, and inserting some additional tables for a better overview of the measurements and instrumentation. The authors are also asked to reduce the number of megabits in their 10.3 MB-long article by reducing the resolution of the photos.

A crucial problem is that the description of the self-invented and built Gerdien-type air ion counter, as well as the applied measuring method, is too short and some of its aspects are missing at all. Therefore, it is hard to understand how the size distribution of air ions was obtained from the integral ion counter measurements. So, we recommend adding a more detailed description of the measurement technique

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and measurement methods. We also recommend using a proper terminology of air ion classes (small or cluster ions, intermediate ions, large ions), introduced in page 25299, throughout the manuscript to avoid misunderstandings. Waterfall-generated ions cover the size range from about 1 nm up to 30 nm, from the cluster ion size range up to large aerosol ions; in some cases, the changes up to 350 nm in the size distribution can be seen. Therefore, the authors are asked to specify the ion group or size range of ions under discussion, instead of only using the term “WF ions”.

Specific comments

Specific comments are given below in the order of the paper’s sections (chapters).

Introduction

1.1 Air ions in the environment

Page 25298, lines 22-23. We cannot agree that the background concentration of air ions in the environment is about a few hundred per cubic centimeter. Probably the authors mean here only the cluster or small ion concentrations. For typical concentrations see Hirsikko et al. (2011).

Page 25299, lines 1-5. The authors should specify the category of ions they are talking about - cluster or small ions.

Page 25299, lines 5-7. The meaning of the sentence is hard to understand and it should be rewritten. We cannot understand how the lower mean mobility of positive cluster ions causes their lower concentration in the atmosphere. Due to the atmospheric electric electrode effect the concentration of positive cluster ions near the ground should be higher compared to the negative one. This is a generally known fact. The lower mean mobility of positive ions plays a secondary role here, it only results in lower attachment rates of positive ions to the surfaces (aerosols, ground surface,

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vegetation canopy, etc) compared to the negative ones. Also, the beta-coefficient is called an effective attachment coefficient, which is an integral over the size distribution of aerosol particles (see Laakso et al., 2004; Hõrrak et al., 2008).

Page 25299, line 10. The reader is referred to the original work by Hõrrak (2001). In addition to this PhD thesis it would be very advisable to refer to some more widely distributed journal article, e.g. Hõrrak et al., (2000).

Page 25299, lines 14-15. Only the cluster ions are supposed to carry one elementary charge, but not all the aerosol ions. Thus, the concretization is needed. The charging probability for multiple charges on atmospheric aerosol particles is still low up to about 40 nm.

2 Methods

2.1 Instrumentation and measurements

Page 25300, section "Instrumentation and measurements". As mentioned above, more information is needed about measurement methods to understand and properly interpret the Gerdien counter (CDI-06) measurements. If we understood it correctly, the measurements have been performed at certain limiting mobilities, which correspond to the limiting diameters (or critical diameters) of 0.9, 1.5, 2 nm. The direct measurement data contain some information about the mobility distribution of air ions, but this information cannot be extracted without mathematical data processing. If any mathematical procedures have been used to derive ion mobility or size distribution form direct measurements of three different CDI-06s, these should be outlined in this section. As a comment we can say that the derivation of the ion mobility distribution from the integral counter measurements is a complicated task. The second order derivatives of the volt-ampere characteristic curve of the ion counter measurements should be found (see overview paper by Hirsikko et al. (2011)). Regarding instruments installation, nothing is said about the electrical grounding (earthing) of spectrometers, which is also im-

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portant in field measurements. The radon activity-concentration counters RAD-7 and the Gamma Scout counter, as well as all the meteorological devices should also be described in this section. Radon counters are first introduced only in Page 25307.

Page 25300, line 19. The discrete sizes of 0.9, 1.5, 2 nm are given here, but the meaning of these discrete sizes remains unclear for the reader. It should be mentioned that ion concentrations were measured at certain limiting (or critical) diameters of 0.9, 1.5, 2 nm. In addition to limiting diameters also the values of the corresponding limiting mobilities should be given. The limiting mobility is a characteristic of the ion counter and it enables a better comparison with other data than the diameter, which depends on the used mobility-size conversion algorithm and particle charge. We recommend presenting all the technical parameters of ion counters (voltage applied to the mobility analyzer, air flow rate, etc) operated under different regimes in a separate table.

Page 25300, lines 24-25, regarding the fragment of the sentence “The critical mobility (μ_c), i.e. the smallest ion mobility to be measured (maximum size), is defined as. . .”. This sentence is not correct. It is known that the Gerdien-type integral ion counter has a relatively flat transfer function, where the limiting (or critical) mobility determines its breaking point (Tammet, 1970; Flagan, 1998). Accordingly, the ions with mobilities above this limit are measured in the "saturation regime", while ions with mobilities below this limit are measured in the "conductivity regime" in proportion to their electrical mobility. Therefore, the “smallest ion mobility” is, in principle, equal to zero (or infinite particle size).

Page 25301, lines 8-10. It is not obvious what the authors mean with the end of this sentence. Perhaps they mean “approximately equal air ion concentrations”?

3. Measurements and discussion

3.1. Study of 5 different WFs

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Page 25302, regarding the section “3.1. Study of 5 different WFs”. Overview of the various measurement campaigns carried out during a three-year period at five different waterfalls is missing. It is recommended to add one table, which contains data about the time and duration of the campaigns, used instrumentation (CDI-06, SMPS, etc), number of measurements obtained etc. This table can, in principle, be companied also with Table 1.

Page 25303, lines 11-14. In order to draw this conclusion, the data in Table 1 should be complemented by additional data. It is recommended to include data about the characteristics of waterfalls (height, number of cascades, amount of water flow, surrounding topography), as well as data about ion background measurements in the vicinity of waterfalls. This information can be found only for some waterfalls in the text, but for the comparison, it is better to concentrate it into one table.

Page 25303, lines 23-26. The authors have pointed out that “one of the processes that contribute to WF ion generation is charge separation via aerodynamic break-up of micrometer-sized water droplets into nano-sized aerosols (Zilch et al., 2008).” A similar statement or assumption can be found in Page 25304, lines 5-8. We doubt that purely mechanical aerodynamic break-up or fragmentation by bubble bursting can generate nanometer size particles from large water drops. The microphysical mechanism behind this phenomenon is still unknown (see Tammet et al., 2009). Do the authors know any mechanical methods to generate nanometer size particles?

3.2 Ion inventory at the Krimml WF

Page 25304, lines 15-16. Regarding the sentence “The height of the last WF cascade provides maximal velocity to water droplets as the flow reaches 53 ms^{-1}“. The reader may think and ask also about the speed of falling water droplets of different size. For example, the terminal velocity of about 6-millimeter raindrop in the atmosphere was found to be approximately 10 m/s.

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Page 25304, lines 25-28 and page 25305, lines 1-5. This paragraph belongs to the section “Instrumentation and measurements”.

Page 25306, lines 1-14. This paragraph mainly belongs to the section “Instrumentation and measurements”.

3.3 Ion measurements at the Krimml WF

Page 25306, line 15. The fragment of the sentence “Gradients of 0.9, 1.5 and 2 nm size positive and negative integral ion concentrations” considering specific discrete sizes and “integral ion concentrations” cannot be understood by the reader.

Page 25306, lines 23-24. Taking into account the uncertainty of the measurements, there is no need for changing the values of alpha- and beta-coefficients to explain the qualitative difference between positive and negative WF-generated ion concentrations.

Page 25306, lines 25-26. A short lifetime of WF ions, not allowing propagation to larger distances, could also be a reason for “Equalization of the smallest positive and negative air ion concentrations”.

Page 25307, line 9. The radon dose rates are given in units of mSvh^{-1}, but for comparison with the RAD-7 data it is recommended to present the data also in the activity-concentration units (Bqm^{-3}) if possible.

Page 25307, line 12. It is not obvious which one of the “air ion concentration peak at the reference point (547 m)” is mentioned. Please indicate the corresponding peak size in Figure 9.

Page 25307, line 15 and line 28. The discrete size of ions (0.9 nm) is not understandable for the reader here. Please consider recommendations given for the section “Instrumentation and measurements”.

Page 25307, line 26. We did not understand the part of the sentence “clustering took

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place into a size window at around 120 nm”. What does “clustering” mean here?

3.4 Ion measurements at the Gartl WF

Page 25308, line 20. The sentence “Figure 13 depicts the negative ion concentrations vs. aerosol size” sounds a bit strange – “ions” versus “aerosol”. Actually, the authors mean the ion concentration distribution by their size, assuming that ions carry one elementary charge.

4 Conclusion

Page 25309, lines 5-6. Regarding the statement that “ion size distribution was found to be almost identical”. The statement is incorrect. It follows from Figures 9 and 13 that the size distributions have a similar shape, but the concentrations are significantly different.

Page 25309, line 7. A lower limit of the size range of 0.2–25 nm is probably a mistake. It was not mentioned anywhere in the text before. Also, it is difficult to understand how this lower limit was found from the integral ion counter measurements. An explanation is needed.

Page 25309, lines 10-11. Ions with discrete mobility-sizes (0.9, 1.5 and 2.0 nm) should be appropriately designated.

Page 25309, lines 10-11. The statement “The generation of 1.5 nm ions was less dominant than those at 0.9 and 2.0 nm” should be clarified. This statement is based on the size distribution data in Figures 9 and 13, but it seems to be inconsistent with the data in Figures 7 and 11. In both cases, the ion concentration data have been given.

Page 25309, lines 12-14. Considering the explanation of the variation of positive ion concentration generated by different waterfalls. The statement that these variations can be ascribed to the increase in the small ion recombination coefficient is rather

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strong. This is not sufficiently explained for being stated in the conclusions. We also have some doubts about the correctness of the statement itself.

Page 25309, lines 14-16. In the conclusion, the authors have listed numerous parameters that could affect the concentration of waterfall-generated ions, but many of them remained mainly without proof in this paper. The authors should show that these parameters can really be considered as factors, or list in the conclusion only those, whose effect is proven.

Page 25309, lines 17-18. Regarding the sentence “These parameters determine the quantity of aerosolised water in the form of the ion neutralization coefficient”. It is hard for the reader to understand the meaning of the end of the sentence “aerosolised water in the form of the ion neutralization coefficient”.

Page 25309, lines 18-21. Hopefully the authors mean "bubble break-up on aqueous surfaces” and “splashing of water on solid surfaces". Please consider a revision. We did not find much discussion in the paper about the contribution of different mechanisms (bubble break-up on aqueous surfaces, splashing of water on solid surfaces, aerodynamic break-up of droplets) to the generation of “waterfall ions” to allow concluding that the first two are more important than the third one. If possible, please consider it in the revised manuscript.

Page 25309, Conclusion. Unfortunately, we did not find any conclusions about the spatial distribution of the concentration or its gradients of waterfall-generated ions measured by the CDI-6, as well as the unipolarity coefficients of ions in the Conclusion.

Figures

Page 25318, Figure 6, Page 25319, Figure 7 and Page 25323, Figure 11. Reading the figure captions, one can understand that there are “ion concentration gradients” given in these figures, but really there are ion concentrations.

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Page 25321, Figure 9 and Page 25325, Figure 13. It remains unclear for the referee, as well as for the reader, how the data-points of the size distribution below 5 nm were determined from the integral ion counter measurements.

Page 25324, Figure 11. It is interesting to know, why the measurements were limited up to the distance of 120 m from the Gartl waterfall, while at the Krimml waterfall the measurements were made up to about 550 m (see Figures 7 and 11). Taking into account the initially higher concentration of ions at the Gartl waterfall, it is expected that their influence can be found also at longer distances. Is there any orographical reason for stopping the measurements at 120 m and believing that the situation does not change farther off? The first coinciding data point of positive and negative ion concentrations shown in Figure 11 at 120 m may be occasional.

Minor comments

Page 25301, line 6. Instead “depicted” there should probably be “selected”.

Page 25304, line 4. Instead of “aerosols” we recommend using of “WF-generated aerosols” or “WF-generated intermediate ions”.

Page 25305, line 3. Instead of “correlation factor” we recommend using of “correlation coefficient”.

Page 25307, line 9. A new paragraph should be started, beginning with “As outlined in the “Methods”,. . .”.

Page 25307, line 26. We recommend starting a new paragraph with “Here the WF-related ion. . .”, because it seems to be not connected with previous sentence.

Page 25309, line 7. Regarding “nano-sized and intermediate ions”. Please use the well known terminology, “small or cluster ions” instead of “nano-sized ions”.

Page 25309, lines 20. Instead of “aero dynamic”, there should be “aerodynamic”.

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