Interactive comment on “Characterization of ions at Alpine waterfalls” by P. Kolarž et al.

Anonymous Referee #5

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General recommendation

The authors investigated ion concnetrations close to Alpine waterfalls. Evaporating water will produce negative ions in the air and as a consequence leave positive charges behind in the water that hasn’t yet evaporated. Usually these positive ions deposit on the ground. Therefore, large negative ion concentrations are seen at the waterfalls and druing the rainfall. What makes this study to stand out is that several waterfalls have been studied and the instruments cover a large size range. Based on the manuscript the main motivation and aim of this study is not clear.

I recommend publication in ACP. Nevertheless, there are some issues to be resolved, mainly related to some corrections and providing more details, which help understanding the methods and the results.

First of all we would like to thank the referee for the useful and constructive comments on the manuscript. We have accepted most of the comments and discussed each single point. The referee comments have been pasted below in italic (black) while our answers are colored in red and sections regarding the main text in italic (red).

1. General comment

1.1. What is the main motivation and aim of this study? Where would you apply the results from this study? Add this information to the introduction and conclusions.

Physical characterization of WFs is part of a study that focuses on the influence of ions and aerosols generated by waterfalls on human health, i.e. on human lung function and physiology. Spatial and size distribution, polarity ratio, ion lifetime and size are crucial for defining charged particles that are penetrating human lungs. Article concerning this subject is submitted.

We agree that our motivation is missing so we added the following text to the introduction:

> *The motivation of this study was to investigate the spatial and mobility distributions of WF-generated ions which are influenced by location - specific WF physics and topographic structures in the region of the Austrian Alps. The physical characterization of WF-generated ions was part of a randomized control clinical study, which focused on the influence of these ions and WF generated aerosols on human physiology. Within this study we analysed five WFs to investigate the different properties and distribution of ions originating from each WF.*

Furthermore, we added the following text into the Conclusion (new measurements of daily and seasonal variation at Krimml WF are added in section 3.2):

> *In this study, spatial, time and size distributions of WF generated ions on several alpine WFs are presented.*
>
> *Although the daily as well as seasonal variations of WF-related water masses (maxima versus minima differ by one to two orders of magnitude) is reflected in the fluctuating ion concentrations, concentration maxima of generated ions are more related to the geological structure of the WF than on the amount of water pouring down. Nonetheless, the combined mix of parameters such as topographic configuration (height and shape of the WFs and surrounding rocks topography), quantity of water and local meteorological conditions are reflected in the detected ion concentrations of the fall.*
>
> *Spatial distribution of aerosolised ions of alpine WFs is attributed to air flow trajectories, which typically follow the course of the river bed. In the case of the Krimml WF, generated ions reached distances up to 500 m which implies maximal WF-related ion lifetime of approximately 120 s.*

1.2. Ion mobility versus size: Why did you choose to use Tammet’s mass diameter instead of Millikan-Stokes mobility diameter as you mentioned in the text? Nowadays, in air ion researcher community

Millikan mobility diameter is commonly used (see e.g.Mäkelä et al. JCP 1996, Hirsikko et al. ACP 2011). This can cause some difference in results when comparing results with for example Laakso et al. 2007. You mention that you converted ion mobility to radius (!) in the manuscript (p. 25299, line 16). Unify terminology. Make sure that all sizes that you report are actually the same size, diameter to be exact (Tammet diameter?), for all instruments (CDI and SMPS).

As stated in Hõrrak (2001), the Stokes-Cunningham-Millikan equation is sufficiently accurate above a particle diameter of 3 nm, and an improved procedure (Tammet, 1995) is available for smaller particles. Also, use of Tammet algorithm allowed us to compare our results with those obtained by Laakso et al., (2007), Luts et al., (2009) and Tammet et al., (2009) who were dealing with Lenard ions, and also used Tammet mass diameter.

Size of bigger ions (5.5 to 358 nm), were obtained by default SMPS algorithm using Stokes Millikan mobility diameter. Combination of these two approaches was made in order to obtain the most accurate results.

Thus, we also changed the wording of the introduction as follows and added two more sentences:

> *Ion mobility (measured by CDI–06) to mass diameter conversion was based on the formulation given by Tammet (1995).*
>
> *SMPS default conversion algorithm is using Stokes Millikan mobility diameter.*

1.3. *Background in measured signal due to high relative humidity: Typically, the electrical mobility spectrometers are very sensitive to high relative humidity which causes high noise levels to cover the real measured signal. Explain in more details why this not a problem here. p. 25302, lines 4-7: How did you solve the problem with WF's droplets (water spray) soaking the electrodes of the CDI, which caused current leakages in the electrode system and amplifying electronics? Especially, concentration calibration with very high concentration (up to 50000 ions cm^{-3}) should have been included into this study. This is also related to specific comment3.*

Current leakage caused by humidity on Gerdien detectors (CDI-06) is an obvious process that rapidly corrupts measurements, i.e. instruments either work well either saturates or shows senseless ion concentrations that are very easy recognizable. Current leakage on the collecting electrode due to high relative humidity is prevented by collecting electrode rubin ball fixing system. Additional precautions like protective casing (big aluminum box shown in Fig 2, heating tapes that heat amplifier electronics, outer and polarizing electrode and ball fixing sags (used occasionally due to high power consumption), sealing and silica gel desiccants in electronic housing, etc. were used to avoid current leakage on our electrode systems and amplifiers (Kolarž, et al., 2011).

Nevertheless, WF generated aerosols do not produce a 100% percent humidity as one can expect. Relative humidity during the day (from the moment the sun hit the ground) was in the range between 70 to 80% which gave us sufficient time to complete the measurements. Our overall success-rate was that more than 90% of measurements completed, under the precondition that measurements were not performed during early morning hours or late in the evening or during rainy conditions.

Accordingly, we added the following sentence into the main text:

> *Relative humidity at this and all other WFs during the measurements varied in the range between 60 and 80%, which is below the upper operational limit where measuring will become impossible due to current leakage on electrode and electronics insulating surfaces of the instruments.*

Concerning the last comment, we do not share the reason why we should do separate calibration procedures for low and high concentrations. Concentration of 50000 ions cm^{-3} equals about $8 \cdot 10^{11}$ A which is not considered as excessively high signal. In addition, this was the maximum concentration ever recorded and the result of gusts that were brief in duration and consequently – in any case the uncertainty is rather high. That is why we did not used those values for analysis but only to complete the picture.

1.3. Basic characterization of all 5 measurement sites and WF's is missing: Chapter 3.1, especially p. 25302, lines 24-25: It would be crucial to add a summary table with all the measurement sites listed *including the control sites. Where, when (how many days total of good data) and what (quantities) was measured on each site? What kind of WF is at the site (height of cascade etc.)? Add this information to the manuscript.*

We added following paragraph into section 3.2. along with Table 1:

> *However, the topography of the Austrian Alps often implies glaciers that feed their liquid phase into creeks. Runoffs often make their way downhill in turbulent conditions through the valleys. The collecting torrent form little cascades that also generate WF ions leading to variable ion distribution spectra in their embedded areas. Measurements at every WF have been carried out during midday along existing footpaths.). Table 1 lists the height, average and maximal water flow of each WF, while positive and negative air ion averages and maximal number concentrations of are given in Table 2. An important feature of the artificial WF at the river Möll regards the manner water makes its way downhill - there it pours straight past a manmade barrier into the bottom pond, so that no water splashing on rocks does take place. At all sites ion concentrations were measured at places of maximal ionization and moderate wetting of the instruments. The highest average concentration increase of negative ions up to 2 nm in comparison to control point concentration – was found to be 80–fold. Evidently, alpine WF ion concentrations fluctuate depending on the WF sampled. Ion averages at the examined sites typically depend on the amount of water flowing, which is subject to seasonal variations and oscillate with the melting of the glaciers. In contrast, the origin and structure of the ions generated at reference sites arise mostly from natural radioactivity, which is subject to diurnal changes and related to the fluctuation in concentrations of radon, thorium and their progenies.*

Table 1. WF height (approximate), average and maximal water flow.

WF name	*Height (m)*	*Average water flow (m^3/s)*	*Maximal water flow (m^3/s)*
Krimml	*140 x 100 x **140***	*5.1*	*17.5*
Stuiben	*150 x **150***	*1.6*	*2*
Bad Gastein	*310 (measured cascade **50**)*	*1.83*	*6.8*
Gartl	*100 (50 x **50**)*	*0.6*	*0.9*
Artificial (river Möll)	***5***	*1.33*	*4.66*

In the abstract you state that "During a 3-year field campaign..." but there is no time series presented in the paper. This is very confusing.

In Section 3 we added overview of the measurement times:

> *The study of WF-related aerosol inventory lasted for three years, from 2008 till 2010. At the very beginning measurements on several different WFs have been conducted - as shown in section 3.1. After locating the WF in Krimml as the most promising and convenient, spatial measurements at this WF have been done during June and September 2009 - shown in section 3.2. In section 3.3 measurements of ion size distribution are presented in detail.*

1.4. Seasonality in ion concentrations: How would you assume that the seasonality affects you results? You were not able to measure at all sites at the same time. What kind of seasonal behaviour the WF's have? How would this affect the ion concentrations?

This is why you actually would have needed to measure the long time series from one selected site.

We added whole new set of measurements we carried out at Krimml WF during the project named „Immediate effects of waterfall ions on physiological parameters in the upper airways" during July and September of the year 2009. WF generated ion measurements were carried out simultaneously with exposure of every proband. Measuring details are added to Section 3.2. as listed below:

> *During the 2009 field campaign, a series of 7-day measurements of ions ≤2 nm have been performed and included two separate field campaigns: 10-26th of July as well as 09-23rd of September. Measurements were carried two times a day, and regards a sampling session during the morning hours (around noon) and one in the afternoon (around 16 h) for one hour each. This was done*

> *simultaneously along with the medical survey to monitor immediate responses of exposed individuals. According to the data provided by the authorities of the local hydro power plant, which is just 700 m downstream from the fall, the average water flow in June was 14.4 m^3s^{-1} whereas in September it was 5.5 m^3s^{-1}. The former is 2.6 times larger than the latter and directly correlates with the period of intensive glacier melting during summer. Daily variations of negative WF ion concentrations were within the 8% range. However, the average ion concentration in July (10944 ions·cm^{-3}) was 1.5 times higher than in September (7502 ions·cm^{-3}) of that year. While daily differences in ion concentrations are the consequence of local meteorological parameters, the significant difference between July and September are related to the reduced water masses during September. In any case, the drastic reduction in the amount of water flow affects negative ion generation and sinking ratio to a much greater extent than the suppressed generation in ion concentrations.*

Also, sentence about seasonality is added in the Conclusion:

> *Although the daily as well as seasonal variations of WF-related water masses (maxima versus minima differ by one to two orders of magnitude) is reflected in the fluctuating ion concentrations, concentration maxima of generated ions are more related to the geological structure of the WF than on the amount of water pouring down.*

2. Specific comments

2.1 p. 25298, lines 23-24: You have to let reader know where you found the numbers you report in the introduction. Add citations. E.g. Hirsikko et al. paper in ACP 2011 and reference therein might be very useful to you.

We added 23 references. List is given below in the section New Added References.

2.2 p. 25299, lines 14-15: What does the Aplin, 2008 citation really mean? In atmosphere the charged particles are not single charged. Especially the ones you measure with the Grimm SMPS (>20 nm). Is this citation related to mobility-to-diameter or voltage-to-mobility conversion?

In our experimental setup we assumed that each aerosol size class is dotted with one single negative charge - even though we are all well aware that larger diameters can easily occupy multiple charges. The use of a single SMPS device does not allow us to determine the exact amount of charge residing on particles of a given size class. The only way to determine the true value would be to use two SMPS' in series; within such a tandem design, the first DMA would be set to a given aerosol size class while the second DMA would screen the real quantity and along with the CPC also the corresponding diameter. By pooling the data of all 44 individually screened size classes into one graph containing all size classes one obtains the charge-diameter relationship for the entire spectrum between 5.5 - 350 nm. Such a design is rather inappropriate for field measurements; it should be done in the lab - as long as a 2nd DMA with a controller unit is available.

What does this mean for our field study? since we operated our setup without the ^{241}Am neutralizer, it implies that the particles with multiple charges detected by the SMPS shift from the original size bin to one that corresponds to a net smaller size bin (due to the multiple charge loading, such a particle behaves like a smaller one); thus, the spectrum obtained with the SMPS in the larger size range (>50nm) is smaller in amplitude than it really is. By looking at the graphs below file you will realize that most scans using the ^{241}Am radiation source (causing the aerosol to be discharged according to the Boltzmann distribution pattern) the 100 nm fraction is well represented, whereas the 10 nm range is completely stripped off the spectra. This indicates that in comparison, the size distribution is present in principle, yet the actual number concentration of the raw aerosol (not using the ^{241}Am radiation source) can not accurately be determined with the setup we used.

We added one sentence concerning this subject:

Effect of multiple charges is assumed to be significant in the case of heavy large particles >50 nm (Hõrrak, 2001) which is considered as artefact (i.e. uncertainty).

> *By operating the SMPS with and without the ^{241}Am neutralizer, it was possible to uncover the differences in the spectra. Besides the sharp increase in sub-20 nm size range when the SMPS was*

> *operated without the neutralizer, the remainder of the spectra (20-350 nm) still reveal similar trends – regardless whether the neutralization source was attached or not.*

What kind of inversion algorithm did you use for the Grimm SMPS data when measuring naturally charged ions? I assume that you took only the diffusional losses into account in the inversion – not e.g. charging efficiency. Add this information to the manuscript.

Here we did not alter the software at all. Using the built-in algorithm, it enabled us to compensate for instrumental losses (DMA & CPC efficiencies). There is no other way to interfere with the software (v1.34). However, this can be a different story when using the new labview-based version, as this enables the user to manipulate many more parameters. However, the latter was not yet available when we performed the field studies.

2.3 p. 25031, lines 8-11: How was the CDI's with different cut-off sizes intercompared or calibrated? Please if the exist some study refer to it. Now the sentence that states: "It is expected that simultaneous measurements of all three instruments on places where no WF related ions are present should show approximate air ion concentrations." This doesn't sound convincing.

According to comments of this and referee number four, we added following text in section 2.1. in order to explain our ion detector setup, calibration and measurements:

> *Integral aspiration counter also known as Gerdien condenser is the most commonly used tool for atmospheric conductivity measurements (Gerdien, 1905). Its operation is based on the polarizing voltage that is applied between two aspirated coaxial electrodes. Ions of the same polarity as polarising voltage are deflected toward the collecting electrode depositing charge ($q=1.62\ 10^{-19}$ C) and generating current (I):*
>
> $$I=n\cdot q\cdot Q \qquad (2)$$
>
> where Q is air-flow through the electrodes.
>
> *The limiting mobility of the aspiration condenser (μ_c), determines the ion mobility above which all ions should be collected on the collecting electrode, and is defined as a combination of polarizing voltage (U) and air flow:* (equation 2 in Discussion manuscript*).*
>
> *Concentrations of small and intermediate ions with mobilities smaller than 0.3 $cm^2V^{-1}s^{-1}$ (mass diameter 2 nm) were measured using three identical Gerdien condenser detectors (CDI-06), made at the Institute of Physics, Belgrade. Using the dynamic property of ionic motion in an electric field, the polarizing voltage and air flow on three identical CDI-06 detectors was set to measure small cluster, big cluster and intermediate ions with the following limiting mobilities: 0.3, 0.55 and 1.18 $cm^2V^{-1}s^{-1}$, which corresponds to the boundary mass diameters of 0.9, 1.5 and 2 nm (Eq.3). Selected limiting mobilities correspond to characteristic ion-sizes originating from WFs, which is around 2 nm (Laakso et al., 2007) and by background ionisation sources as nuclear decay as well as cosmic rays - with the latter up to 0.9 nm (small cluster ions).*
>
> *The relatively large electrode cross–section (R_1=0.028 m, R_2=0.005 m) along with the collecting electrode length (L=0.4 m) provides a relatively high air flow (0.0048 m^3s^{-1}) and sufficiently intense signal to use a relatively small measuring resistor (R_M=1 GΩ) thus resulting in a reduced measuring uncertainty. The collecting electrode is at virtual ground potential; with the measuring system, including the polarization circuit, is galvanically separated from the power supply and the PC communications port (USB). Each of the three detectors has its own power supply, temperature, pressure and relative humidity sensor, and also features a sophisticated such as dynamic zeroing (eliminates zero offset drifts during measurements), live data acquisition, with programmable data averaging, sampling and polarity change options (Kolarž et al., 2011).*
>
> *According to Eq.1, the detector amplifier (i.e. A/D converter) output signal to air ion concentration conversion factor was calibrated prior each measurement using a Keithley Picoampere 261 source. The limiting mobility was calculated using Eq.3. Correction due to ion repulsion of small ions at the inlet of the detector (Anderson et al., 1991; Misaki, 1961) on detector measuring ions with lower mobilities (0.3 $cm^2V^{-1}s^{-1}$), i.e. with highest polarizing voltage, was experimentally obtained and*

> *incorporated into measuring signal to air ion concentration conversion factor. Ion mass mobility (measured by CDI–06) to diameter.*
>
> *Correction of ion concentration for each mobility class was obtained using second derivation of current to voltage characteristics (Horrak et al., 2008; Flagan,1998) measured at the place where sufficient concentration of WF generated ions were present, but their fluctuations due to gusts was moderate. Estimated correction was incorporated into measuring signal to air ion concentration conversion factor. Further derivation of ions with limited mobilities of 0.55 and 1.18 $cm^2V^{-1}s^{-1}$ from integral values was obtained using the subtraction method (Dhanorkar et al., 1992); gaining ions in size windows of 0.36-0.9 nm, 0.9-1.5 nm and 1.5-2 nm.*
>
> *Although the determination of ion mobilities using integral Gerdien counters is not as accurate and narrowband as for e.g. a DMA (differential mobility analyzer), portability and robustness compensate that disadvantage and allow measurement on nearly impassable terrains.*
>
> *Away from the WF, in conditions where mostly small cluster ions prevail (except in cases of nucleation bursts of intermediate ions), two detectors that are measuring ions with lower mobilities should measure equal or slightly higher ion concentrations as one that is measuring small ions (Laakso et al., 2007). As stated above, similar applies to the ratio of positive and negative background ion concentrations on the reference sites where n^+/n^- should be about 1.12.*
>
> *Relative humidity at this and all other WFs during the measurements varied in the range between 60 and 80%, which is below the upper operational limit where measuring will become impossible due to current leakage on electrode and electronics insulating surfaces of the instruments.*

Please, move the CDI1-2 intercomaprison results from p. 25304, lines 25-> into chapter 2.1.
We moved it.

2.4 p. 25309, line 8: Did you actually measure 0.2 nm ions? I suppose this is in Tammet diameters? Those are really small ions. How did you measure those? You haven't mentioned ions diameter of 0.2 nm earlier in the paper.
Ion diameter of 0.2 nm is a mistake. The actual limit is 0.36 nm, which is the smallest traceable ion diameter according to Hõrrak et al. (2000).

3 Technical corrections
Chapter 2.1: Please add manufacturer and intrument model information always when introducing instruments (e.g. for Grimm SMPS, CPC, DMA (length), and anemometer).
Chapter 2.1: What is the time resolution of the instruments?
The instrumentation we used: the SMPS consisted of a CPC (model 5403) attached to a middle sized DMA column (5540 26 Reischl Vienna type).

Combining the triplet readings of the CDI with that of the SMPS, one obtains a composite graph as shown in figure 9 as well as 13. Each data series representing the aerosol inventory for a given distance from the WF has been obtained by gathering at least three SMPS-scans - each scan takes about three minutes to cover the full size spectrum from 5.5 to 350 nm, split into 44 size class bins. Since each bin consists of nine separate measurements (minus the initial and the last recording - to account for dynamic measurement principle during switching from one DMA-voltage, respectively size bin to the next) an overall set of 924 individual measurements spread over 44 size bins - constitute for the 5.5-530 nm scanning window - have been gathered at a given location. At the same time, the GCS, with its second by second mode of acquisition, logs around 2700 readings simultaneously for each of three fixed size bins at the lower end of the particle spectrum, i.e. 0.9, 1 and 1.5 nm. Hence, each graph is a composite of averaged CGS and SMPS data.

Concerning time resolution of the ion detectors we have added following sentence in section 2.1.

> *The time resolution of CDI–06 detectors was set on 2 seconds for all measurements so that sufficient statistics and data analysis can be achieved.*

p. 25301, line 13: What does RPM stand for?
Revolutions per minute (RPM)

References used in the comments section:

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New added references:

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