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## Aerosol nucleation induced by a high energy particle beam

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[1] We have studied sulfuric acid aerosol nucleation in an atmospheric pressure reaction chamber using a 580 MeV electron beam to ionize the volume of the reaction chamber. We find a clear contribution from ion-induced nucleation and consider this to be the first unambiguous observation of the ion-effect on aerosol nucleation using a particle beam under conditions that resemble the Earth's atmosphere. By comparison with ionization using a gamma source we further show that the nature of the ionizing particles is not important for the ion-induced component of the nucleation. This implies that inexpensive ionization sources - as opposed to expensive accelerator beams - can be used for investigations of ion-induced nucleation. **Citation:** Enghoff, M. B., J. O. P. Pedersen, U. I. Uggerhøj, S. M. Paling, and H. Svensmark (2011), Aerosol nucleation induced by a high energy particle beam, *Geophys. Res. Lett.*, 38, L09805, doi:10.1029/2011GL047036.

### 1. Introduction

[2] Aerosol and cloud research is one of the most critical frontiers of climate science [Shindell *et al.*, 2009; Bodenschatz *et al.*, 2010] and the direct radiative forcing and indirect cloud albedo forcing from aerosols remain the dominant uncertainty in the radiative forcing of the atmosphere. Among the uncertainties are the mechanisms behind formation of new aerosols from the gas phase and several schemes have been suggested [Curtius, 2006] such as ternary nucleation with amines [Kurtén *et al.*, 2008]. Observations show that sulfuric acid, together with water and other gas phase molecules play an important role in atmospheric nucleation [Lee *et al.*, 2003; Kulmala *et al.*, 2006] and these systems are also the basis of many theoretical nucleation works [Wyslouzil *et al.*, 1991; Hanson and Lovejoy, 2006].

[3] The importance of ion-induced nucleation for the Earth's atmosphere is a subject of intense discussions [Yu, 2010; Sloan and Wolfendale, 2008] which is being studied both in field measurements [Kulmala *et al.*, 2010] and in modeling work [Pierce and Adams, 2009; Kazil, 2010]. Previous laboratory studies using gamma rays have found a positive correlation between ionizing radiation intensity and aerosol concentration in air [Raes and Janssens, 1985; Svensmark *et al.*, 2007; Enghoff *et al.*, 2008], but systematic laboratory experiments on aerosol nucleation with ionizing

particle radiation have so far only been performed with very high radiation doses far above natural atmospheric levels using  $\alpha$ - and  $\beta$ -sources [Adachi *et al.*, 1996] or high intensity electron [Hakoda *et al.*, 2003] and proton beams [Imanaka *et al.*, 2010]. Preliminary measurements with a more realistic 3.5 GeV/c pion beam have been reported to show indications of ion-induced nucleation [Duplissy *et al.*, 2010], but suffered from unstable conditions in the reaction chamber. A new experimental facility dedicated to these studies is under commissioning at the CERN Proton Synchrotron (PS) [CLOUD Collaboration, 2000] and is likely to resolve these problems.

### 2. Experimental Methods

[4] The experiments took place in a 1 m long, 50 L cylindrical, electropolished stainless steel reactor, previously used by Enghoff *et al.* [2008]. A mixture of pure humidified synthetic air is continuously flowed through the vessel at a rate of 3.1 L/min together with trace amounts of (typically 2 ppb) sulfur dioxide (SO<sub>2</sub>) and approximately 55 ppb ozone (O<sub>3</sub>) with the relative humidity held at about 50%. UV light was used to initiate the photochemistry in the chamber leading to an in situ production of sulfuric acid. The temperature was kept at 21.2 ± 0.04°C and the pressure at 1–2 mbar above room pressure.

[5] A low-intensity beam of 580 MeV electrons from the Aarhus Storage Ring ASTRID [Myers *et al.*, 1998] at the University of Aarhus was used as the ionizing source. The energy of the electron beam falls within the main part of the natural cosmic ray spectrum and the induced ion concentrations cover the full range of atmospheric values.

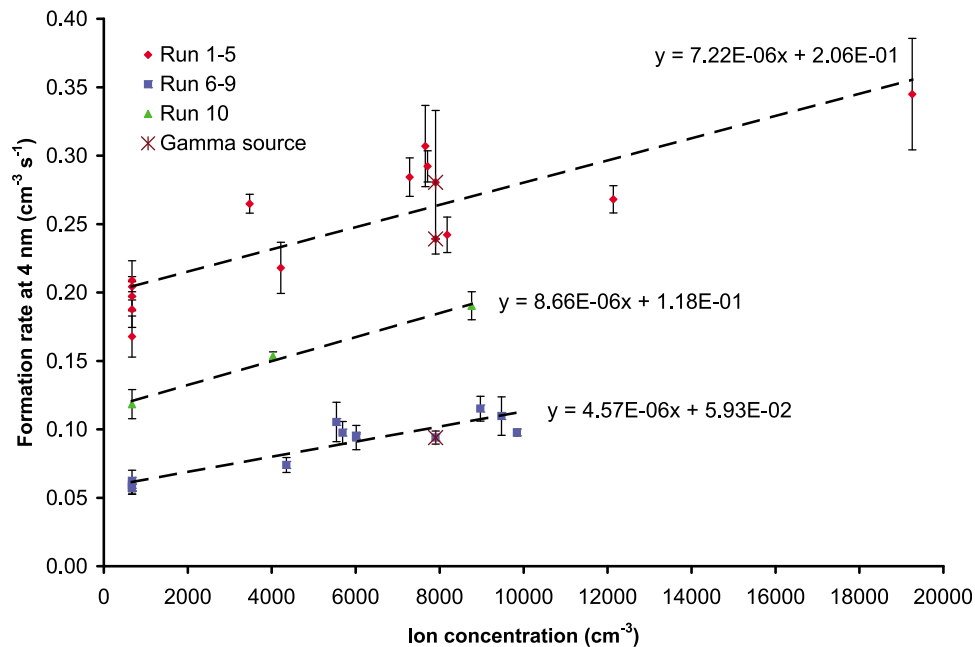
[6] Production of aerosols was measured using a condensation particle counter (CPC) with a nominal 50% cutoff diameter of 4 nm. The chamber volume could be ionized using the 580 MeV electrons going coaxially through the chamber covering an elliptical area with semi axes of 4.4 and 6.1 cm (FWHM) at the front end of the chamber. The ionization level from the beam was adjustable by changing the amount of beam extracted and, as shown by the estimated energy loss (see auxiliary material), the electrons pass through the volume of air almost undisturbed.<sup>1</sup> Alternatively a 33.5 MBq Na-22 gamma source could be used. For some of the runs a 5 cm thick scintillator was placed in the beam, 7.5 m upstream of the chamber, dispersing the beam by an additional 6 cm at the front of the chamber.

[7] The procedure for each experiment involved turning the UV lamps on for 10 minutes giving a burst of sulfuric acid and then recording the resulting particle number. The time between UV bursts was 60 minutes, in order to allow

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**Figure 1.** Formation rate (at 4 nm) versus ion concentration as measured by the CPC (see text for details). Error bars show statistical uncertainties.

the aerosol concentration (as measured by the CPC) to reach zero by the end of each burst, with the aerosols being lost to walls and by dilution.

### 3. Results and Discussion

[8] The measurement program consisted of varying the ionization levels by adjusting the electron beam, investigating the effect of dispersing the electron beam, and comparing the aerosol formation (observed at 4 nm) induced by, respectively, the gamma source and the electron beam. A range of ionization levels from background levels to  $\sim 700 \text{ cm}^{-3} \text{ s}^{-1}$  were tested and the results can be seen in Figure 1, where the formation rate measured by the CPC is shown as a function of ion concentration. The runs have been divided into 3 groups and it is seen that an increase in ion concentration causes an increase in the aerosol formation rate for all three groups of runs. Also shown for each group is the best linear fit to the data. Runs 1–5 were made using air from the same cylinders. Between run 5 and 6 the air supply was changed, causing a large shift in the number of aerosols produced in a burst and between runs 9 and 10 a shift occurred again. This shift is seen on Figure 1 as a vertical displacement of the 3 groups of measurements, and we speculate that the shift is due to varying amounts of trace impurities in the purified air cylinders. The impurities are able to pass the filters and influence the neutral component of the nucleation process. There is also a smaller variation of the slopes of the linear fits in Figure 1 between the 3 groups of measurements, but they do not seem to vary in a systematic way with the vertical shift. While we cannot exclude that the ion-induced component of the nucleations are also affected by the impurities, the effect thus seem to be smaller on this component, and does not change the main observation that there is a clear effect of the ion concentration on the formation rate.

[9] During run 6 the effect of dispersing the beam was tested. The formation rate at 4 nm changed from  $0.095 \pm 0.003 \text{ cm}^{-3} \text{ s}^{-1}$  to  $0.094 \pm 0.009 \text{ cm}^{-3} \text{ s}^{-1}$ , showing that there was no significant difference between the two settings. This also shows that the gases in the chamber were well mixed.

[10] The sulfuric acid concentrations are found to be  $\sim 7 \cdot 10^8 \text{ cm}^{-3}$  (+100/–50)% for runs 1–5 and  $\sim 6 \cdot 10^8 \text{ cm}^{-3}$  for runs 6–10, with run 10 having a slightly higher concentration than runs 6–9 (see auxiliary material). This could explain the changes in aerosol production rates observed during the experiments.

[11] In all runs the effect of the ions is to produce additional aerosol formation over a wide range of ion concentrations from background levels to  $19,000 \text{ cm}^{-3}$ . Model results [Kazil and Lovejoy, 2004] indicate that the nucleation rate should peak at high ionization levels, but apparently this range is not reached at the present experimental conditions, which may be a result of our rather high  $[\text{H}_2\text{SO}_4]$  values. The error bars shown are statistical uncertainties and the absolute uncertainties are much larger, but since our purpose is to investigate the relative variation of the nucleation rate with ion density, we find the statistical uncertainties to be most relevant. A linear relationship (which has been observed before in a smaller range of ion densities by Svensmark *et al.* [2007]) would show that the nucleation rate depends on ion concentration, but we cannot for example exclude a square relationship, which would indicate that nucleation depends on the ion production rate and any conclusions would also be complicated by linear wall losses. An extrapolation of the curves to zero ion density would give the nucleation rate due to processes not involving ions, but further experimental work at low ionization values is needed to resolve this.

[12] We caution that the combination of a finite residence time in the reaction chamber and a detector cutoff larger than the diameter of the critical cluster has the effect that

only a fraction of the nucleated particles will grow to detectable sizes as recently shown by *Sipilä et al.* [2010]. In our experiment the possibility of a faster growth rate of the ions may artificially enhance the measured particle concentrations when using the ionizing sources compared to no sources. From an estimated residence half-life of 200 s, and with  $[\text{H}_2\text{SO}_4]$  concentrations in the range  $6\text{--}7 \cdot 10^8 \text{ cm}^{-3}$  we conclude from *Sipilä et al.* [2010, Figure 3] that we reach mean particle diameters above 3 nm, which is close to the detector cutoff around 4 nm, but may still leave the majority of the aerosols undetected. Since our measurements were made under stable conditions where only the ion density was varied, our observation that ions enhance the formation rate at 4 nm remains solid in spite of the large absolute uncertainties; however, the measured relative enhancement should be considered an upper limit. *Sipilä et al.* [2010] also found that the rate of homogenous nucleation measured with an instrument similar to ours should vary with the 6th power of  $[\text{H}_2\text{SO}_4]$ . If the difference between our runs 1–5 and 6–9 is due to a change in  $[\text{H}_2\text{SO}_4]$  by about a factor of 1.2, this should result in a homogenous nucleation rate difference by a factor of 3 which is close to the values found by extrapolating the rates shown in Figure 1 to zero ion concentration.

[13] Using an analytical model (see auxiliary material) we have calculated the unobserved nucleation rate  $J_1$  from the observed formation rates  $J_4$ . This has large inherent uncertainties and gives absolute 1 nm nucleation rates in the order of  $1 \text{ s}^{-1} \text{ cm}^{-3}$ , which is about two orders of magnitude lower than previous laboratory measurements under similar conditions and with only background ionization [see *Sipilä et al.*, 2010, and references therein]. Some explanations for this discrepancy could be that our sulfuric acid concentrations and our counting efficiency are lower than estimated. A possible remedy would be to normalize our nucleation rates to the literature values, but we have chosen to give our estimated absolute values with the added note that only the relative variations should be considered accurate.

#### 4. Conclusions

[14] An important result from this work is (as seen from Figure 1) that nucleation induced with the ionization from the gamma source experiments are indistinguishable from those using the electron beam. Compared to the 580 MeV electrons the gamma rays have rather low energies and the electrons emitted through Compton scattering will ionize very locally, whereas the 580 MeV electrons have a mean energy loss rate close to the minimum (minimum ionizing) and will ionize along their path. The gammas may however ionize more than one atom due to secondary gammas emitted in the Compton process. Nevertheless our data show that the fraction of nucleation events due to the electron beam or the gamma rays is an effect of the produced ions and that the nature of the ionizing particles is not important. This demonstrates that while a particle beam can be used for studies of the ion-effect on aerosol nucleation future laboratory studies of atmospheric aerosol nucleation can be greatly facilitated, since they can more easily be done with gamma radiation sources instead of using complicated and expensive accelerator beams.

[15] Further we conclude that under the present experimental conditions ( $P = 1$  atmosphere,  $T = 21.2^\circ\text{C}$ ,  $[\text{SO}_2] =$

2 ppb) there is a clear evidence of ion-induced nucleation as a source of aerosol production, which corroborates earlier measurements using only gamma radiation as the ionizing source [*Svensmark et al.*, 2007; *Enghoff et al.*, 2008]. However we also stress that this is a qualitative result and that further work at lower values of  $P$ ,  $T$ ,  $[\text{H}_2\text{SO}_4]$ , and  $[\text{SO}_2]$  is required to perform an extrapolation to real atmospheric conditions, in particular since our  $[\text{H}_2\text{SO}_4]$  concentrations of  $6\text{--}7 \cdot 10^8 \text{ cm}^{-3}$  are at least an order of magnitude above typical clean-air concentrations of about  $10^7 \text{ cm}^{-3}$ . Using the lower estimated  $[\text{H}_2\text{SO}_4]$  concentration of  $6 \cdot 10^8 \text{ cm}^{-3}$  in runs 6–9 we find that the effect of changing the ion density from  $\sim 700$  to a typical atmospheric value of  $\sim 1,500 \text{ cm}^{-3}$  results in a relative increase in formation rate at 4 nm of approximately 7%, but modelling results [*Kazil and Lovejoy*, 2004] show that lowering the values of the present experimental parameters will influence both the critical cluster size and the particle production and may both correlate and anti-correlate with the ionization level.

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