

# SKY-ZERO: Study of aerosol nucleation in an ultra-low ionisation environment

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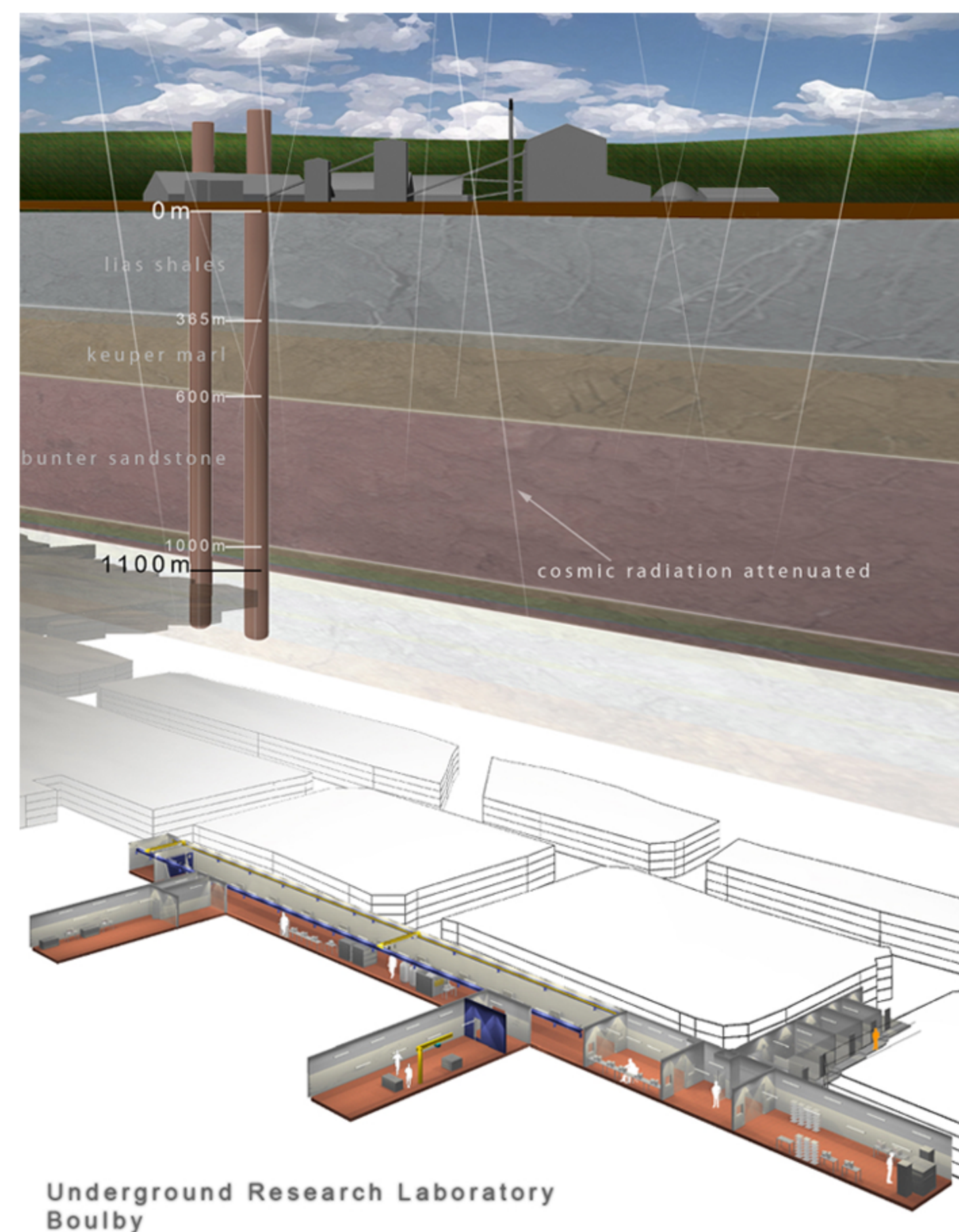
## INTRODUCTION

Do cosmic rays play a role in cloud formation? Studies of the role of ionising radiation in atmospheric aerosol formation are important for improving our understanding of one of the potential variables responsible for changes in global temperature and climate.

In this work we have studied aerosol formation at ultra-low ionisation levels, using the existing deep underground science facility at Boulby mine, UK. At 1100 m depth, with a corresponding factor  $10^6$  reduction in cosmic ray muon flux, the Boulby facility is an ideal place to study the role of ions in aerosol nucleation.

By exposing a controlled volume of air to varying levels of ionising radiation, and with the minimum ionisation level vastly reduced compared to normal surface laboratory conditions, we have provided both a validation of earlier studies of ion-induced nucleation and extended the measurements to ionisation levels approximately 3 order of magnitudes lower than any earlier study.

Getting this close to zero ionisation allows us to distinguish between the neutral and charged contribution better than previously. The effect is explored over a range of sulphuric acid concentrations.



Underground Research Laboratory Boulby

## SETUP

We used a 50 L electropolished stainless steel chamber. Collimated UV light entered through a Teflon foil to start the oxidation of  $\text{SO}_2$  into sulphuric acid that condense with water to form aerosols. Aerosols and trace concentrations of ozone and  $\text{SO}_2$  were measured along with temperature, pressure, and humidity. A continuous flow of 3.1 L/min of clean, humidified, synthetic air went through the chamber, which was kept at 1 mbar overpressure at all times. A 17 MBq Cs-137 gamma source was used to increase the ionisation in the chamber.

Each measurement was initiated with the production of a 'burst' of  $\text{H}_2\text{SO}_4$  following exposure of the chamber to UV light, resulting in the production of aerosols. The peak aerosol concentration lasted for a few minutes before decaying exponentially due to wall and dilution losses.

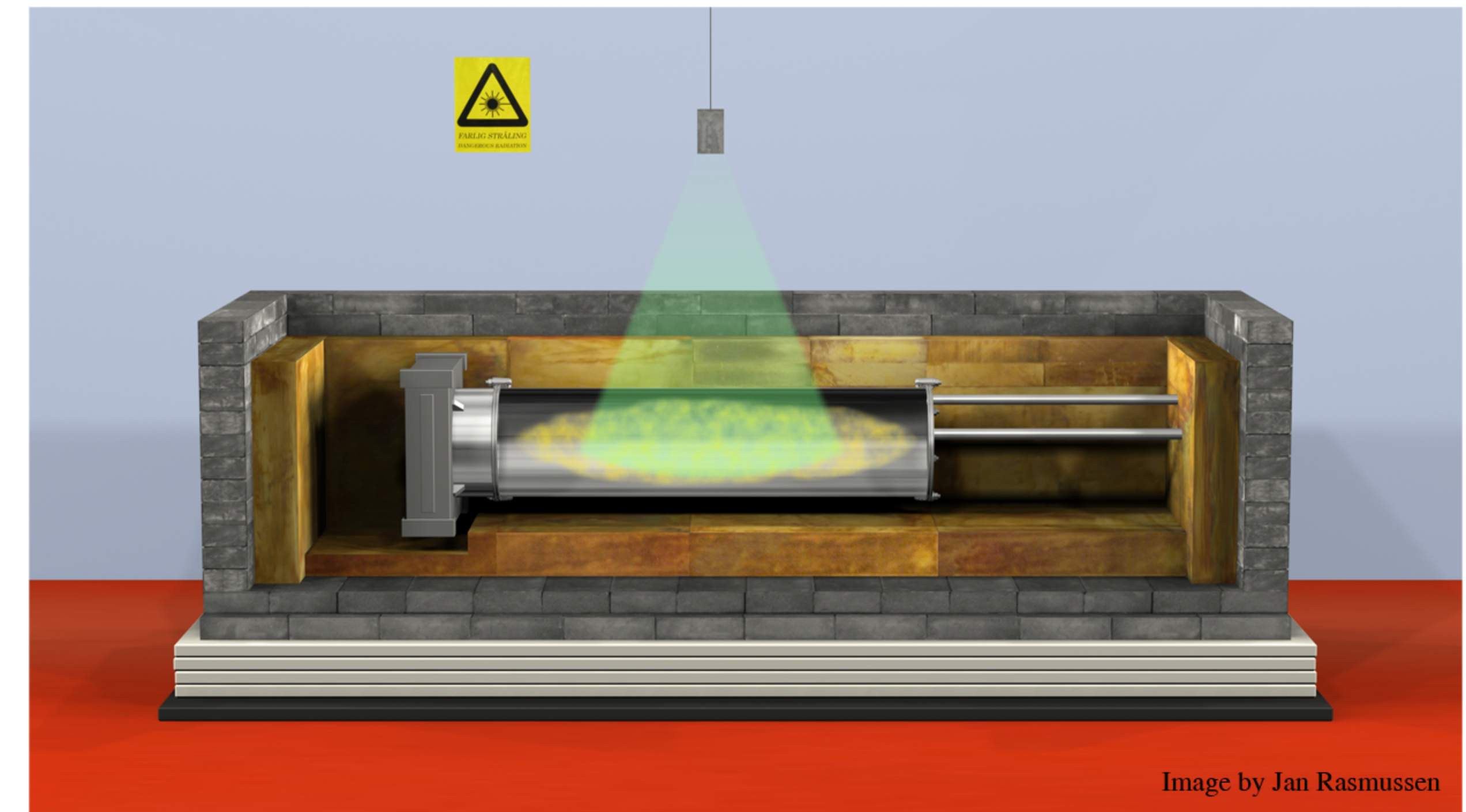


Image by Jan Rasmussen

## ION CONTROL

- 1.1 km of rock shielding reduced cosmic ray muon intensity by a factor  $10^6$ .
- 10 cm of lead and 10 cm of copper shielded the chamber from gamma radiation from 5 sides. For very low ionisation measurements the 6th side could be closed giving a total reduction in ion production rate through gammas of greater than a factor  $10^4$ . We calculate an ion production rate of  $< 0.04 \text{ cm}^{-3} \text{ s}^{-1}$  due to gammas, with the 6th side open and  $< 2 \cdot 10^{-5} \text{ cm}^{-3} \text{ s}^{-1}$  with the 6th side closed.
- Stored Synthetic air, filtered with activated charcoal ensured Radon levels and associated ionisation levels were a factor  $10^3$  or more below typical surface levels. Inner surfaces of the chamber were also electropolished to reduce ionisation from plated-out long-lived Rn daughters and we measured the resulting concentration. Ion production due to radon was thus  $< 0.007 \text{ cm}^{-3} \text{ s}^{-1}$ .

## ANALYSIS

For each series of measurements we performed a number of bursts with minimal ionisation, followed by bursts at various ionisation levels, and ending with bursts at minimal ionisation again. These bursts are used to correct for any drift, as shown in Fig. 1 where the black peaks result from minimum ionisation bursts ( $0.04 \text{ cm}^{-3} \text{ s}^{-1}$ ), blue from surface ionisation levels ( $2.6 \text{ cm}^{-3} \text{ s}^{-1}$ ), and red from high ionization ( $32 \text{ cm}^{-3} \text{ s}^{-1}$ ).

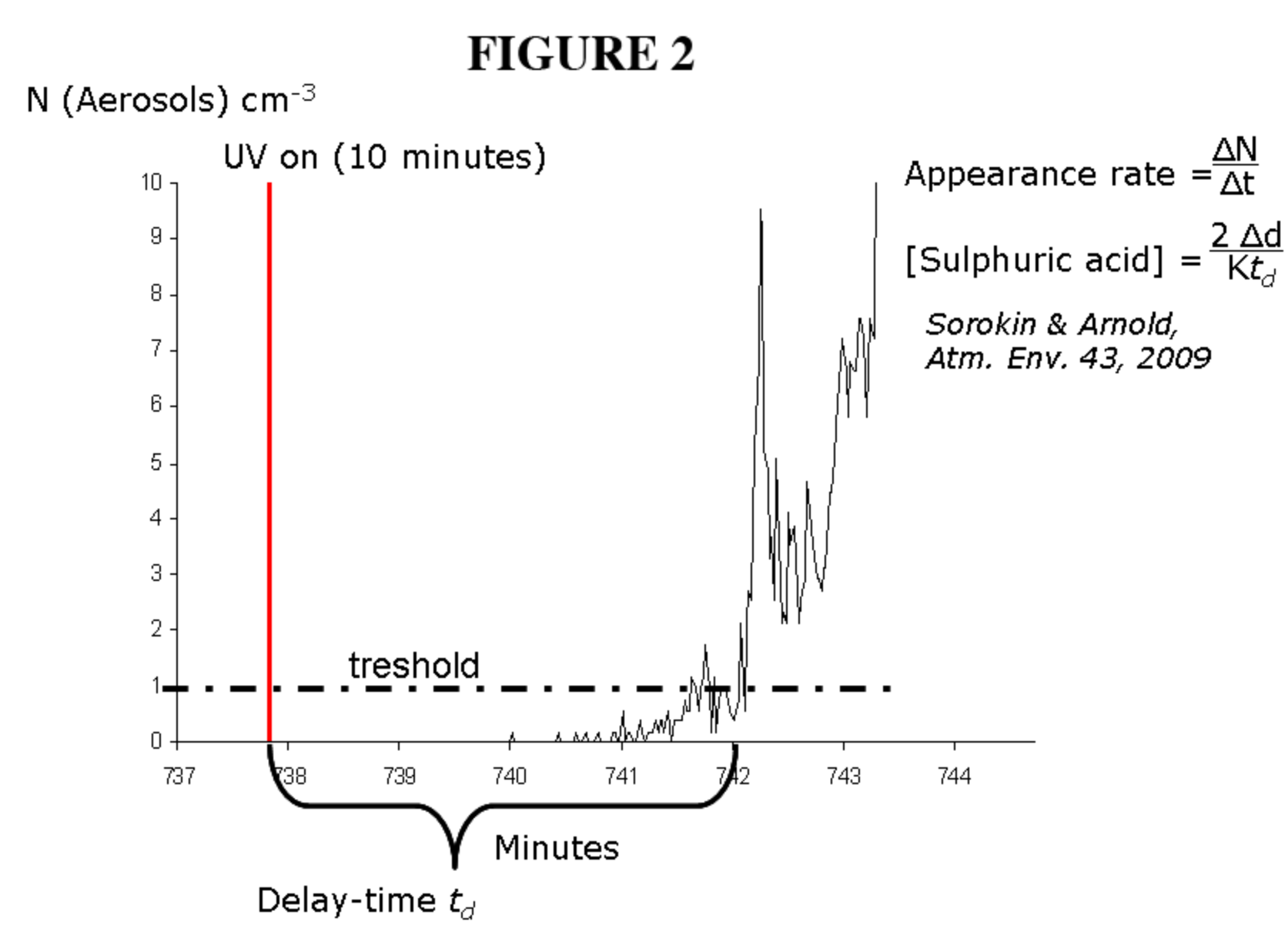
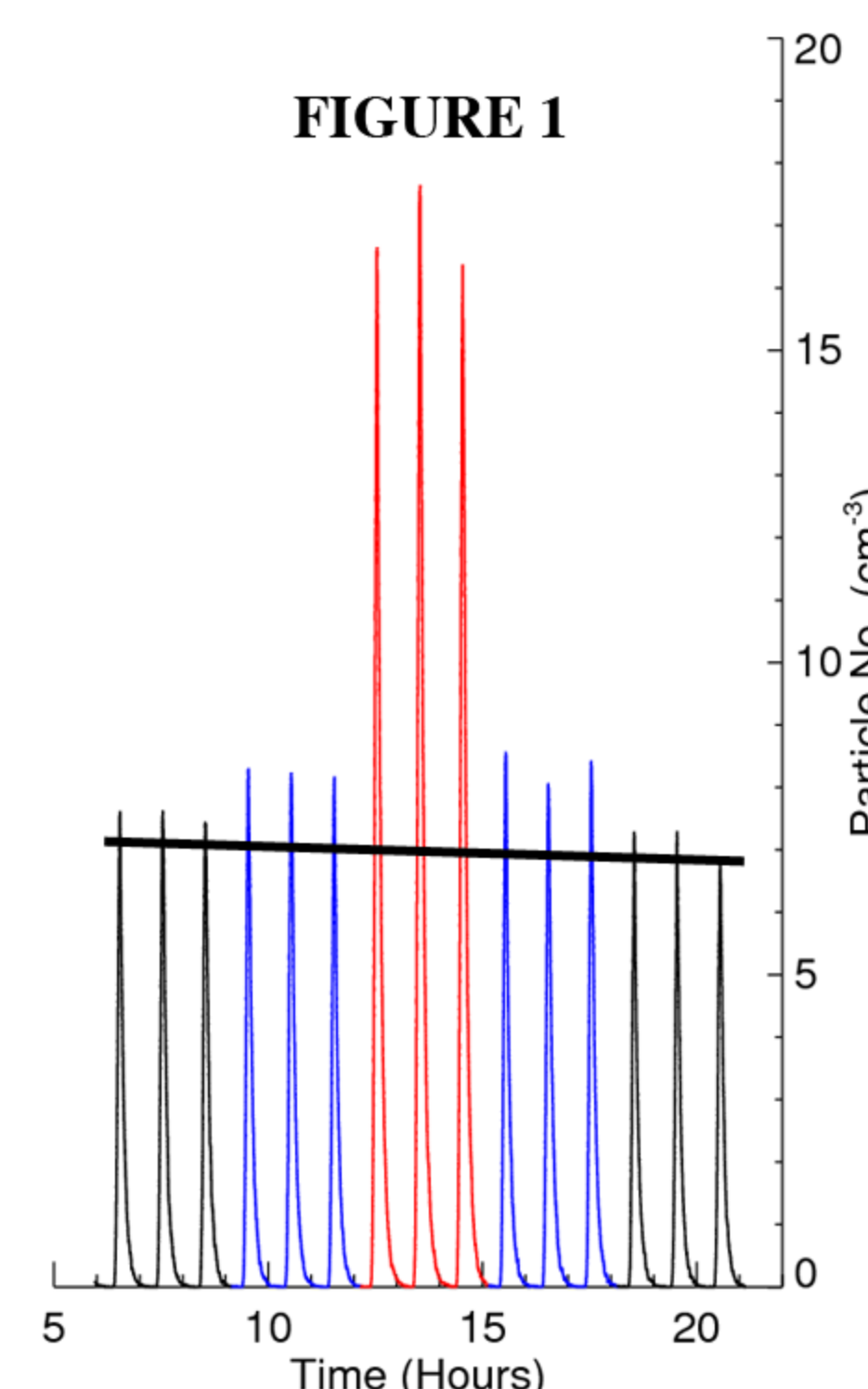


Figure 2 shows the start of a peak. We calculate the appearance rate of aerosols above the detection limit ( $\sim 4 \text{ nm}$ ) as the smoothed concentration at the top of the peak, divided by the timespan between turning the UV lights on and reaching the top of the peak. The sulphuric acid concentration is derived from the growth rate of the aerosols, determined by measuring the time it takes for the concentration to reach and stay above a threshold value of  $1 \text{ cm}^{-3}$ .



Once the appearance rate at  $4 \text{ nm}$  ( $J_4$ ) is known we need to find the true nucleation rate at about  $1 \text{ nm}$  ( $J_1$ ). We approach this using two separate methods.

**Loss rate method:** The loss rate of the aerosols is found from the decay of each individual peak. We use the estimated sulphuric acid concentration to find the size of the aerosols at a given time. This is combined with a size dependent loss (scaling as  $1/d$ ) to find  $J_1$  from  $J_4$ .

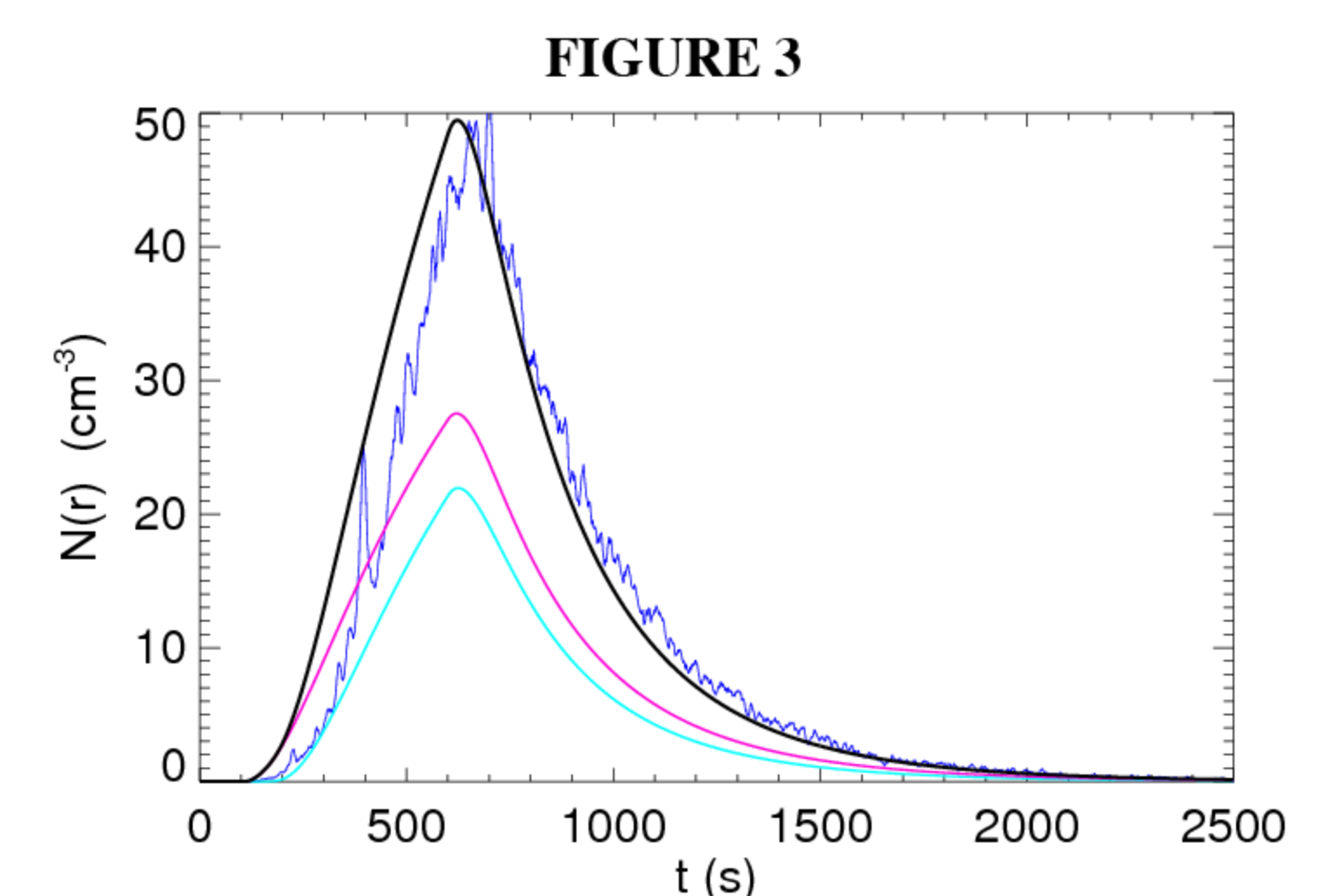
**Analytical model:** An analytical solution to the condensation equation has been found. Input parameters are loss rates, sulphuric acid concentration, and critical cluster size.  $J_1$  is kept as a free parameter and the value best fitting the measured peak is found. Using  $J_1$  from the low ionization measurements we can estimate the  $J_1$  resulting from ions.

## RESULTS

Figure 3 shows an example of output from the analytical model. The dark blue line is the data, which has been recorded with enhanced ionisation. The turquoise line is the calculated neutral contribution, the pink line is the ion contribution, and the black line is the combined result from the model. The sulphuric acid concentration was about  $5.5 \cdot 10^8 \text{ cm}^{-3}$ .

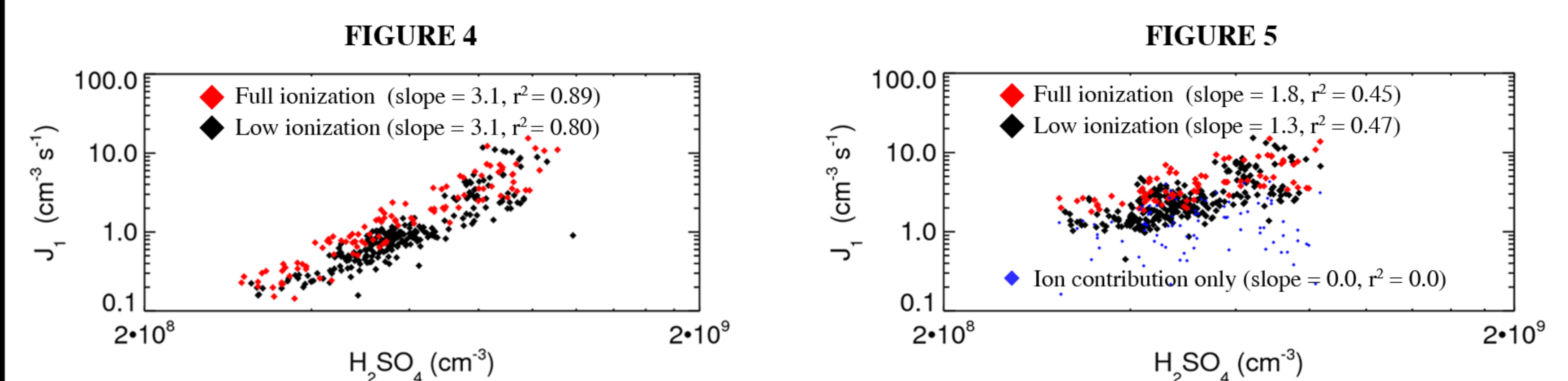
The fit reproduces the overall structure of the peak and the decay, but the initial rise appears a bit early, which is due to the model predicting an early rise of the aerosols formed by ions.

Growth rates for charged particles are enhanced according to Yu and Turco, JGR 106, 2001.



A total of 258 peaks with minimal ionization and 110 with full exposure to the gamma source have been analysed. Figure 4 and 5 show the output from the two methods of analysis, with Fig. 4 containing results from the loss rate method and Fig. 5 from the analytical model. The true nucleation rate  $J_1$  is shown as a function of the sulphuric acid concentration, in a log-log plot.

The slope of the plots are 3.1 for both lines in Fig. 4. In Fig. 5 we get 1.8 and 1.3 for the low ionisation and full ionisation, respectively. Interestingly the ion contribution seems to be more or less independent of the sulphuric acid concentration. The difference between the two methods makes any definitive conclusion difficult, however, the results from the analytical method are more in line with other observations that yield slopes between 1 and 2.



## AKNOWLEDGEMENTS

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