Extending Estimating Hydrogen Content in Atom Probe Tomography Experiments where H_2 Molecule Formation Occurs

Supplementary Material

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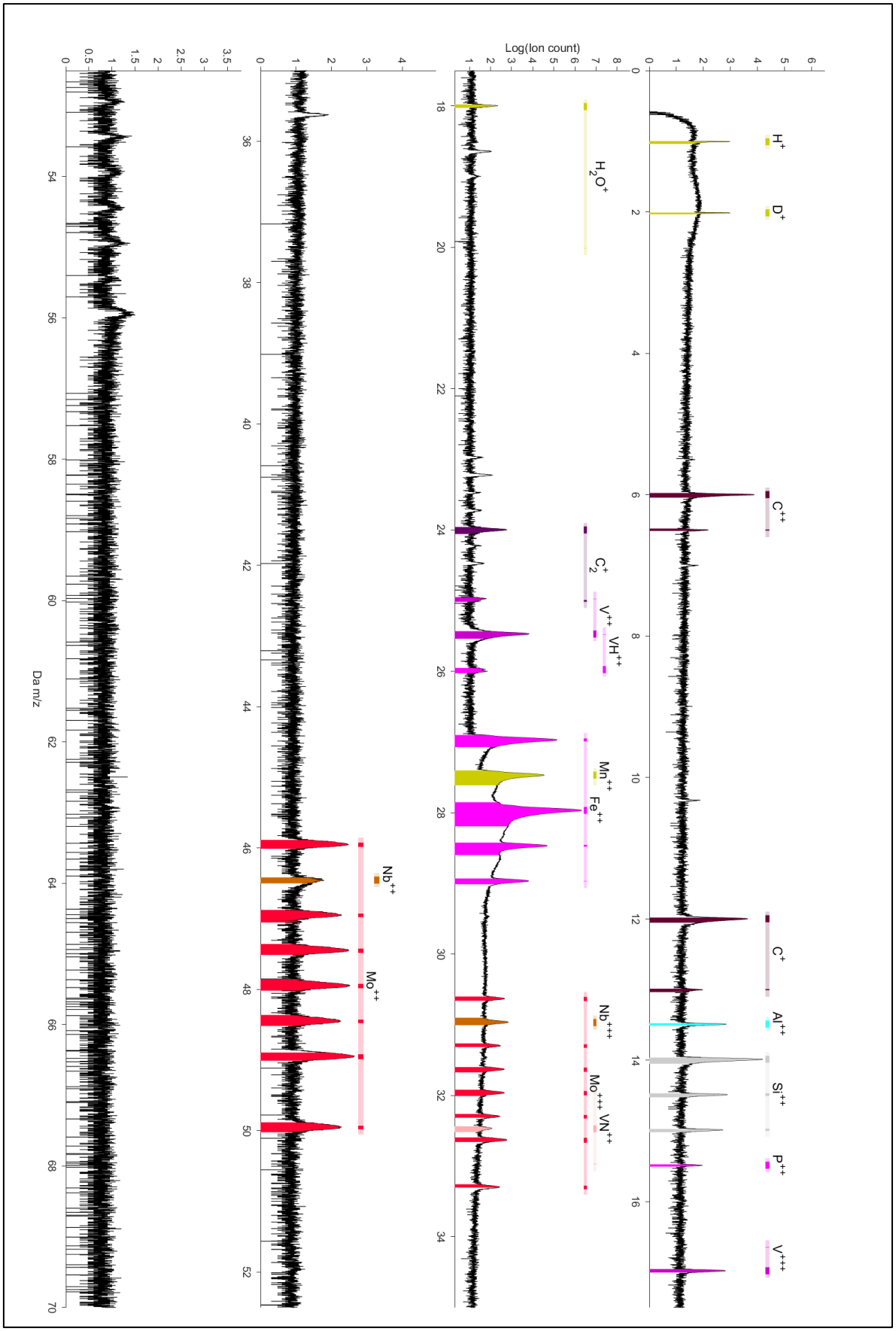
S1: Nominal Composition of the sample Material

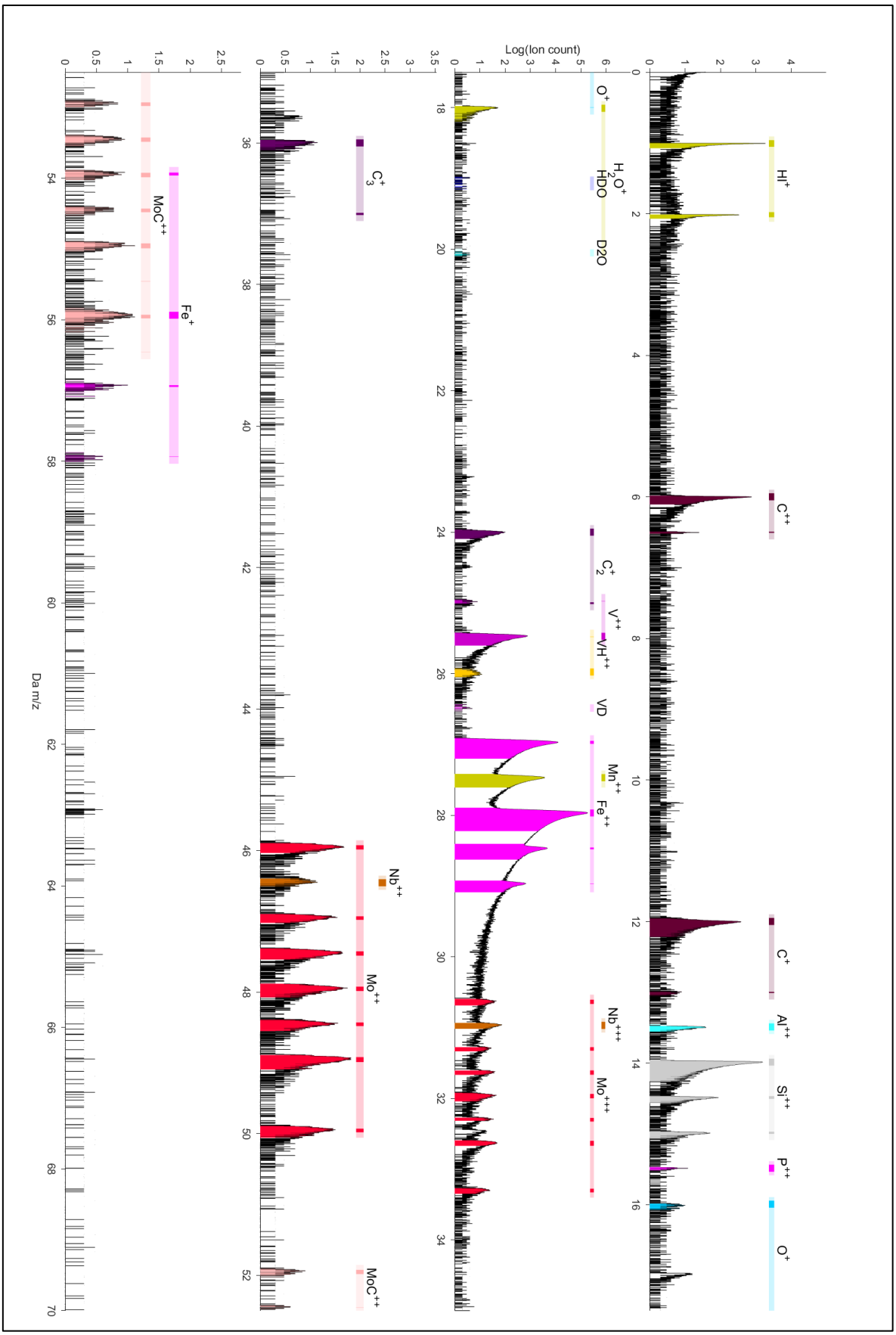
Component	Mn	Mo	V	C	Nb	Al	Si	N	Fe
Weight-%	1.56	0.51	0.25	0.096	0.056	0.05	0.026	0.004	Balance
Atom-%	1.59	0.30	0.27	0.45	0.034	0.1	0.05	0.02	Balance

Nominal composition of the sample material

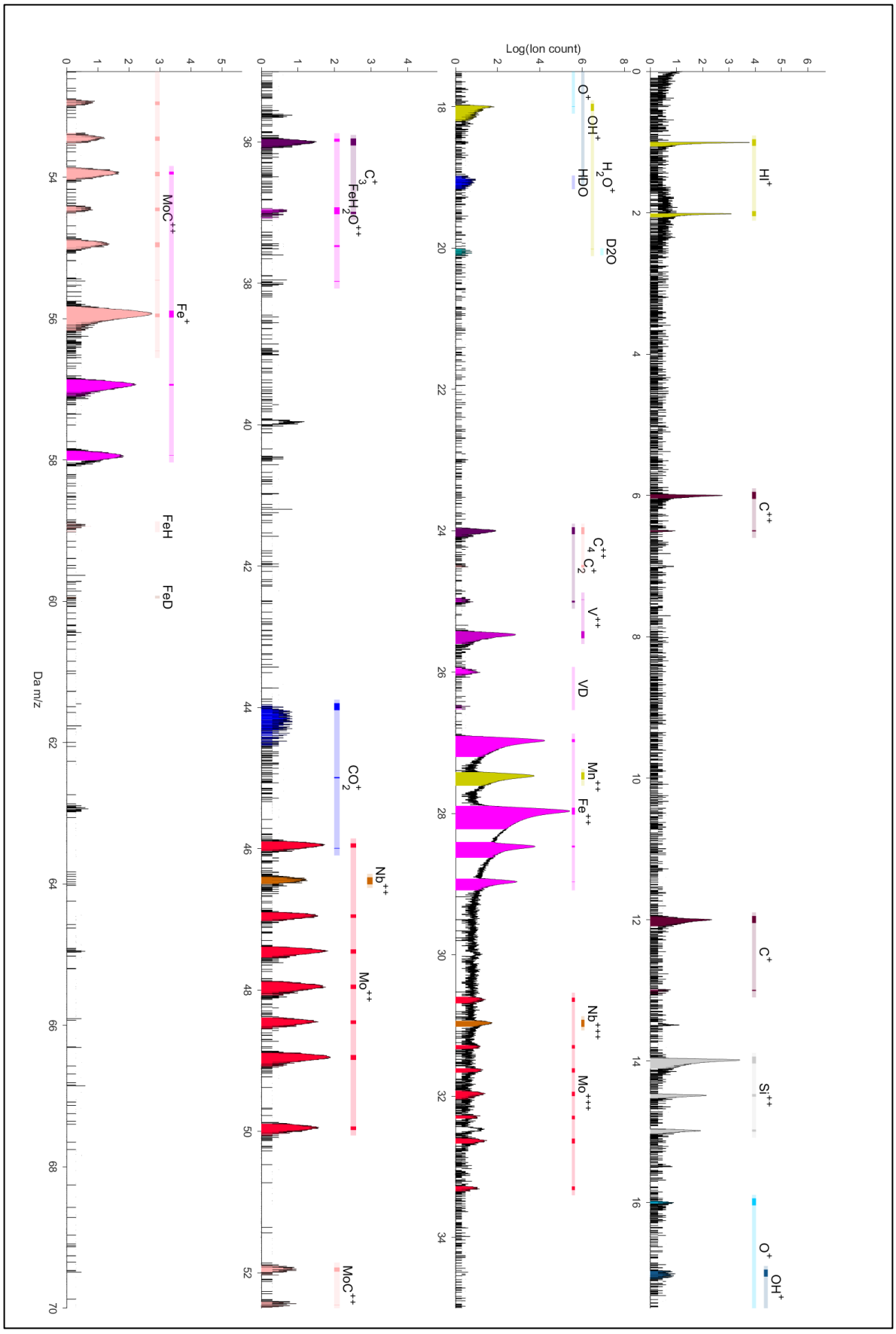
S2: Mass-charge spectra of deuterated Steel Samples, Ratio method



A: Spectrum of deuterated steel, pulsed voltage evaporation

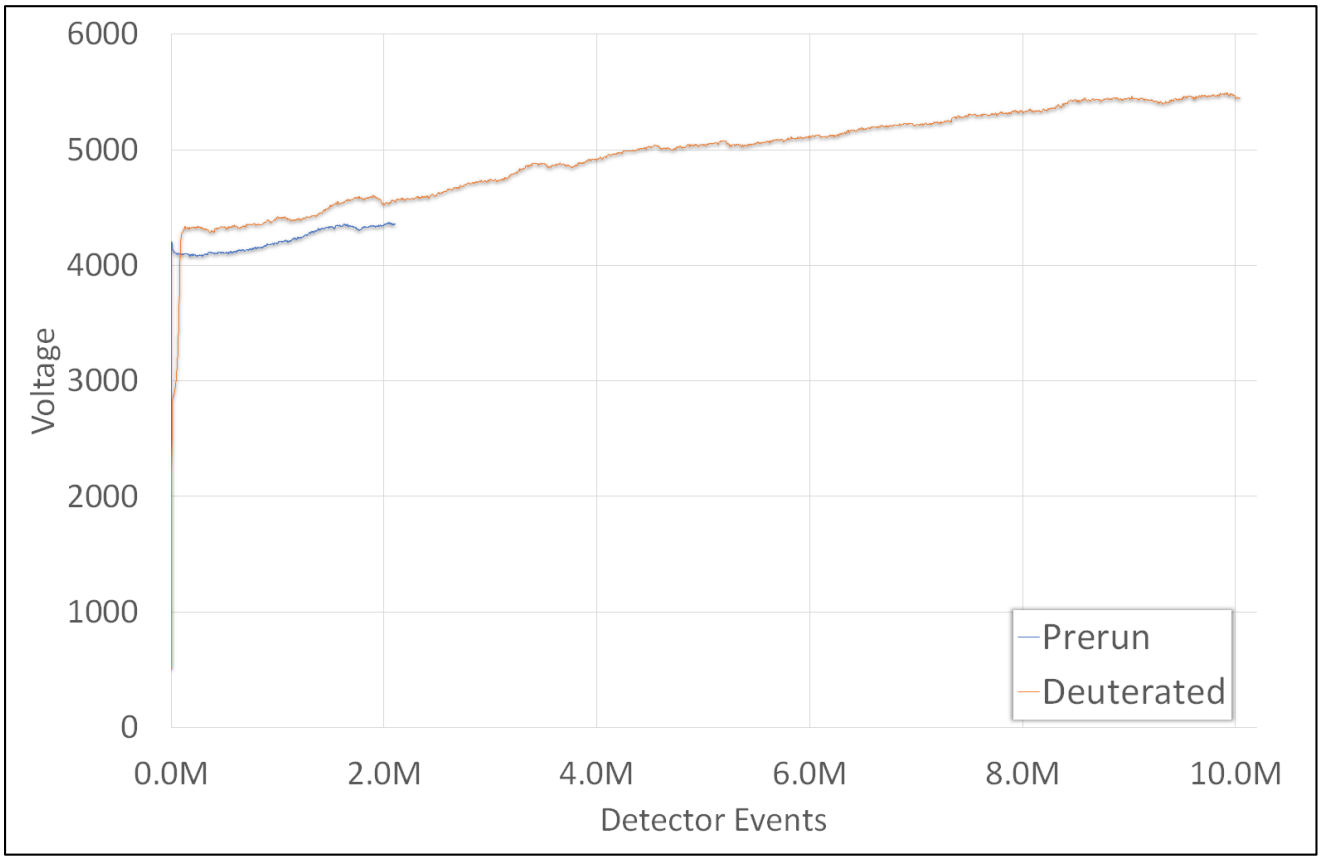


B: Spectrum of deuterated steel, laser assisted evaporation, 0.2 nJ

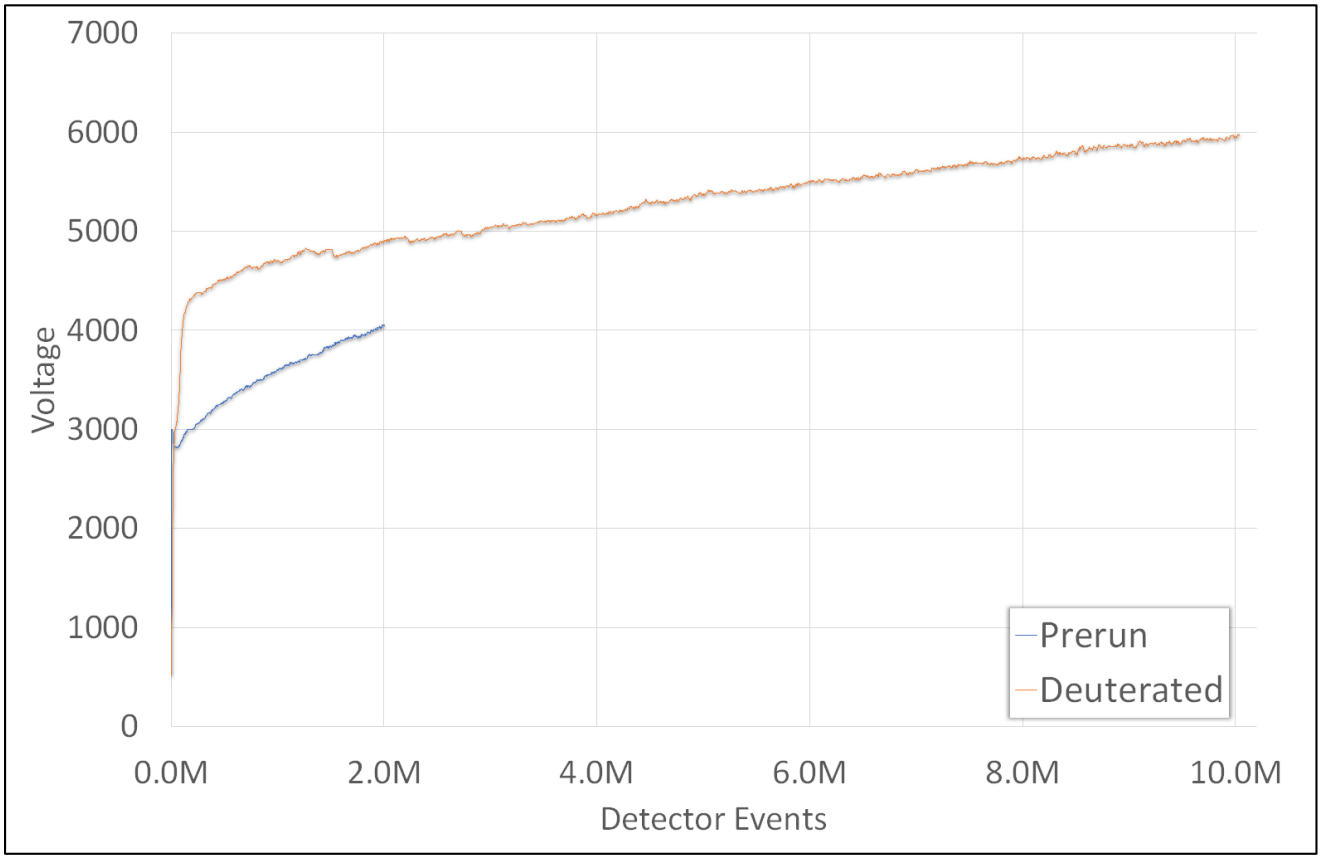


C: Spectrum of deuterated Steel, laser assisted evaporation, 0.4 nJ

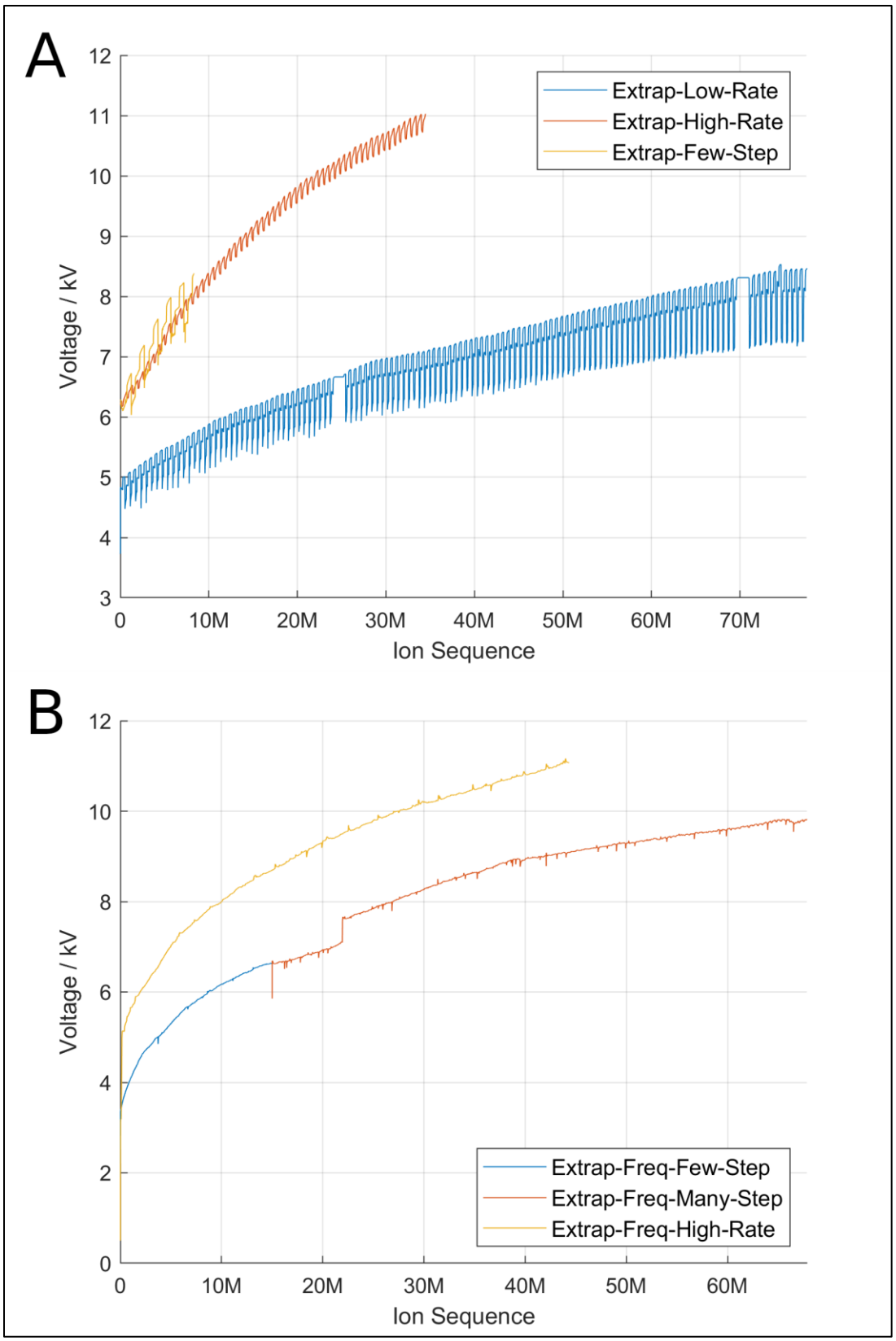
S3: Voltage Curves



A: Voltage curves in prerun and deuterated experiment at 0.2 nJ laser energy (Ratio-Low-Laser)

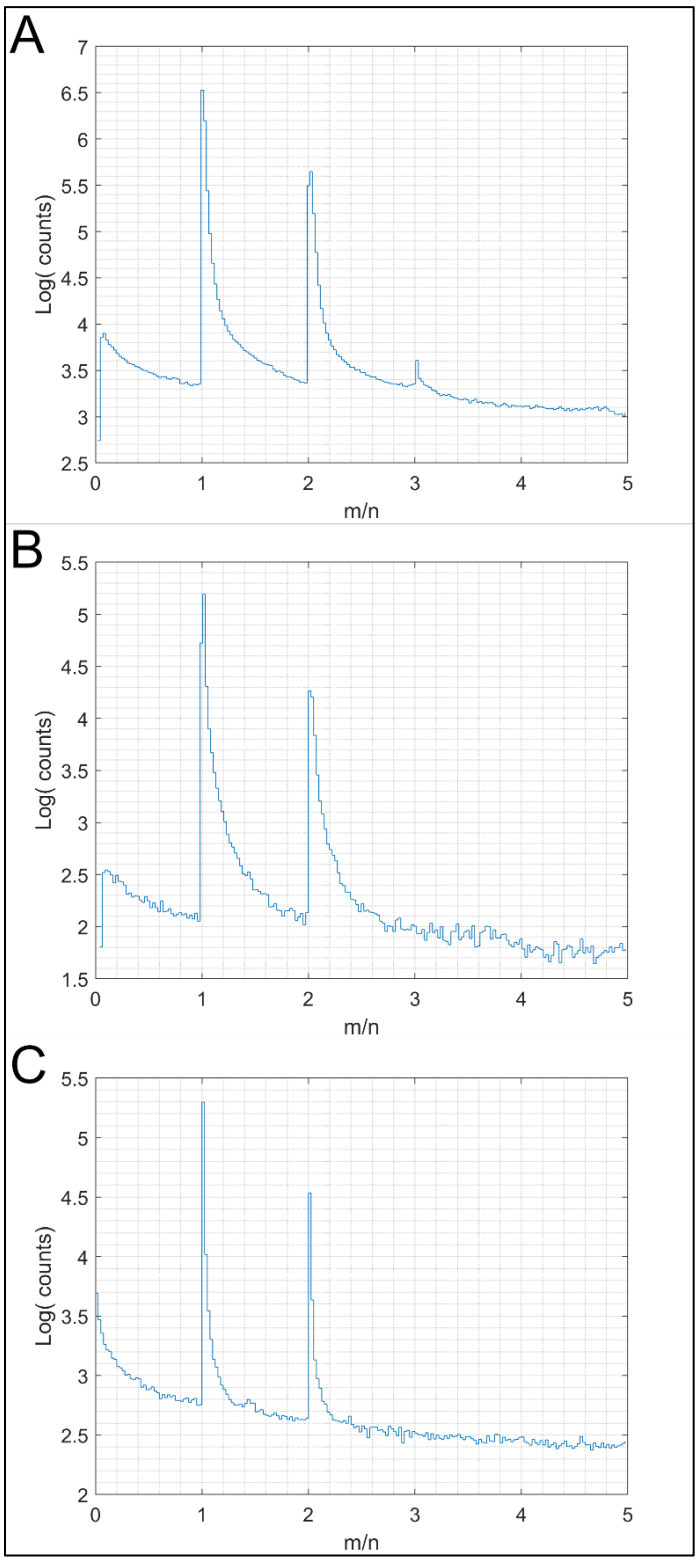


B: Voltage curves in prerun and deuterated experiment at 0.4 nJ laser energy (Ratio-High-Laser)

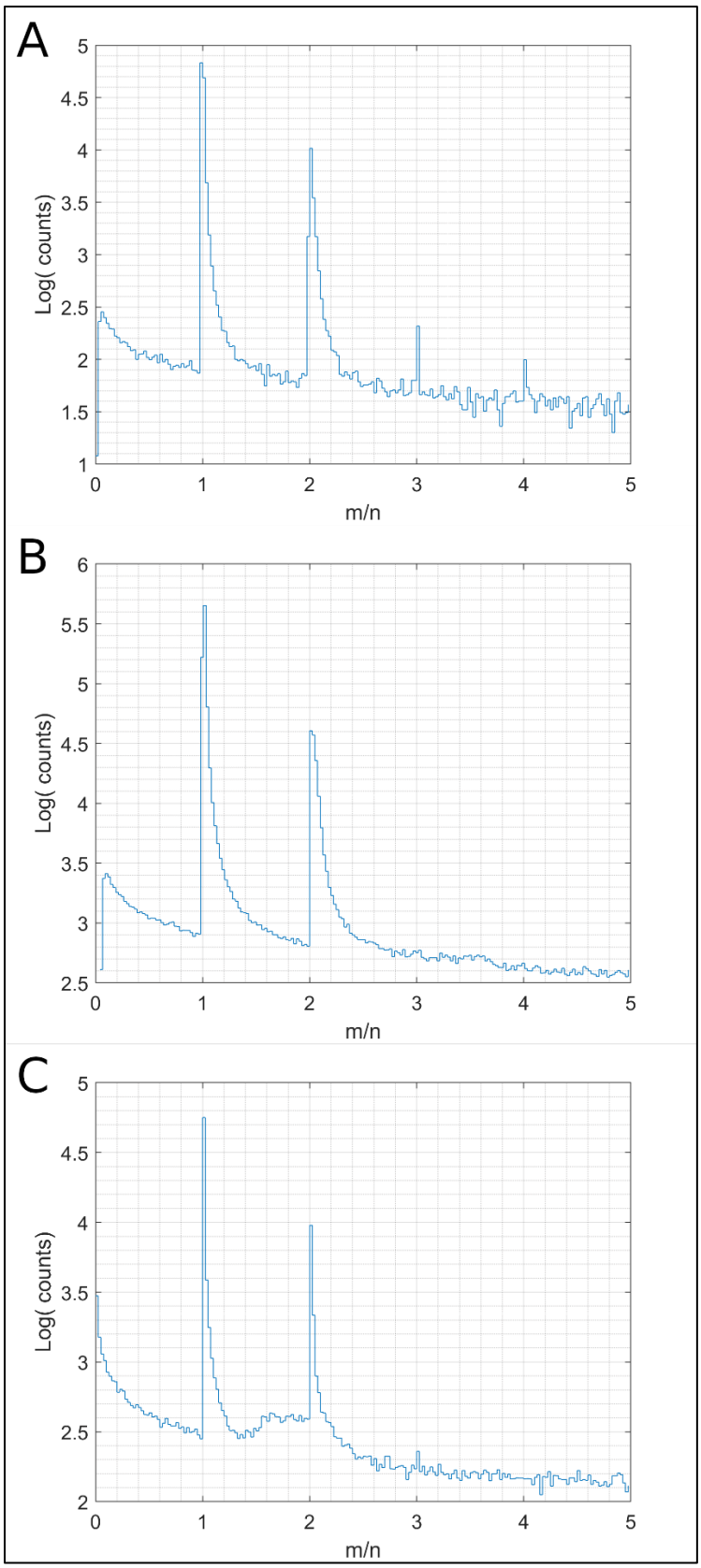


Voltage experiments with cycled evaporation rate (A) and cycled frequency (B). it is seen that the cycling of the evaporation rate influenced the voltage in the cycled Evaporation rates experiments and lead to a “ripple”, while the cycled frequency curves are smooth. In A, two short phases during the Extrap-Low-Rate where the voltage adjustment failed are seen. In B, the experiments Extrap-Freq-Few-Step and Extrap-Freq-Many step are merged into one curve as both experiments were conducted sequentially on the same tip. A small partial fracture in the shape of a short, steep voltage increase is seen in Extra-Freq-Many-Step.

S4: Mass-charge spectra of Steel samples, Cycled Evaporation Experiments



Hydrogen Spectra of Vanadium Carbide Steel samples in cycled evaporation rate experiments, A: Experiment with Detection rate cycled between 0.1%, 1%, 10% every 200000 ions (Extra-Low-Rate, evaporation rate 0.125%, 1.25%, 12.5%), B: Experiment with evaporation rate cycled between 3%, 8%, 10% aver 200000 ions (Extrap-High-Rate, evaporation rate 3.75%, 10%, 15%), C: Experiment with Detection rate cycled between 0.07%, 0.7%, 7% every 500000 ions (Extrap-Few-Step, evaporation rate 0.125%, 1.25%, 12.5%).



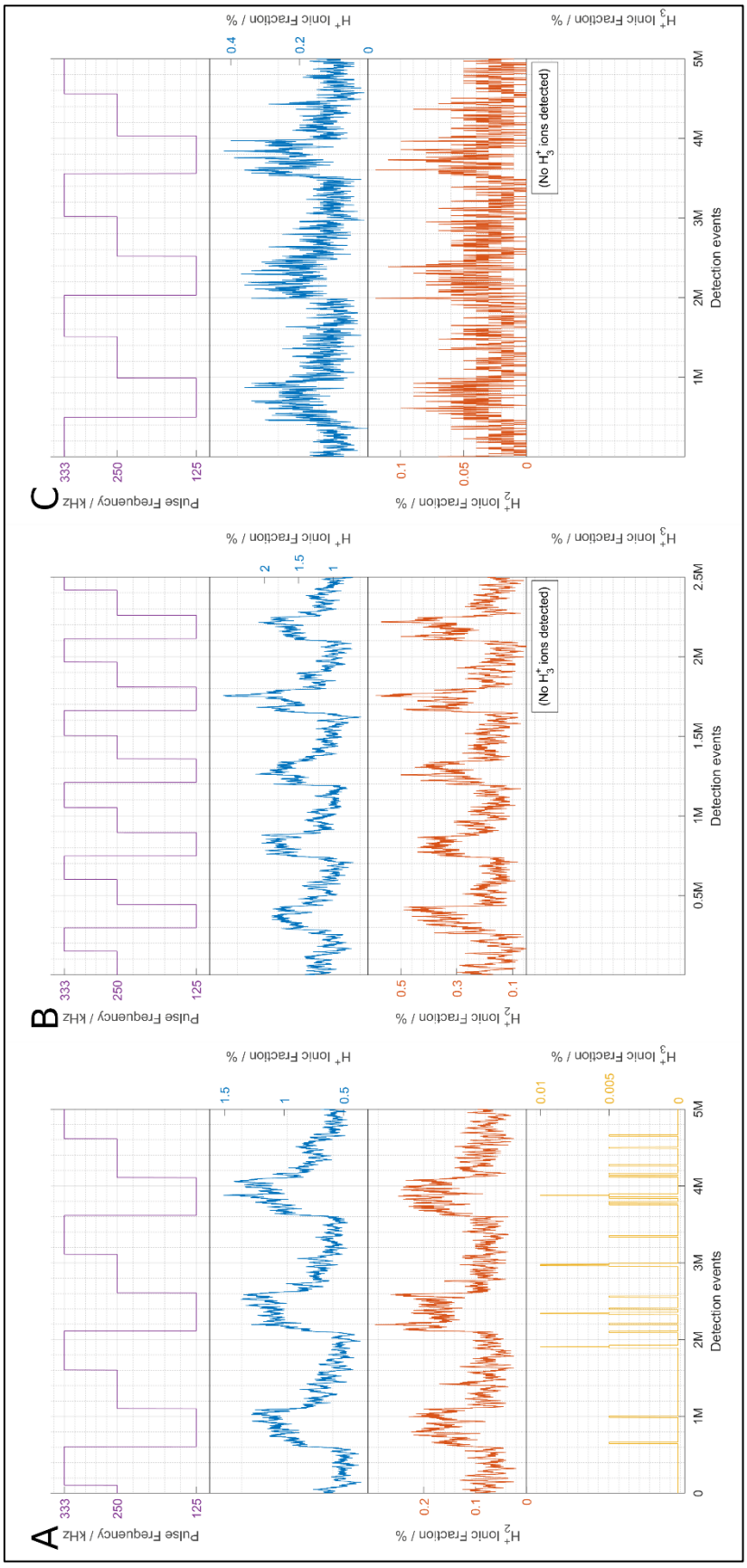
Hydrogen Spectra of Vanadium Carbide Steel samples in cycled frequency experiments, A: Experiment with Frequency cycled between 125, 250, 333 kHz every 500000 ions (Extra-Freq-Few-Step, Target Evaporation Rate 1.25 %) B: Experiment with frequency cycled between 125. 250, 333 kHz every 150000 ions (Extrap-Freq-Many-Step, Target Evaporation rate 1.25 %), C: Experiment with frequency cycled between 125, 250, 333 kHz every 500000 ions (Extrap-High-Rate, Target Evaporation Rate 10 %).

S5: Target and Actual Rates, Cycled Evaporation Experiments

	Low-rate experiment Extrap-Low-Rate			Low-rate experiment Extrap-High-Rate			Low-rate experiment Extrap-Few-Step		
Target detection rate	0.1 %	1 %	10 %	3 %	8 %	12 %	0.07 %	0.7 %	7 %
Average observed (actual) detection rate	0.1 %	0.86 %	5.3 %	3.5 %	5.7 %	8.6 %	0.07 %	0.71 %	5.64 %
Target evaporation rate	0.125 %	1.25 %	12.5 %	3.75 %	10 %	15 %	0.125 %	1.25 %	12.5 %
Average observed (actual) evaporation rate	0.130 %	1.08 %	6.7 %	4.3 %	7.1%	10.8 %	0.125 %	1.27 %	10.1 %

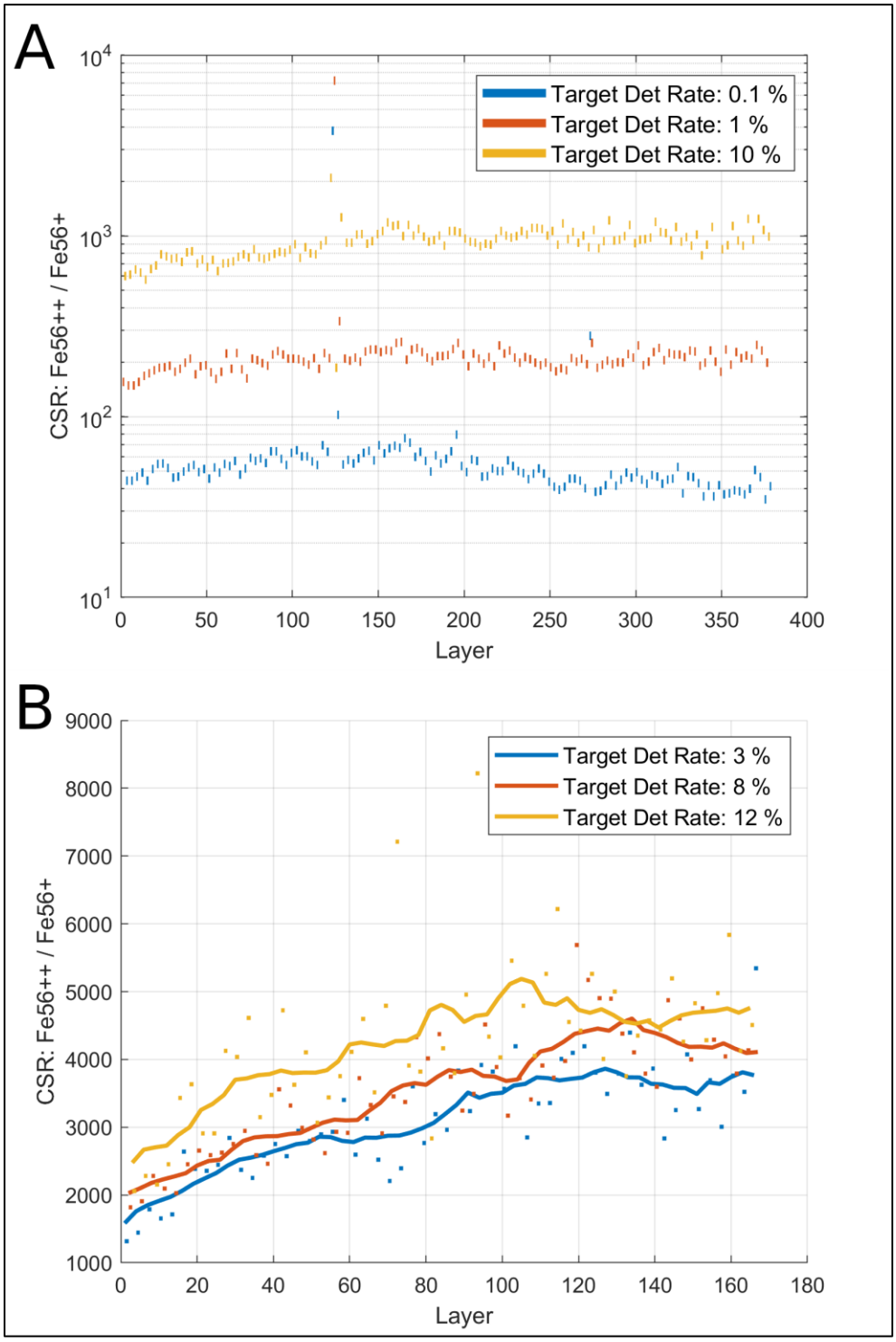
Target and observed detection and the corresponding evaporation rates in the cycled evaporation rate experiments. It can be seen that target and observed rates deviate further at high rates than at low rates.

S6: Hydrogen evolution in cycled Frequency experiments

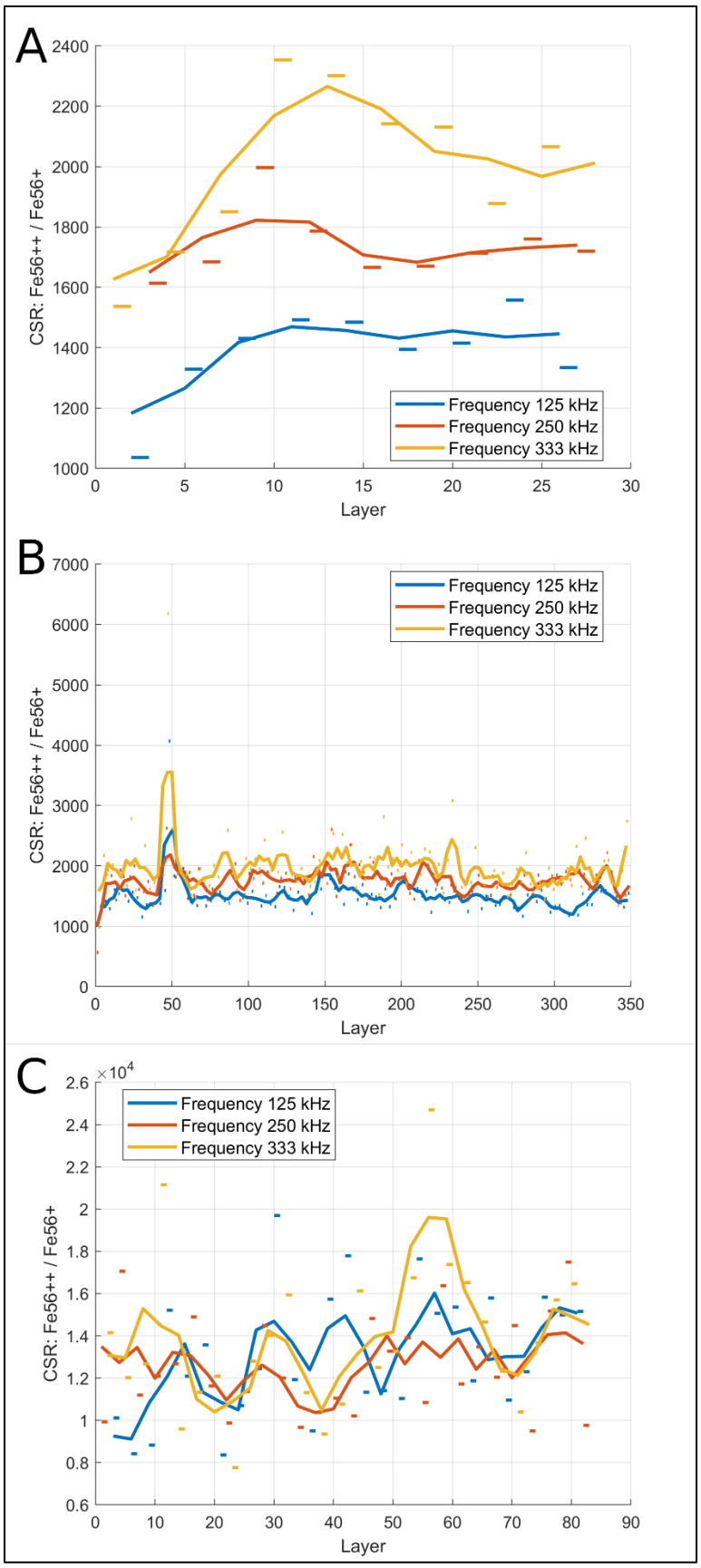


Hydrogen evolution in the cycled frequency experiments Extrap-Freq-Few-Step (A), Extrap-Freq-Many-Step (B) and Extrap-Freq-High-Rate (C). It is seen that the concentration of H^+ shows some tail-off after changes in the frequency in A and B, but no such observation in made in H^+ in C or in any of the H_2^+ curves

S7: Charge State Ratio Curves

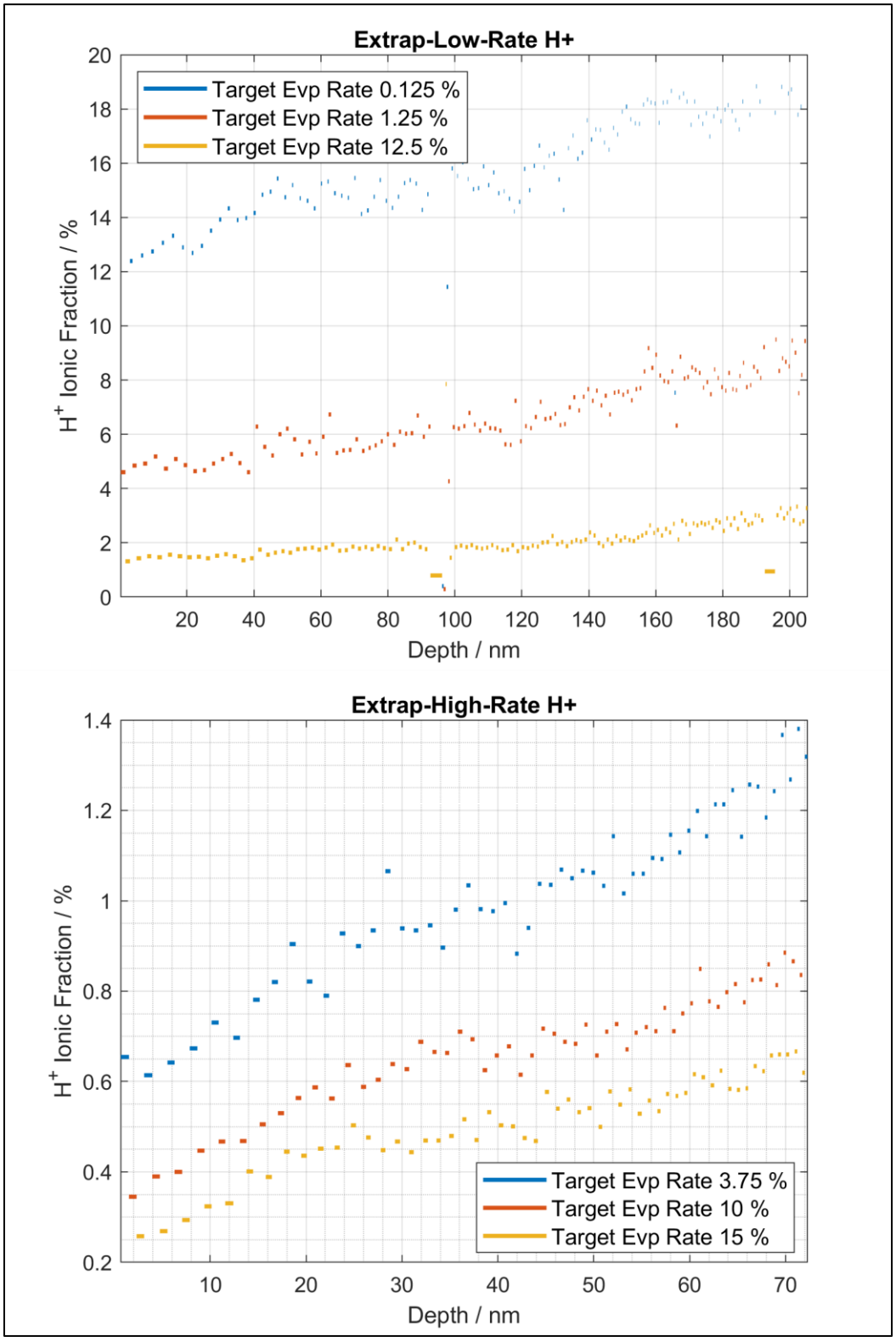


Charge-Statio ratios (CSR) of the Fe-56 isotope over the layers in the cycled evaporation rate experiments Extrap-low-Rate (A) and Extrap-Hogh-Rate(B). For Extra-High-Rate, the moving average across 10 layers for each target detection rate is included. It is seen that the CSR for the different target detection rates in Extrap-Low-Rate are very distinct, meanwhile the trend in Extrap-High-Rate is less prominent.

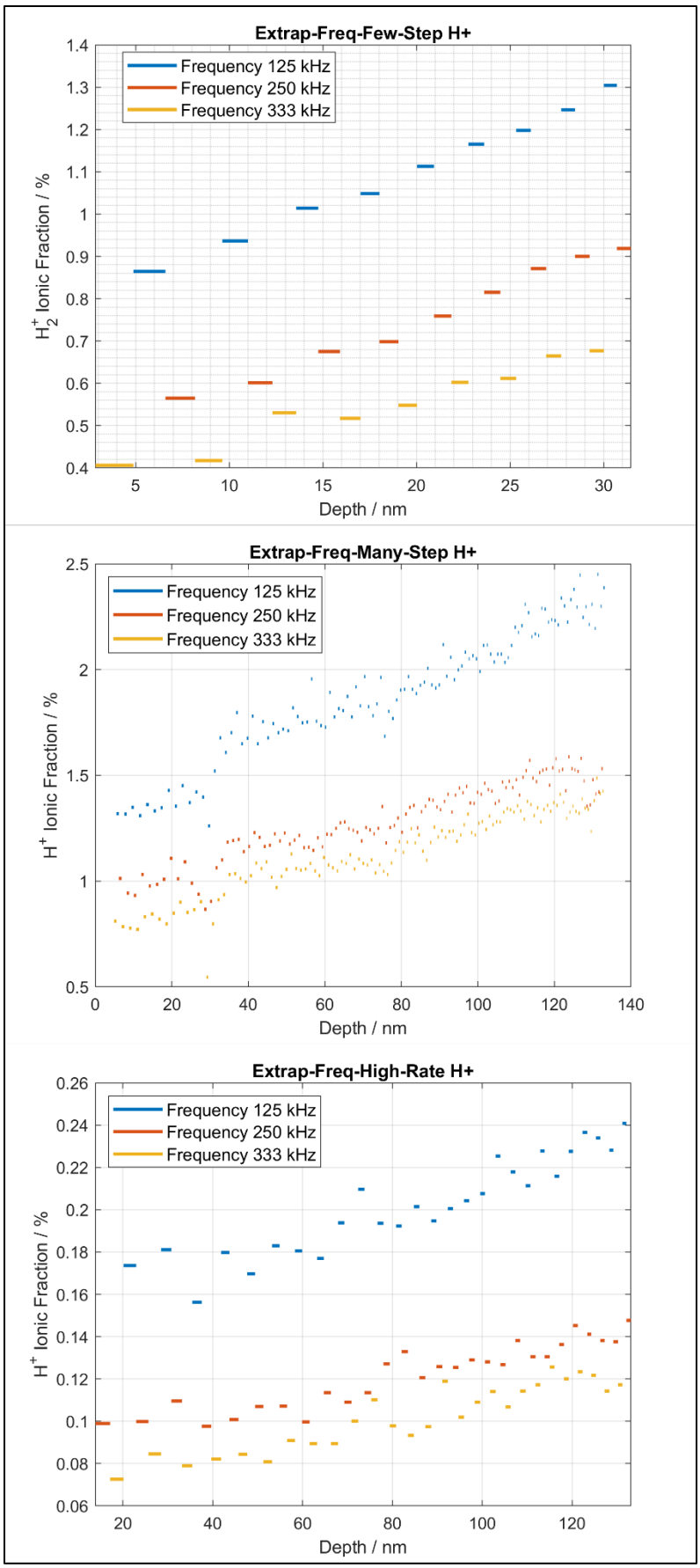


Charge-Statio ratios (CSR) of the Fe-56 isotope over the layers in the cycled frequency experiments Extrap-Freq-Few-Step (A) and Extrap-Freq-Many-Step(B) and Extrap-Freq-High-Rate (C), and moving average CSR across three layers for each frequency. It is seen that the CSR for the different target detection rates in Extrap-Freq-Few-Step and Extrap-Freq-Many-Step are slightly different while n0o such trend is seen in Extrap-Freq-High-Rate.

S8: Measured H+ content throughout experiments

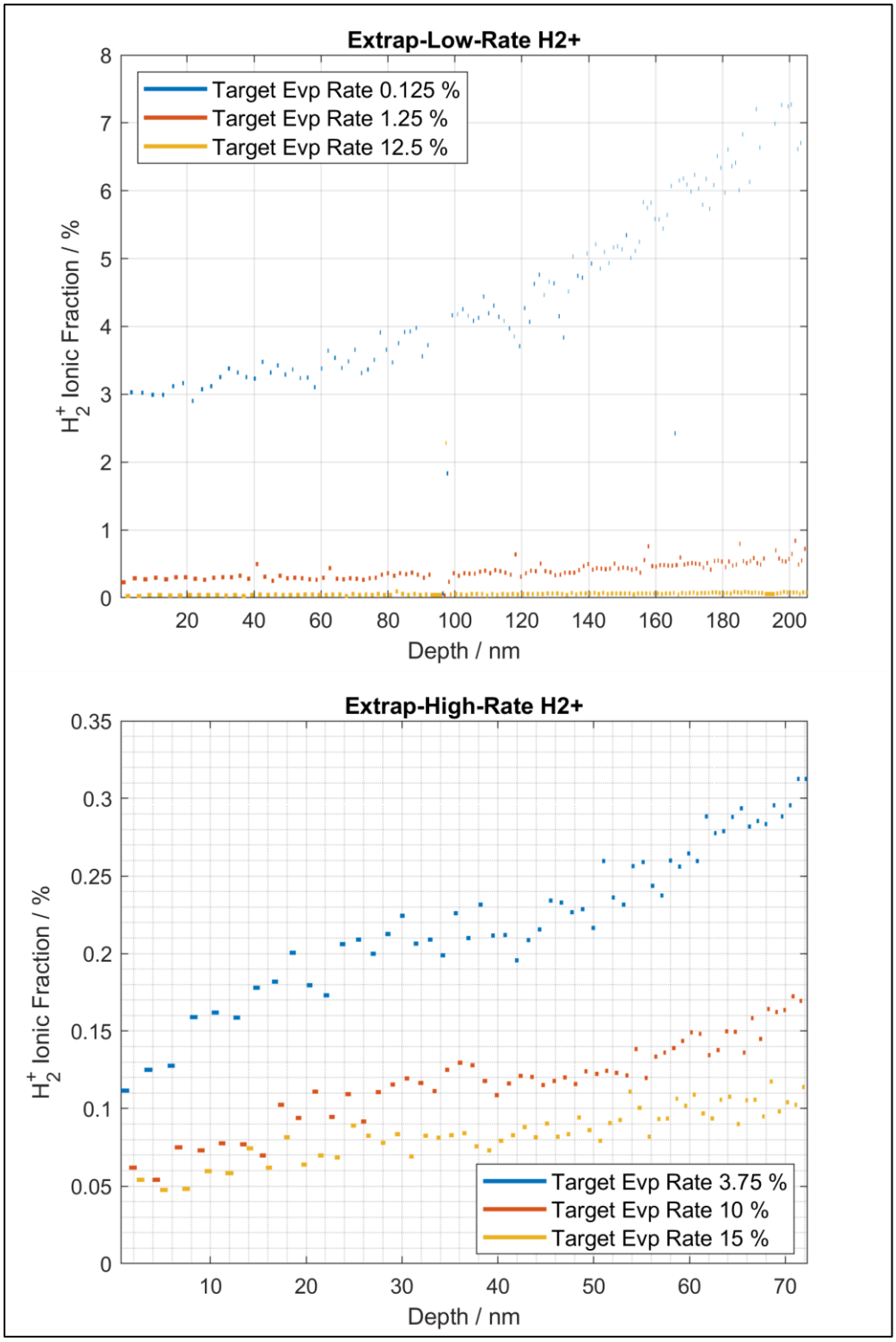


Detected ionic fractions of H^+ throughout the layers in the cycled evaporation rate experiments. It is seen that the amount of H^+ is clearly different for the different target evaporation rates and increases as the experiments progress.

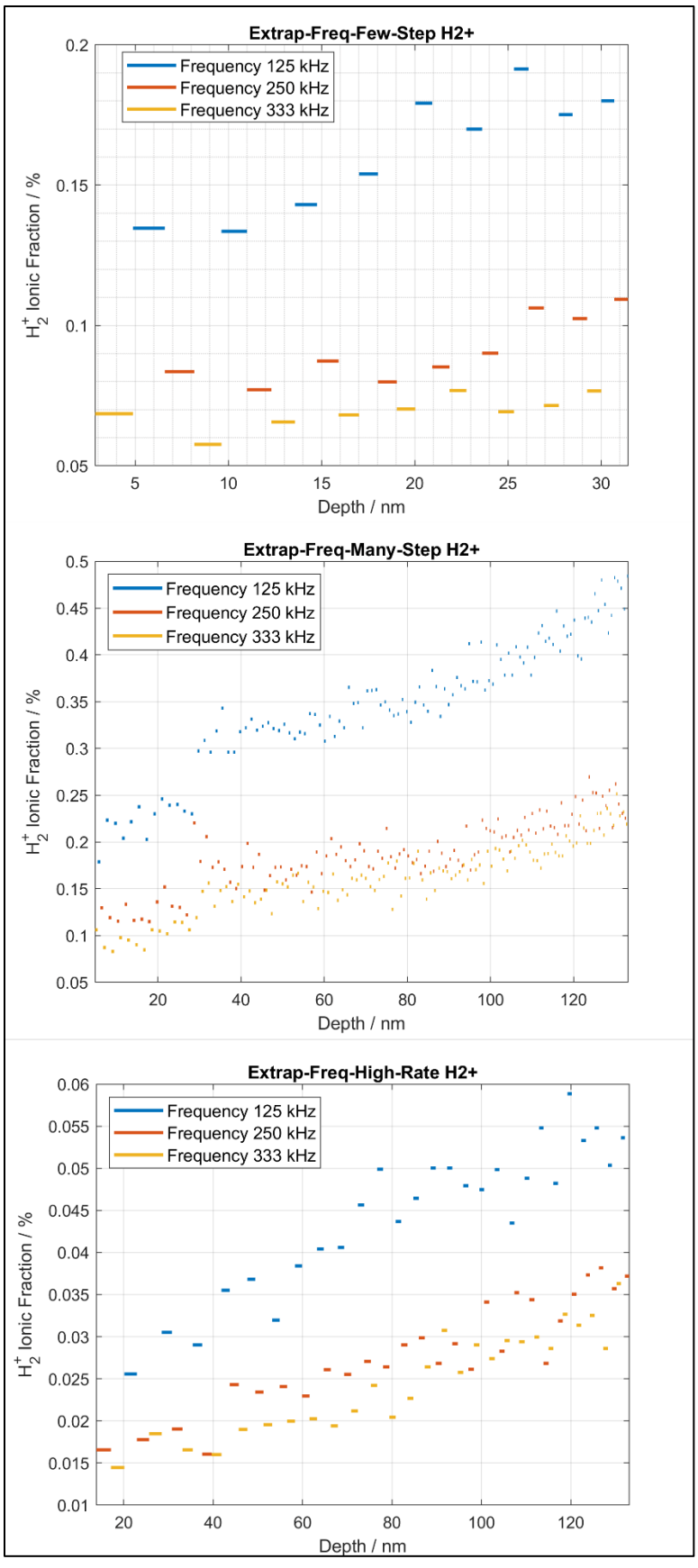


Detected ionic fractions of H^+ throughout the layers in the cycled frequency experiments. It is seen that the amount of H^+ is clearly different for the different frequencies and increase as the experiments progress, similar to the cycled evaporation rate Experiments.

S9: Measured H2+ content throughout experiments

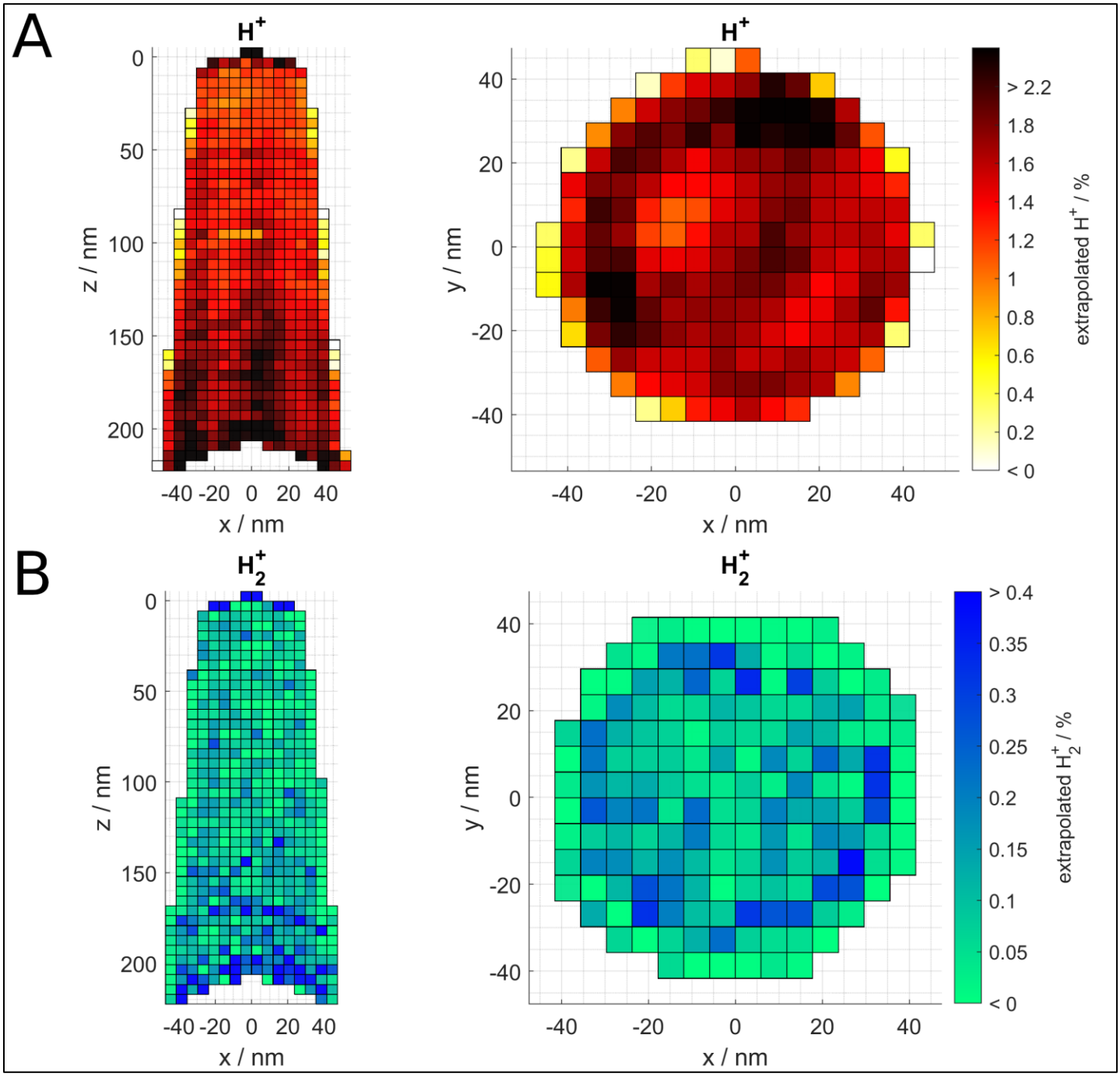


Detected ionic fractions of H_2^+ throughout the layers in the cycled evaporation rate experiments. Similar to the H^+ fractions (Supplementary Material S8), the amounts of H^+ are different for the different evaporation rates and increases as the experiments progress.



Detected ionic fractions of H_2^+ throughout the layers in the cycled frequency experiments. Similar to the H^+ fractions (Supplementary Material S8), the amounts of H^+ are different for the different evaporation rates and increases as the experiments progress.

S10: Voxel-based extrapolation in Extrap-Low-Rate



Voxel-based extrapolation of H^+ (A) and H_2^+ (B) in Extrap-Low-Rate. Structures resembling patterns from pole segregation are observed.