We greatly appreciate the thoughtful comments from referee Daniel Wielandt, an expert in noble gas mass spectrometry who has published high-precision noble gas isotope ratio analyses on similar mass spectrometers. The review is reprinted below in its entirety in italics with author responses shown by indentation where appropriate.

Interactive comment on "The Isotopx NGX and the ATONA Faraday Amplifiers" by Stephen E. Cox et al.

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General comments: This is a well-written article on an important novel amplifier technology called ATONA that provides an otherwise currently unavailable combination of low noise and high dynamic range for Faraday cup measurements of ion beams. The technology could significantly improve both current and future mass spectrometers, and is therefore of general interest to all mass spectrometry specialists. I however believe that its impact could be improved by including some additional information as mentioned below. Alternatively, the suggestions in general comments should be addressed in future publications.

The article focuses on comparing the ATONA to current 10E11, 10E12, 10E13 and a hypothetical 10E14 ohm amplifier, or rather their idealized Johnson-Nyquist noise characteristics, for the purpose of multicollector noble gas measurements. ATONA outperforms ideal i.e. model 10E13 ohm amplifiers with respect to signal-noise ratio for 10 second integrations which is (most likely) an appropriate integration time for many measurements, and approaches an ideal (and currently commercially unavailable) 10E14 ohm amplifier for a 100 second integration which is most likely to long to properly sample and back-project a noble gas ion beam evolution to T0. The high dynamic range and low noise-fast response is definitely an improvement as compared to traditional amplifiers. This versatility means that amplifiers do not need not be physically or electronically switched among Faraday cups for different applications, which is an additional advantage that complements their low-noise characteristics. An ATONA could also be useful for single detector instruments that still have merit due to the high sensitivities afforded by the small volumes of such instruments.

Although the comparison with traditional amplifiers at low signal intensities is appropriate, the paper could benefit from a more stringent comparison with ion counters where the noise characteristics at low signal intensities are dominated by Poisson i.e. counting noise of the individual ion arrivals. This noise is inherent to counting atoms or ions and cannot be avoided. An interesting question is therefore under which beam intensity x time i.e. accumulated charge conditions the "baseline" noise in an ATONA becomes comparable to this inherent and unavoidable counting noise that will also be present and superimposed on zero-beam i.e. electronic baseline noise? This would seems to be an appropriate lower dynamic range where ion counters would (decisively?) outperform ATONA in terms of precision (but not necessarily accuracy). This number could presumably be calculated based on the 1-10-100 second zero-beam measurements that have already been carried out. It may also be possible to tease out that information from e.g. figure 7, but it is better that it is presented.

> It is true that the ATONA cannot replace ion counters for measuring very small signals. The baseline noise of the ATONA, while greatly reduced compared to other Faraday

amplifiers, is still larger than the near-counting-statistics noise level of an ion-counting multiplier. We show this, for example, in Figure 7, and we have added new discussion to the introduction clarifying the circumstances under which an ion-counting multiplier remains preferable. We also discuss in Section 3.2 that the ion counter remains necessary for very small signals on the NGX. We have also added a shot noise calculation to Figure 7.

The paper would also benefit if the working principles of ATONA were more thoroughly discussed (without disclosing confidential information). The patent documents contain a lot of public information that could be condensed into a description of the technology. I think the mass spectrometry community would be more likely to adopt the technology if they could understand it better, rather than using it as a "black box" technology where one might run into an unpredictable problem. As a naive non-engineer I personally would like to know how leakage current is reduced. Is there a maximum charge than can be accumulated before "discharging" if that is even the appropriate term? Are there hysteresis effects in the capacitor that make it particularly hard to drive out or sense low buildups of charge that might adversely affect linearity at low signal intensities? Can charge buildup in the Faraday-amplifier system start to deflect incoming ions, changing the peak shape thereby affecting e.g. pseudo-resolving peak-shoulder measurements. Does the "firmware" make decisions on sampling rate or readout parameters, switching between different regimes that depend on beam intensities?

We have endeavored to address these issues in the manuscript, and we address the specific points raised here in more detail in the response below.

Regarding leakage current: We assume that the reviewer is referring to the leakage current through the capacitor when there is non-zero charge accumulated. This leakage current is caused by migration of electric charges through the volume of the dielectric when an electric field is applied and creates non-linearity in measuring the accumulated charge over time, as part of the charge is lost through leakage during the measurement. Isotopx addressed this by first, the use of proprietary extremely low leakage dielectric for the feedback capacitor, then cooling the amplifier to reduce the already very small leakage current and then measuring its parameters to further compensate for the leakage. As a result, this error current is less than 1aA (10^{-18}A) for input currents above 1pA (10^{-12}A) creating <1ppm non-linearity. For smaller input currents the error current is reduces proportionally, 10^{-19}A for 100fA (10^{-13}A), 10^{-20}A for 10fA (10^{-14}A) and so forth, still maintaining <1ppm non-linearity. We have added this information to the manuscript.

Maximum charge: There is a maximum charge that can be accumulated by the feedback capacitor, which is determined by the value of the capacitor and the working voltage of the amplifier. However, the ATONA simply discharged the capacitor when it reaches the maximum value, a scenario that does not affect the measurement process. Only the rate of change of the transimpedance amplifier output voltage and therefore the rate of change of the accumulated charge is measured. This rate of change does not depend on the maximum charge value and the maximum value of the measured current depends only on the dynamic properties of the amplifier and subsequent data acquisition circuitry.

Hysteresis: Dielectric hysteresis may be defined as an effect in a dielectric material similar to the hysteresis found in a magnetic material. This causes a static shift in the capacitor voltage for a certain charge dependent on the history of previous charges/discharges. Isotopx addressed this by the use of proprietary dielectric with paraelectric properties and with negligible hysteresis. As a result, the effect of capacitance-voltage hysteresis on output voltage is unobservable.

Charge buildup: The Faraday buckets are directly connected to the input of the inverting amplifier. This fixes the voltage of the bucket at zero volts all the time regardless of the accumulated charge and therefore does not create any change or deflection in the incoming ions beams. We have added this information to the manuscript.

Firmware/black box decisions: No. The firmware neither changes any acquisition parameters, e.g. sampling rates, voltage ranges, measurement regimes or any other parameter, nor switches/changes any hardware values or components for any reason. This is done to preserve continuity, linearity, and repeatability of the measurements throughout the entire dynamic range.

Throughout: The term Johnson-Nyquist noise is used in line 114, but then subsequent usage is about Johnson noise. Should abbreviate it JN-noise at first usage, and then refer to it as such subsequently.

We have adjusted the text as suggested for better clarity.

When discussing the performance using air and cocktail standards, it would be nice to have the approximate beam intensities tabulated in e.g. fA as that it the unit that is already reported for noise measurements.

We have added this information where appropriate.

Specific: First paragraph i.e. 8-22 could perhaps use a statement regarding engineering tradeoffs regarding multicollection versus volume/sensitivity, i.e. the increase in volume that tends to occur with multicollection and the related drop in sensitivity. This is one reason why single collector instruments still have a role. In fact, the versatility of the ATONA seems to make it very well suited for that role; this is only aided by its rapid response as discussed later.

We agree with both the principle of the statement regarding the value of small volume instruments and with the possible role of the ATONA in such instruments, both because of its dynamic range and because of its response time. We have added statements to this effect to the second paragraph of the manuscript.

Second paragraph, line 30. Mention of long settling time for high value resistors is relevant in case of dynamic measurements, but static multi-collection of noble gases all but removes the settling time issue since any single resistor only measures one very slowly evolving beam. This should be mentioned in order to be fair to the current generation of high-ohm multi-collector equipped instruments.

While it is true that settling time is less important for multicollector instruments, it can be long enough on some high-gain RTIAs that without mitigation it affects the settling time of the measurement on the time scale of gas inlet. We have added this caveat to the manuscript.

Paragraph 6, Line 80. Could the authors perhaps make a back of the envelope error propagation calculation of how much of the air correction error on a blank subtraction on their instrument would arise from the 36Ar using a ion counter versus the ATONA? Or conversely the calculations suggested in the general comments regarding comparison of counting noise vs zero beam noise? This would be highly relevant for e.g. Ar or Ne dating of young samples where samples or fractions may be comparable in intensity to blanks.

We have added such a calculation for a typical young basalt sample.

Paragraph 7, Line 84. If possible, it would be nice if the patent were hyperlinked.

We have provided a hyperlink.

Paragraph 9 A formulation of Johnson-Nyquist noise with some appropriate reference and description would be useful for non-specialists.

We have added additional information.

Paragraph 11, Lines 130-140. This is a bit hard to read, and the reporting would benefit from a data-table showing the noise characteristics for 1, 10 and 100 second integrations with ATONA and 10E11-14 resistors. In such a way, one could focus on describing the noise "crossover" points for the various detector technologies that most readers would be searching for anyway as seen in figure 4.

We have added the requested table as Table 1.

Figure 6 (and figure A1) It is hard to identify the ranges, could the color code somehow be complemented by a change in marker style? It might also be a good idea to write the ranges as from 200% to 0.36% rather than between 200% and 0.36%.

In response to this comment and a comment from Kuiper, we have reorganized the figures so that the analyses are grouped by signal size rather than by analysis order, which we hope will also address the difficulty in distinguishing them from one another. The point about the ranges being inclusive is noted and this change has also been made.

Figure 7 We should expect a number of inflection points where all faraday mass spectrometer technologies gradually switch to follow a slope determined by counting noise (N^0.5) rather than signal over "baseline" JN or kTC noise (N^1). The linear error envelopes could give the erroneous impression that Faraday-based technologies can eventually outperform counting noise at high intensities, this should be avoided.

We have added a line showing the calculated uncertainty limit for a time-zero regression through data affected only by shot noise.

Table 1 The table should include the intensity of the smallest ion beam intensity i.e. the 36Ar intensity in fA. It would also be nice to have (and discuss) an MSWD to compare internal *precision and external reproducibility for all the measurements. A calculated average for the different intensities would also be nice, and could be plotted to evaluate non-linearity.*

The uncertainties shown at the bottom of the table are in fact the population uncertainties. We have added a line also showing the averages as requested, which should also make it more clear that what is meant by the 1-sigma uncertainties at the bottom (this is also in response to a comment by Kuiper). The averages are all well within uncertainty of one another, but an experiment aimed at properly assessing isotope ratio linearity would require many more measurements for the smaller signal sizes.

Table A1 The table should include the intensity of the smallest ion beam intensity i.e. the 38Ar intensity in fA. It would also be nice to have (and discuss) an MSWD to compare internal precision and external reproducibility for all the measurements. A calculated average for the different intensities would also be nice, and could be plotted to evaluate non-linearity.

Data for 0.36% measurements seem improbably precise, are they missing a digit

See notes from above. And yes, thank you for catching that—the table was hanging off the edge of the page, truncating the internal uncertainties for these measurements. The tables have been modified to fit the page better.

It would also be nice to discuss the presumably significant decrease in precision when going from 5.2% aliquots to 2.6% aliquots and lower. Is this a characteristic of ATONA, or is it due to error propagation effects from subtraction of blank 38Ar + H37Cl?

While the uncertainties increase for smaller signal sizes, the relationship is as expected and is primarily governed by the measurement uncertainty of the smaller ion beam. We discuss this further in Section 3.2 and show it in Figure 7.