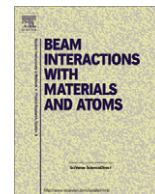




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Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimbAMS of ^{36}Cl with the VERA 3 MV tandem accelerator

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ABSTRACT

Recent progress with compact ionization chambers has opened new possibilities for isobar suppression in accelerator mass spectrometry (AMS). Separation of ^{36}Cl ($t_{1/2} = 0.30$ Ma) at natural isotopic levels from its stable isobar ^{36}S became feasible at particle energies of 24 MeV, which are also accessible for medium-sized tandem accelerators with 3 MV terminal voltage like VERA (Vienna Environmental Research Accelerator). Investigations with an ionization chamber revealed how physics favors isobar separation even at energies below the maximum of the Bragg curve. The strong energy focusing effect at high energy losses reduces energy straggling significantly and isobar separation steadily increases up to almost full energy loss. With an optimized detection setup, sulfur suppression factors of 2×10^4 have been achieved.

Refraining from the additional use of degrader foils has the benefit of high transmission to the detector (~16%), but requires a low sulfur output from the ion source. Therefore several backing materials have been screened for sulfur content. The dependence of the sulfur output on the AgCl sample size has been investigated as well.

Precision and accuracy have been thoroughly assessed over the last two years. Since drifts in the spectra are efficiently corrected by monitoring the position of the ^{36}S peak, the reproducibility for high ratio samples ($^{36}\text{Cl}/\text{Cl} > 10^{-12}$) is better than 2%. Our blank value of $^{36}\text{Cl}/\text{Cl} \approx (5 \pm 5) \times 10^{-16}$ is competitive to other labs. ^{36}Cl has become a routine AMS-isotope at VERA.

Recently we also explored novel techniques for additional sulfur suppression already in the ion source. While results with a small gas reaction cell in front of the sputter target were discouraging, a decrease in the sulfur/chlorine ratio by one order of magnitude was achieved by directing 300 mW continuous wave laser beam at 445 nm towards the cathode in the ion source.

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1. Introduction

Isobar suppression by utilizing the different energy loss of ions in matter is limited for medium-sized AMS facilities like VERA with 3 MV terminal voltage. When the stable isobar forms negative ions, only light AMS-isotopes like ^{10}Be – with a large relative difference in atomic number from the isobar ^{10}B – could be measured [1]. Heavier radionuclides like ^{36}Cl , where ^{36}S also forms negative ions, were out of reach until a few years ago. Recently however, the use of very homogeneous SiN-foils as windows for gas ionization chambers has opened many new possibilities for particle identification in AMS at low energies [2,3], since the energy straggling introduced is significantly smaller than with e.g. mylar foils [4]. A good overview of relevant physical processes in gas ionization chambers at lower ion energies is given in [5].

Pushing VERA to 3.5 MV terminal voltage and using terminal foil stripping allowed to perform the first exposure dating measurement of ^{36}Cl with a (nominal) 3 MV facility [6]. With an extended detection setup, the same isobar separation was already achieved at the nominal 3 MV terminal voltage [7]. Improvements of the ion source and its regulation as well as the target geometry were other important steps towards routine measurements of ^{36}Cl . To optimize the performance of the detection setup, the main physical processes governing the isobar separation of ^{36}Cl and ^{36}S in gas ionization chambers were studied, especially the influence of the detector gas. Some advances have already been published in [8], others are shown below. The present work focuses on isobar separation below the maximum of the Bragg curve and its consequences for the detection setup and the performance of ^{36}Cl measurements at 3 MV terminal voltage. With a well-established setup, the separation of ^{36}Cl and ^{36}S also provides a good test case for the application of novel methods for isobar suppression with reactive gases or laser optical filtering and interesting first results have been achieved.

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2. Isobar suppression below the Bragg maximum

To first order, the capability of distinguishing particles of interest from interfering background by their difference in energy loss when passing through matter is determined by the ratio $\Delta E_{12}/\sqrt{\delta E_1^2 + \delta E_2^2}$, where ΔE_{12} is the total difference in energy loss of the two species (the distance between the peaks in the

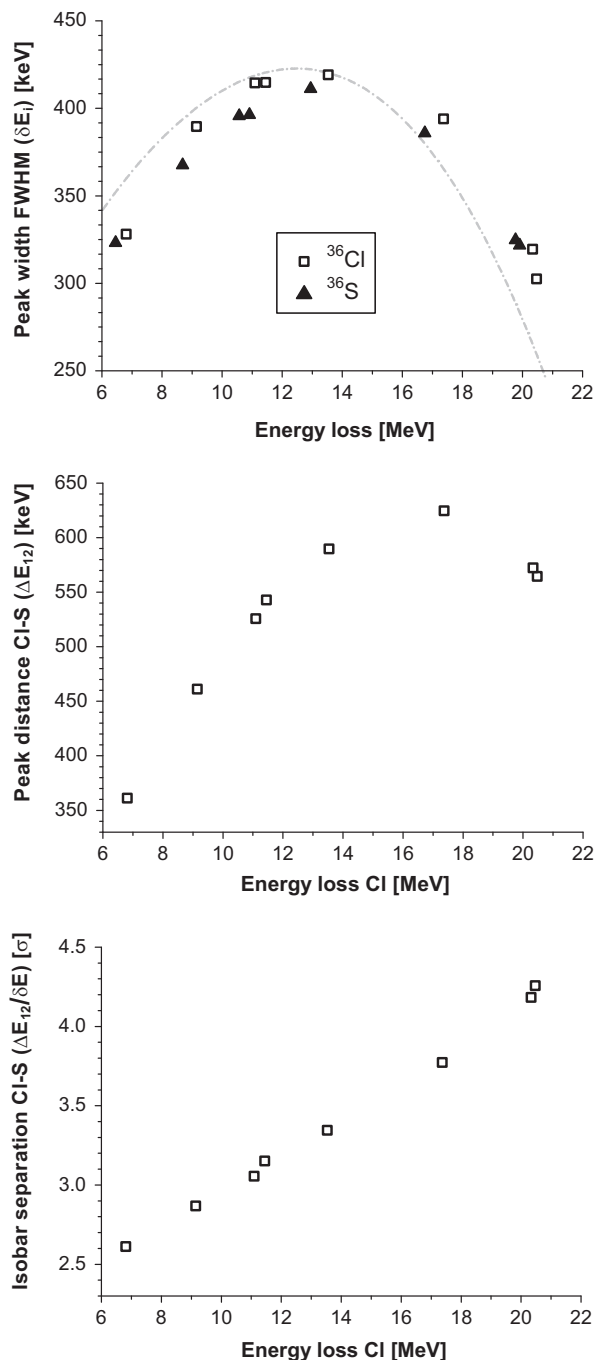


Fig. 1. Isobar separation of ^{36}Cl and ^{36}S in isobutane at 23.4 MeV initial energy after passage through the detector window. The peak widths (energy straggling, δE_i), the distance between the two peaks (ΔE_{12}) and the isobar separation $\Delta E/\delta E$ (with $\delta E = \sqrt{\delta E_1^2 + \delta E_2^2}$) are plotted as a functions of energy loss. The gray dotted line in the upper figure shows the estimate of the peak width based on the semi-empirical straggling formula for Cl by [11]. Below the maximum of the Bragg curve, the energy focusing effect at high energy losses leads to an increase in isobar separation even after the crossing point of the energy loss curves at ~ 16 MeV energy loss. The best separation is achieved at almost full energy loss.

spectrum) and δE_i are the widths of the individual peaks (determined by energy loss straggling and the energy resolution of the detection setup). For isobar suppression with the $\Delta E - E$ method in an ionization chamber, ΔE_{12} is given by the difference in energy loss due to different stopping powers. With a specific initial particle energy, ΔE_{12} can only be optimized by changing either the gas pressure or the lengths of the anodes, which both also affects the widths δE_i of the measured energy loss distributions. In order to find an optimum configuration, the widths δE_i have to be studied more closely. For all but the lightest ions, preamplifier noise and straggling in the entrance window are negligible even below 1 MeV initial particle energy and the energy resolution is therefore limited by the contribution of the detector gas [5]. With particle energies of ~ 24 MeV for ^{36}Cl and ^{36}S , the dominant process for energy loss in the gas is the interaction of the projectile's screened nuclear charge with the target electrons (electronic stopping). Only at the very end of the particle range, a significant contribution to the energy loss comes from nuclear stopping. A widely used approach to describe energy straggling in electronic stopping is the Bohr formula [9], where the straggling δE is proportional to the square root of the target thickness and the square root of the target's atomic number, but independent from the incident energy (strictly, this is only true for high particle energies where the projectile is fully stripped). Since the energy straggling always increases along the flightpath ($\delta E \propto \sqrt{\Delta E}$), it would be best for maximum isobar separation to measure up to the crossing point of the respective energy loss curves (maximum distance of peaks). After the crossing point, the distance of peaks starts to decrease and the ever increasing straggling deteriorates the separation even more. However, at lower energies and higher energy losses, the variation of the stopping power S with decreasing energy along the ion's flightpath has to be taken into account [10]. Below the Bragg maximum the stopping power is decreasing with decreasing energy and the correlation of energy losses in various intervals results in a focusing effect. Ions with lower energy loss than average in the past intervals of the target experience a higher stopping power in the coming section and vice versa. Schmidt-Böcking and Hornung [11] experimentally verified this effect and also gave a formula for it $\delta E_{\text{corr}} = \delta E * \frac{S(E_0 - \Delta E)}{S(E_0)}$ (δE_{corr} is the correct estimate of energy straggling, δE the straggling estimated from the straggling in a thin layer, e.g. Bohr straggling, and S the stopping power at the given energy).

Fig. 1 shows our experimental results for δE_i and ΔE_{12} of ^{36}S and ^{36}Cl at an initial energy of 23.4 MeV after passage through the gas detector window (3 MV terminal voltage) as a function of energy loss obtained with isobutane as detector gas. Below the Bragg maximum, the strong decrease in energy straggling at high energy losses more than compensates for the decrease in distance of peaks after the crossing point of the energy loss curves and the isobar separation still increases. The highest isobar separation is achieved at almost full energy loss. This allows high isobar suppression factors already at relatively low initial energies. Energy tails in the spectrum, that arise at almost full energy loss, can deteriorate the separation if the ion of interest has an atomic number higher than its isobar (which is the case for chlorine and sulfur). These tails can be suppressed by a special detection setup described below.

3. Experimental setup

3.1. Detection setup, data acquisition and drift correction

We currently use two detectors to discriminate between ^{36}Cl and ^{36}S : a split-anode ionization chamber with silicon nitride entrance and exit window (5×5 mm, 100 nm thickness) and a 256

pixel silicon strip detector for residual energy measurement. Accepting only events that produce signals in both detectors allows to efficiently remove high energy tails in the ionization chamber spectra caused by angular scattering. We investigated several detector gases with respect to energy straggling and angular scattering and now use isobutane with 30% argon. This counting gas provides a sulfur suppression of 2×10^4 at 80% transmission through the ionization chamber and 50% total Cl detection efficiency. More details on our detection setup and the properties of the counting gases can be found in [8]. The data acquisition system records all events of both sulfur and chlorine in the detectors to allow post-measurement analysis of the data including changes of the region of interest. Sample spectra from a reference material with an isotopic ratio of $^{36}\text{Cl}/\text{Cl} \approx 10^{-11}$, a blank with no ^{36}Cl and a true sample with $^{36}\text{Cl}/\text{Cl} \approx 10^{-13}$ are shown in Fig. 2. Apart from two energy loss signals from the ionization chamber (ΔE_1 , ΔE_2) and the residual energy in the silicon strip detector (E_{res}), we also measure the pulse width of the ΔE_1 signal at 10% of the signal height to reject disturbing pile-up events. In order to mimic the pulse height of a ^{36}Cl event, two sulfur atoms have to enter the detector with a certain time in between them such that the second signal still coincides with the tail of the first signal. The resulting pulse has the same height as a ^{36}Cl event but a very long width at the 10% level and is thus easy to identify. At higher count rates this pile-up rejection enhances the ^{36}S suppression usually by roughly 50%.

Since the ^{36}Cl region of interest needs to be very tight for decent sulfur suppression and thus $\sim 30\%$ of the ^{36}Cl events are cut away, drifts in the spectra caused by electronics or changes in the detector gas can easily deteriorate precision. Even with the ionization chamber filled and closed completely, we typically observe drifts in the spectra of 2 channels (3‰) per day. Therefore we implemented a special drift correction where the position of the ^{36}S peak, which is much more pronounced than the ^{36}Cl peak, is continuously evaluated and all spectra are shifted accordingly. The limitations of this method were tested by operating the detector with gas through-flow and a pressure regulation that achieves a constant pressure within 1%. The results are shown in Fig. 3. The pressure fluctuations caused by the regulator lead to drifts in the

uncorrected spectra of ± 4 channels within 100 s. With the drift correction enabled, the peak positions are stable with a deviation of less than ± 0.5 channels. This clearly demonstrates how the drift correction can even compensate short fluctuations in the spectra making it an important tool to achieve good overall precision.

Together with 16% yield for the 7+ charge state in terminal foil stripping, the average sulfur output of $^{36}\text{S}/^{35}\text{Cl} \approx 5 \times 10^{-11}$ from good samples (see below) corresponds to a detector count rate of ~ 500 Hz at $10 \mu\text{A } ^{35}\text{Cl}^-$ current. For samples with higher sulfur content, the count rate in the detector sets an upper limit for the beam current during measurement. Despite the pile up rejection described above, the maximum count rate acceptable for our detection setup is ~ 5000 Hz. Higher count rates result in a decreased isobar separation, probably due to a deteriorated charge collection in the ionization chamber caused by the remaining positive ions in the gas. In addition, dead time issues and limitations of our data acquisition system start to play a role at these count rates.

3.2. Ion source, backing material and sample size

Natural ^{36}Cl samples require good chemistry for sulfur removal and the use of appropriate materials for a low sulfur output from the ion source. A “clean” Cl-beam is all the more important if no isobar separation technique (e.g. degrader foil [12]) prior to the detector is used. Precipitation of chlorine as silverchloride after sulfur removal by precipitation of BaSO_4 is an established method [13] and big AgCl targets (>20 mg) show very small sulfur content [6]. However the sulfur output from the ion source strongly depends on the sulfur content of the surrounding material and the size of the AgCl-sample (or more precisely the area covered with AgCl). Therefore most labs use special cathode and/or backing materials (for an overview see Table 1 in [14]) and require ~ 4 –6 mg AgCl per sample, although only a small fraction is used up during a measurement. We screened several backing materials (see Table 1) and got the best results with silver bromide produced in-house at VERA from KBr following the procedure for AgCl described above. In another study, the lower limit of sample size was investigated. Output currents of several $\mu\text{A } ^{35}\text{Cl}^-$ at typical source settings were achieved even for 1 mg samples, however the sulfur output

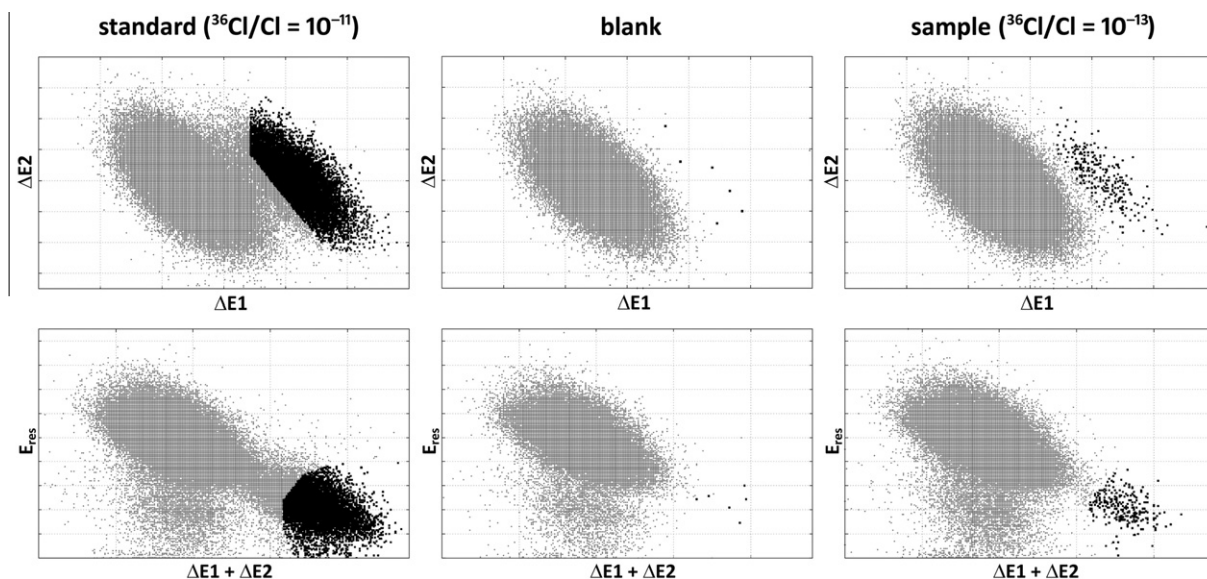


Fig. 2. Sample spectra recorded on three different AgCl samples. Only ions that produced a signal in the silicon strip detector and passed the pile-up rejection criteria are plotted. ΔE_i are the energy loss signals from the two anodes of the ionization chamber and E_{res} is the residual energy signal from the strip detector. All events (mostly sulfur) are plotted in gray and the events in the tight ^{36}Cl region of interest are plotted in black. The chlorine and sulfur peaks are better separated in the total energy loss signal $\Delta E_1 + \Delta E_2$ than in either the ΔE_1 or ΔE_2 signals. The 6 counts on the blank sample correspond to a background of $^{36}\text{Cl}/\text{Cl} \approx 4 \times 10^{-15}$ (prior to sulfur-induced background correction).

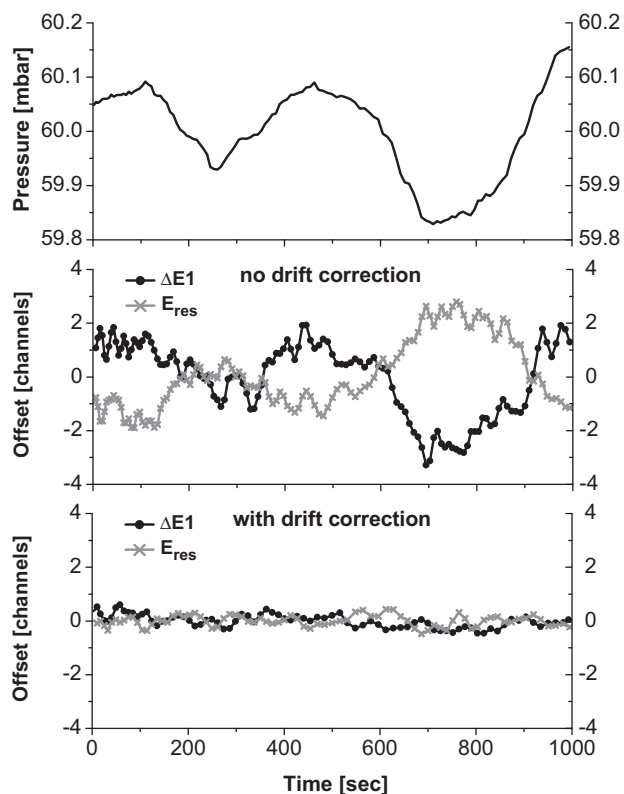


Fig. 3. Drift of the position of the sulfur peak in the energy loss spectra over time with and without peak drift correction. The upper graph shows the intentionally unstable pressure in the ionization chamber over 1000 s. The latter two graphs show the respective drift of the sulfur peak position both for the ΔE_1 signal from the ionization chamber and the residual energy signal from the silicon strip detector E_{res} . Positive offset corresponds to higher energy loss, thus ΔE_1 and E_{res} offsets are anticorrelated. Apparently, the drift correction can compensate even for fast drifts and significantly enhances precision of the measurement.

Table 1

Average sulfur output from the ion source using various cathode and backing materials. All cathodes contained ~ 4 mg of the same AgCl blank material and were sputtered for at least 30 min prior to measurement to remove surface contamination. Best results were achieved with AgBr produced at VERA.

Cathode material	Backing material	Average sulfur output ($^{36}\text{S}/^{35}\text{Cl}^-$) [$\times 10^{-10}$]
Cu	VERA-AgBr	0.5
Cu	Commercial AgBr	2
Al	Ta plate	5
Al	Ta plate (H-baked)	4
Ni	None	6

increases significantly with decreasing sample size. The average sulfur output for various sample sizes is summarized in Table 2. For samples with a $^{36}\text{Cl}/\text{Cl}$ ratio of 10^{-12} or above a higher sulfur induced background is usually acceptable, provided that the ^{36}S count rate is still manageable for the detection system. Samples below 0.5 mg AgCl have been measured successfully. For low ratio samples on the other hand, the increase in sulfur induced background is of course significant and a reduction of sample size below 2 mg AgCl for routine measurements seems only feasible with additional means of sulfur suppression.

For 3 mg AgCl-samples we measure ionization yields above 11–13%. There is usually a fair amount of residual sample material left in the cathode. The high sulfur count rate (>20 kHz) arising once

Table 2

Average sulfur output from the ion source as a function of sample size. All cathodes had the same AgBr backing and contained the same AgCl blank material. All targets were sputtered for at least 30 min to remove surface contamination. The sulfur output quoted is the average over all samples with the same amount of sample material, with at least 2 h of measurement time on each sample.

Sample size AgCl [mg]	Average sulfur output ($^{36}\text{S}/^{35}\text{Cl}^-$) [$\times 10^{-10}$]
> 4	0.5
~ 2	1
~ 1	6
~ 0.5	10

a hole has been sputtered through the AgCl layer, however, does not allow further measurements. At $10 \mu\text{A } ^{35}\text{Cl}^-$ current this corresponds to more than 6 h of available measurement time on each target. This is long enough to acquire ~ 1000 ^{36}Cl events (corresponding to 3% statistical uncertainty excluding sulfur induced background correction and normalization) on a sample with a $^{36}\text{Cl}/\text{Cl}$ isotopic ratio of 10^{-14} . Most targets are larger, thus allowing even longer acquisition times. The high grade of automation of our facility allows for such long measurements, even though only exceptional samples may justify this effort under routine conditions.

The issue of memory effect of our ion source has been discussed in [8] and the choice of appropriate standard material is crucial. In exposure dating, a validated technique to determine the amount of natural Cl in the rock sample is dilution with a ^{35}Cl -spike [15]. In this case, the memory effect also needs to be taken into account for measurement of the $^{35}\text{Cl}/^{37}\text{Cl}$ isotopic ratios.

4. Status and performance

Over the last two years, ^{36}Cl has become a routine isotope at the VERA facility. The accelerator is operated at 3 MV terminal voltage and the ions are stripped to the 7+ charge state with terminal foil stripping, resulting in particle energies of 24 MeV. At $\sim 10 \mu\text{A } ^{35}\text{Cl}^-$ current, one stripping foil usually lasts for one week of measurement time. The terminal voltage is automatically retuned every 24 h to compensate for possible, however seldom occurring thickness changes of the foil. Initial tuning of the machine takes ~ 10 h and is performed with a semi-automated script and the optimization software “automax” [16]. Since part of the high energy side needs to be tuned with the $^{36}\text{S}^{7+}$ count rate in the detector, special AgCl targets containing ~ 50 ppm of AgS have been produced. Use of those special tuning targets assures that most of the sulfur in the beam originates from the sample itself and not from surrounding material and the accelerator is thus tuned for optimal transport of the sample material to the detector. The alternative use of other sulfur rich target materials such as copper or steel for tuning requires the use of a beam attenuator (a perforated steel shield) which, from our experience, significantly changes the emittance of the beam.

The 15–17% particle transmission into the detector compare favorably to other facilities (again, see Table 1 in [14] for details on other labs). The background level of $^{36}\text{Cl}/\text{Cl} \approx (5 \pm 5) \times 10^{-16}$ is also competitive to other labs. With the drift correction enabled, high ratio samples with $^{36}\text{Cl}/\text{Cl} > 10^{-12}$ are typically measured to 0.8% precision for a single cathode. Slight systematic discrepancies between various cathodes containing the same sample material have been observed. They probably originate from different shapes of the pressed AgCl and limit the overall reproducibility to $\sim 2\%$ for those high ratio samples. The reproducibility was derived from the standard deviation of the various results obtained for the same sample material in different beamtimes. All results were obtained on different sputter targets and normalized to the reference material for each beamtime. We consider this the best estimate of the

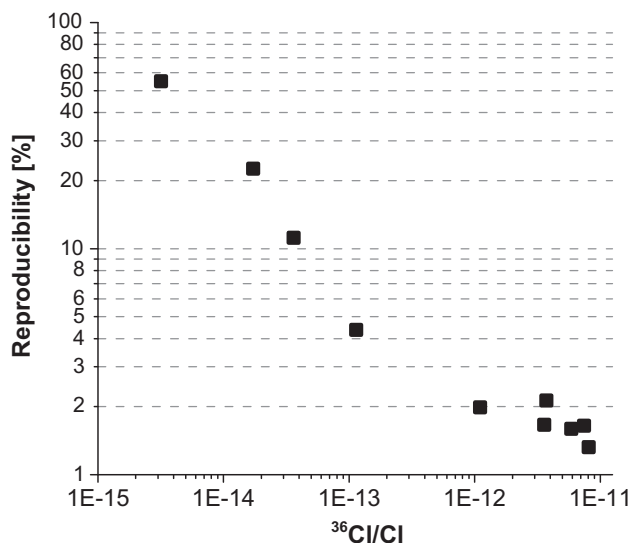


Fig. 4. Reproducibility of ^{36}Cl measurement results including sulfur induced background correction for various $^{36}\text{Cl}/\text{Cl}$ ratios. All samples plotted have been measured at least in five cathodes and in different beamtimes. For high ratio samples, reproducibility is around 2% and limited by systematic uncertainties (see also Fig. 5). For low ratio samples, counting statistics usually limits the precision.

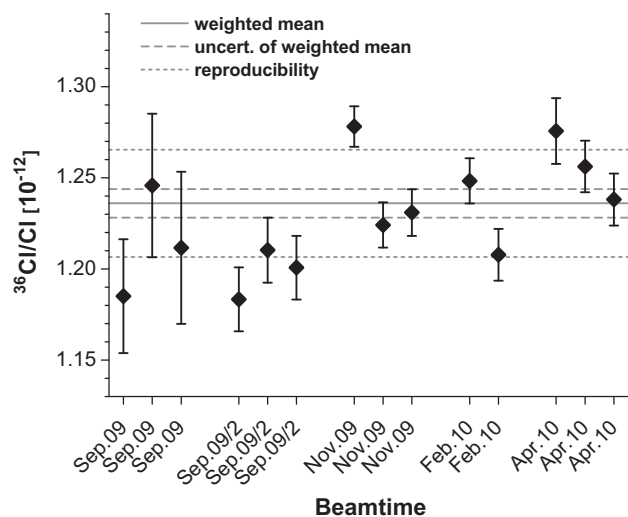


Fig. 5. Comparison of 14 measurement results (already normalized to reference material) for one sample material from several cathodes and beamtimes. The scatter between different cathodes in one beamtime is usually larger than the precisions on the single cathodes would suggest. This is probably due to different shapes of the pressed AgCl targets and limits the reproducibility. There is no additional statistically significant offset between results of various beamtimes.

accuracy of results on a single target of unknown sample material, provided that sufficient counting statistics is available. Fig. 4 summarizes our reproducibility. For samples below $^{36}\text{Cl}/\text{Cl} \approx 10^{-12}$, the reproducibility is mostly limited by the available acquisition time and counting statistics in a typical beamtime of 3 days for a 40 sample wheel (on average 1.5 h per target but adjusted according to the isotopic ratios). For very low ratio samples the uncertainty of sulfur induced background correction starts to play a role as well. Fig. 5 shows the long-term stability of our Cl-measurements over several beamtimes. Apart from the discrepancies between various cathodes mentioned above, there is no statistically relevant offset between the results of different beamtimes. The accuracy of our results has been demonstrated in an interlaboratory comparison [14]. Summarizing, ^{36}Cl measurements at VERA

are definitely competitive to other laboratories and several sets of exposure dating samples have been measured successfully.

5. New techniques for additional sulfur suppression

Currently, a further reduction in the amount of sample material required for a reliable measurement depends on the availability of additional sulfur suppression, preferably prior to the detector to reduce the count rate. Also samples with high sulfur content or isotopic ratios $^{36}\text{Cl}/\text{Cl} < 5 \times 10^{-15}$ would benefit significantly. A common method is the use of a degrader foil in front of an energy- or momentum-sensitive bending element [12]. However, this technique introduces quite high losses due to angular scattering and energy straggling in the foil as well as due to the restriction to only one of the various charge states after the foil. In recent years, two different approaches using gas filled radiofrequency quadrupole ion-coolers yielded sulfur suppression factors $>10^3$ with less than 50% loss of Cl-beam in demonstration experiments: element-selective laser photodetachment of negative ions [17] and the use of charge transfer in a NO_2 filled gas reaction cell [18]. Despite the impressive results none of the two techniques has been applied during real AMS measurements so far. Both techniques require a specially designed injector to accommodate the RFQ-cooler and the total throughput of the cooler is limited to about 10 nA of stable isotope current.

Since one order of magnitude in sulfur suppression would be sufficient for our needs, we have tried to implement both techniques directly inside a standard cesium sputter ion source. In a first approach, a modified cathode arrangement of the sputter source was built. It allows gas to be leaked into a cylindrical gas reaction cell (6 mm long and 4 mm diameter) in front of the sputter target. The reaction cell is formed by a metal cap mounted onto the sample holder. Its interior is dominated by a weak focusing fringe field (~ 30 V from the sample surface to the center of the cell) reaching all the way in from the exit opening. The effect of ~ 0.1 mTorr of NO_2 at the target surface, decreasing through the cell to 10^{-5} mTorr, on the $^{36}\text{S}/^{35}\text{Cl}$ ratio was studied. This was the maximum pressure that allowed operation of the ion source, above, discharges in the source hampered operation. Since NO_2 is a very corrosive gas, the cesium oven was disconnected during this measurement. Still, output currents above $\sim 1 \mu\text{A}$ $^{35}\text{Cl}^-$ current were achieved at ~ 180 W ionizer power. Unfortunately, the sulfur/chlorine ratio from a sample containing AgCl with 1000 ppm AgS was found unaffected (within quite high uncertainties due to random variations) with the gas flow on. Only the total current output decreased by a factor of 5–10, probably due to collisional neutralization on the high density of gas in the source region. Therefore, we didn't further pursue this technique.

In another experiment at the GUNILLA facility in Gothenburg [19], the beam from a continuous wave 1 W blue laser beam was directed onto the sputter target via a mirror situated just outside the ion beam path. The estimated laser power at the target was 100–300 mW, thus comparable to the power transfer of the 3 keV Cs^+ beam. The wavelength of 445 nm (2.74 eV) lies between the electron affinities of sulfur (2.077 eV) and chlorine (3.613 eV), which is required for selective sulfur suppression by photodetachment. While several experiments have been performed with pulsed lasers and different goals in mind [20,21], this is to our knowledge the first experiment with a continuous wave laser in a standard cesium sputter ion source. The target material was AgCl with $\sim 10\%$ AgS. With the laser on, an increase in chlorine current and a decrease in sulfur output were observed, leading to a change in the sulfur/chlorine ratio by one order of magnitude, as shown in Fig. 6. The measured isotopic abundances correspond well to the natural abundances of sulfur and chlorine isotopes and make interference of molecular background (e.g. O_2) very unlikely. However, photodetachment of sulfur was ruled out as the cause for this

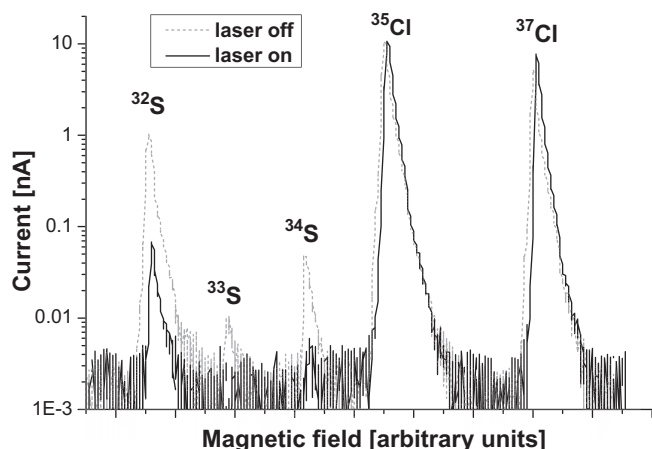


Fig. 6. Mass spectra from an AgCl target with ~10% AgS with (solid black) and without (dotted gray) ~200 mW of blue continuous wave laser (445 nm) focused onto the target in the ion source. The target was sputtered for several hours prior to measurement and the spectra were recorded with 5 min time interval. The laser was turned on 2 min prior to recording the respective mass spectrum. The increase in chlorine current can only be deduced from the ^{37}Cl peak since the ^{35}Cl current was already outside the limits of the amplifier. The laser reduces the sulfur/chlorine ratio by one order of magnitude.

change because of surprisingly long time constants in the order of minutes when the laser was turned on/off and also because similar results were achieved with an IR-laser (1.17 eV). At this point the effect is not understood but it has been recently reproduced for trace amounts of sulfur during a ^{36}Cl measurement at the VERA facility. More detailed results of these experiments in Gothenburg and Vienna will be published separately. Limitations and the underlying physics will be further explored as this method potentially provides the desired additional one order of magnitude sulfur suppression without Cl-beam loss.

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