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TÜBİTAK 1MV Accelerator Mass Spectrometer Designed for ^{14}C, ^{10}Be, ^{26}Al, ^{41}Ca, ^{129}I

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ABSTRACT

A 1 MV Accelerator Mass Spectrometer (AMS) was established at TÜBİTAK, MRC Türkiye in December 2015. The 1MV TÜBİTAK AMS system is based on a Pelletron tandem accelerator, operating up to 1.1MV. Built by National Electrostatics Corporation (NEC), its design was unique at time of production. The TÜBİTAK 1MV AMS is based on the design of an XCAMS 0.5MV AMS also produced by NEC, but with a higher energy tandem accelerator. The higher energy accelerator features better precision (1% or less) for ^{10}Be measurements. This new AMS also has larger magnets than the XCAMS. Larger magnets make it possible to measure ^{41}Ca and ^{129}I at the two-anode gas-ionization detector. In this article, the technical features; the parameters of ion source, terminal and other operations, and measurement quality are explained for accomplished performance of the five isotopic ratios, $^{14}C/^{12}C$, $^{10}Be/^{9}Be$, $^{26}Al/^{27}Al$, $^{41}Ca/^{40}Ca$, $^{129}I/^{127}I$. This article also presents five years of data for $^{14}C/^{12}C$ ratio measurement quality control are represented as well.

1. Introduction

The first AMS facility in Türkiye was established in 2016 at The Scientific and Technological Research Council of Türkiye, (TÜBİTAK), Marmara Research Center, (MRC). The laboratory is called (Türkiye) National 1 MV Accelerated Mass Spectroscopy Laboratory (Doğan et al., 2021). Since 2016, the laboratory has been providing international commercial radiocarbon analysis making it one of only two AMS laboratories in the Middle East and the Balkans. Considering the high demand for radionuclide analysis in the fields of archeology, earth science, environmental science, and criminology in Türkiye and the region, Türkiye‘s first and only AMS laboratory is aiming to address both domestic and international requests. The establishment of the laboratory which includes the 1MV AMS system is financed with national resources by the Ministry of Development, Republic of Türkiye.

The TÜBİTAK 1MV AMS system is designed and produced by National Electrostatic Corporation (NEC) using their Pelletron tandem accelerator technology. The Universal AMS system is the first 1MV AMS from NEC designed for multi species measurements. In this paper, the unique design of the UAMS system, the 1MV Accelerator Mass Spectrometer capable of detecting ^{14}C, ^{10}Be, ^{26}Al, ^{40}Ca as well as ^{129}I, is explained. An overview of the system’s components is also featured. The technical features: The parameters of ion source, terminal and other operations, and measurement quality are explained for accomplished performance of the five isotopic ratios, $^{14}C/^{12}C$, $^{10}Be/^{9}Be$, $^{26}Al/^{27}Al$, $^{41}Ca/^{40}Ca$, $^{129}I/^{127}I$. The five years

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of data for $^{14}C/^{12}C$ ratio measurement quality control are represented as well.

1.1. AMS System Room

The AMS System room is a crucial aspect for providing smooth operation of AMS. If the room where the AMS system is installed is not adequate, it may directly affect the quality of measurement. The AMS system room was thoroughly prepared for installation before the AMS was dispatched for installation: The room floor was checked to be less than ±1 cm slope and ground concrete was prepared to handle the heavy AMS weight. We did not observe any subsidence of the floor in the past 5 years. Any slope forming on the ground would affect the alignment of the AMS. Both the AMS system and the cooling water chiller unit have a stable dedicated AC power setup including a diesel generator and 160KVA UPS which feeds electricity with minimum noise. High electrical noise may lead to defects on AMS parts or shorten their life. Other essential things needed to operate stable AMS are clean compressed air, filtered cooling water, controlled air temperatures, and a low dew point of the room. Fluctuations in humidity and temperature of the room can cause unstable AMS measurements. The TÜBİTAK AMS room is equipped to provide a stable room temperature around 20±1 degrees Celsius and a dew point less than 15. These parameters are continuously monitored at 8 points around the room. After five years of operations, all the parameters mentioned above are strictly maintained.

The room is also large enough to accommodate and service the TÜBİTAK 1MV AMS system which has an outline of 7.4 m x 4 m. (Figure 1)

2. Development of AMS Systems and the TÜBİTAK 1MV AMS System

AMS systems have gone under significant transformations in the last four decades. After showing that rare isotopes can be measured with AMS, some existing accelerator systems were modified, and measurements were made. Measurements were mostly focused on radiocarbon, which was in high demand. During the 1980's, high energy accelerators were used with the capability to measure more than one isotope (Synal and Wacker, 2010). Following this initial phase, lower energy models with an accelerator 1MV or less dedicated for radiocarbon

Figure 1- The AMS system in TÜBİTAK AMS laboratory room.

measurement were introduced (Synal et al., 2000). Commercial availability of AMS systems became more widespread. Now, there are carbon dedicated 0.2 MV AMS systems (Schroeder et al., 2004; Synal et al., 2007). Furthermore, AMS systems with low energy accelerators proved to be sufficient for multi-isotope systems including ^{10}Be, ^{14}C, ^{26}Al and ^{41}Ca (Klein et al., 2006; Zondervan et al., 2015; Macková et al., 2021). Recently, systems with low energy accelerators have become more preferred for routine measurements. Certain minor additions to AMS systems enabled development of techniques that allow measurement by eliminating isobars at lower energies. An important example is the introduction of Si_3N_4 energy degrader foil to measure rare Beryllium isotopes as low as 0.5 MV terminal voltages (Müller et al., 2008). Therefore, Türkiye's first AMS system is conceptualized and designed on this model to be able to measure multiple isotopes and is intended to be further developed in future. The TÜBİTAK 1MV AMS system model is a NEC 3SDH-1 UAMS. The UAMS is a more advanced system based on the XCAMS design which in turn was based on NEC's radiocarbon dedicated compact AMS systems (CAMS) (Southon et al., 2004).

When comparing the TÜBİTAK 1MV AMS hardware features with XCAMS, the first noticeable ones are the 1MV accelerator and larger magnets. Apart from radiocarbon, the TÜBİTAK 1MV AMS can measure rare beryllium, aluminum, and calcium isotopes as well as iodine 129. The 1MV accelerator provides a significant improvement in the precision for Beryllium measurements compared to XCAMS (Zondervan et al., 2015). Also, the larger magnets allow measurements of calcium and iodine isotopes. In fact, the TÜBİTAK 1MV AMS system was ordered to measure only the species mentioned above. However, during production, we requested to add iodine measurement capability to the system. The TÜBİTAK 1MV AMS can be modified to measure more species as future technological developments in isobar separation filters emerge. For instance, AMS systems with 5 MV or higher terminal voltage can measure ^{36}Cl, however recent attempts to extend efficient isobar separation is promising it to be measured in lower energies (Martschini et al., 2011).

The layout of the 1MV AMS system and the details of the components are given in Figure 2.

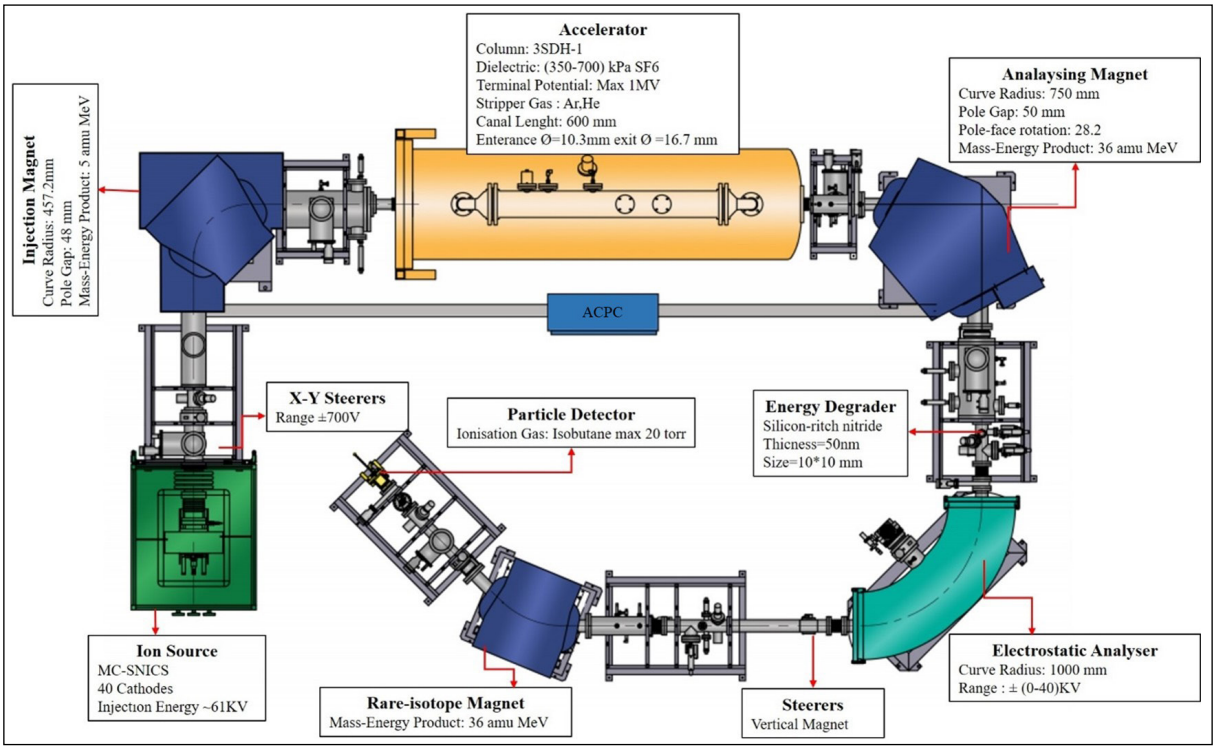

Figure 2- The layout of the 1MV AMS system and the details of the components.

2.1. Ion source

The TÜBİTAK 1MV AMS Multi-Cathode Negative Ion Source (MC-SNICS) holds up to 40 solid samples. The negative ions are accelerated through a potential of 61 kV. The ion source produces up to 100 microampere currents measured in the first Faraday cup, located just after the ion source. Ion source parameters for each isotope are given in Table 1.

2.2. Injection Line Section

An X/Y steerer is located after the ion source, and just before the first Faraday cup. These isotope specific steerers are used to tune the beam along the injection line section with the help of the first beam profile monitor (BPM), which displays the shape of the beam in the X and Y direction. This is the first BPM in the UAMS system and is placed following the Faraday cup before the insulating gap. The insulating gaps are at the image and object point of the magnet. The magnet bias supply (MBS) coupled to the 90° bending magnet does the sequential injection. In the case of Carbon analysis, abundant ^{12}C and ^{13}C ions are integrated at the Faraday cups every 1800 ms and 10000 ms respectively, whereas the rare ^{14}C ions are injected into the Gas Ionization Detector for 90000µs (Doğan et al., 2021). This cycle continues with a repetition rate of roughly 10 Hz. The injection times for each ion species are given in Table 2.

After the magnet, the beam of one ion is injected at once through the Pelletron accelerator. Between the magnet and the mass accelerator are offset Faraday cups to measure the abundant isotope(s). The injection magnet, or low energy magnet, has a radius of 457mm. After the magnet there are two off axis Faraday cups. The first one is used for ^{12}C, ^{13}C, ^{9}Be, ^{40}Ca, and ^{127}I while the other one is used for ^{27}Al. Slits are placed just before the entrance to the accelerator tank.

The sequential injection brings advantages to the AMS system. The average current from the abundant isotopes in the 3SDH accelerator remains small. This allows the use of substantial abundant isotope currents, maximizing throughput and measurement precision, but at the same time minimizes accelerator loading.

2.3. 1MV Accelerator

The one charge state ion's beam is accelerated to a maximum of 2.2 MeV to achieve the necessary energy dispersion through the high energy part of the mass spectrometer. An Einzel lens is placed in the acceleration tank just before the acceleration tubes. Interlocks are incorporated for safe operations. A chain stop was also installed in this system (Figure 3). It gives an opportunity to stop the chain motor if the chain stretches too much. Excess and unexpected stretch of the chain can cause damage to mechanical parts in the tank. 50 psi of SF_6 gas is sufficient for accelerator operation at 0.5MV. However, 80 PSI is required for operation at full gradient at 1.1 MV terminal voltage. The SF_6 gas is circulated in the tank so that it is continuously filtered to keep the gas as pure as possible and cooled with a heat exchanger.

The molecular dissociation is achieved by the accelerator via the stripper gas that can be selected among Helium and Argon from the two bottles placed inside the high voltage terminal. The stripper gas

Table 1- Ion source parameter for each isotope is given.

Isotope Ratio	$^{10}Be/^{9}Be$	$^{14}C/^{12}C$	$^{26}Al/^{27}Al$	$^{41}Ca/^{40}Ca$	$^{129}I/^{127}I$
Cathode	6.00 kV	6.00 kV	6.00 kV	6.00 kV	4.00 kV
Immersion Lens	5.65 kV	5.60 kV	5.60 kV	5.77 kV	3.33 kV
Extractor	15.00 kV	15.00 kV	15.00 kV	15.00 kV	18.00 kV
Focus	0.96 kV	0.8 kV	0.8 kV	0.00 kV	0.96 kV
Bias	41 kV	41 kV	52 kV	42 kV	37 kV
Ionizer	23 A	23 A	22 A	23 A	23 A

Table 2- The injection times for each ion species are given.

Species Measured	^{14}C	^{13}C	^{12}C	^{10}Be as (BeO^-)	^{9}Be as (BeO^-)	^{26}Al	^{27}Al	^{41}Ca as (CaF_3^-)	^{40}Ca as (CaF_3^-)	^{129}I	^{127}I
Injection time (millisecond)	90	10	1.8	1	99	1	99	100	1	1	99

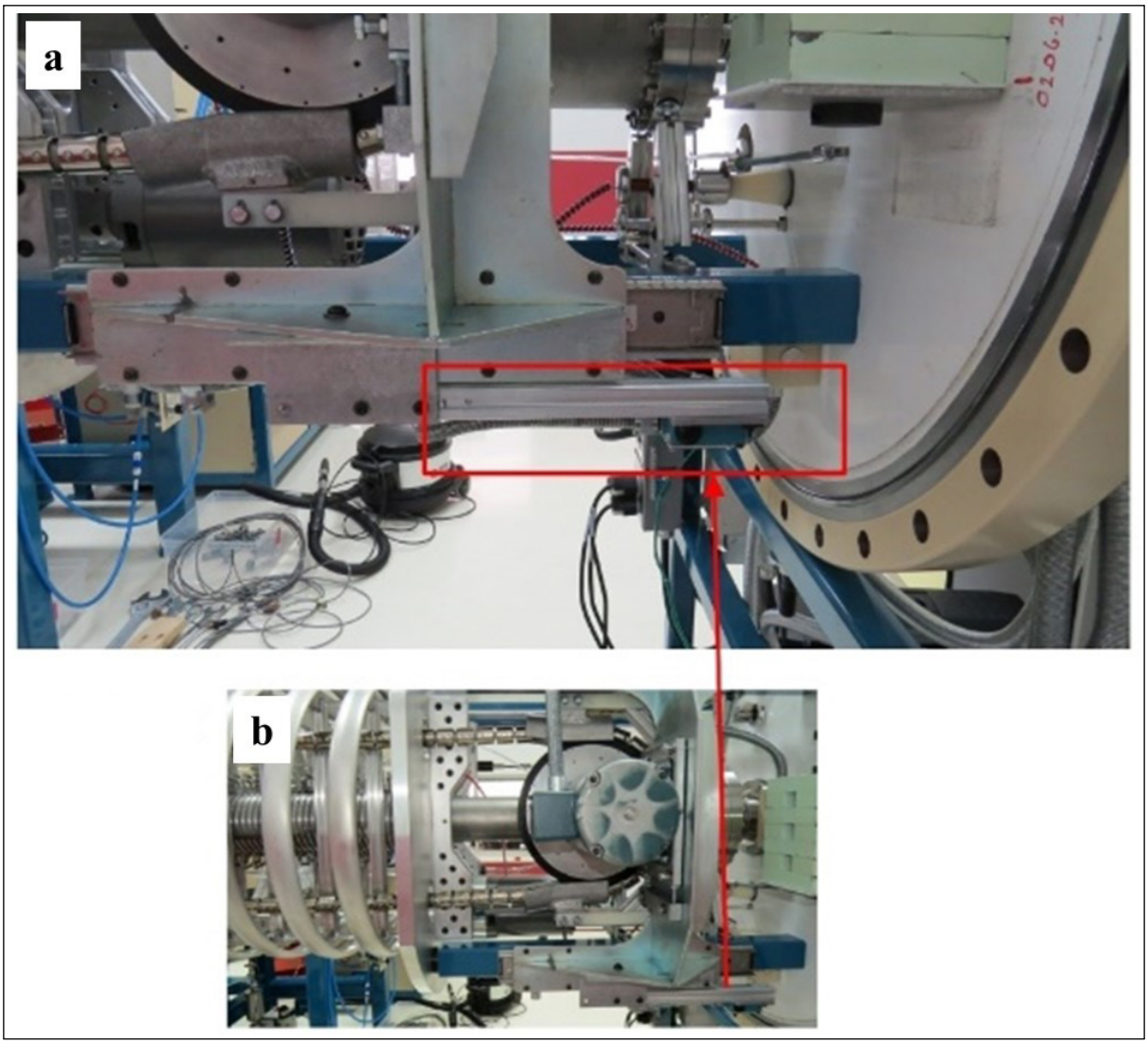

Figure 3- a) A chain stopper mechanism is installed to prevent damage in accelerator tank in case of excessive chain stretching and b) the position of the chain stopper mechanism in accelerator tank.

supply is configured to manually switch between Helium and Argon with a selector knob placed outside the tank. The stripper pressure is remotely adjusted with a metering valve in the high voltage terminal that is connected to a stepper motor on the tank head through a long lucite rod. Argon is used during carbon and aluminum measurements while Helium is chosen for Beryllium, Calcium, and Iodine (Doğan et al., 2021). The Stripper canal is differentially pumped and recirculates argon or helium with the two 300 l/min turbo pumps.

On each side of the terminal there are two 22 gap acceleration tubes. Voltage grading is achieved by 550 MOhm resistors across each gap.

The terminal stability is better than 0.05% of the terminal potential which indicates less than a 500V variation of the terminal voltage at 1 MV. Lost charge is stable below 0.4 micro ampere. Two VFDs were placed outside the accelerator tank to control the charging chain and generator motor. A glass viewport was placed at the top of the tank to monitor any stretching of the chain or sparking from the outside.

Focus adjustments are possible by changing the electric field gradient inside the accelerator with a shorting rod. With this the 3SDH Pelletron Accelerator can be operated at terminal potentials from 0.2 MV to 1.1 MV. The optimum terminal energy at the terminal

and the parameters specific to isotopes being measured are given in Table 3.

2.4. Analyzing Magnet

A Faraday cup is placed after the exit of the accelerator which is utilized during beam tuning. Due to design limitations, this beam line cup does not have a suppressor. This feature is different from other Faraday cups in the system. There is also an electrostatic quadrupole triplet before the analyzing magnet which is used for beam shaping and focusing.

The analyzing magnet has a 75cm radius, 50mm pole gap and ME/Z^2=36amu-MeV. This allows bending higher masses than Carbon and Beryllium. Thus, heavy isotopes such as ^{41}Ca and ^{129}I can reach the gas ionization detector.

After acceleration, the analyzing magnet separates the isotopes of interest, sending the abundant reference isotopes to the off-axis Faraday cups at the image point of the analyzing magnet. Furthermore, the rare isotopes travel down the rare isotope beamline. Fast, precision, gated current integrators with digital sampling circuitry measure the abundant isotopes currents.

There are 3 off-axis Faraday cups where 12C, ^{13}C, ^{9}Be, ^{40}Ca, ^{27}Al are measured. For ^{10}Be measurements a 75 nm energy degrader Silicon Nitride foil is inserted via an actuator. Due to slightly different stopping powers, the ^{10}B isobar counts at the detector can be separated from the ^{10}Be counts.

For the abundant isotope ^{127}I, measurement is done in the low energy side Faraday cup. Since it does not have any isobar, it is possible to use measurement data from the low energy side.

2.5. Electrostatic Analyzer

The second BPM along the UAMS is placed before the ESA, which makes it possible to check the shape and orientation of the beam after the high energy magnet. The ESA has a 100 cm radius which provides a longer flight path compared to the XCAMS. The ESA elements are kept at around +40 kV and –40 kV respectively for carbon measurements. The electrostatic analyzer discriminates ions based on energy and prevents unwanted ions from reaching the final rare isotope detector. The third filter, a 45° magnet in combination with a thin stopper foil before the ESA, is necessary for separating the isobar ^{10}B from ^{10}Be. A magnetic steerer is placed after the ESA

Table 3- The optimum terminal energy and the parameters specific to each element are given.

Rare Isotope	**^{10}Be**	**^{14}C**	**^{26}Al**	**^{41}Ca**	**^{129}I**
Source Output (A) (Species)	1.034×10^{-06}	5.234×10^{-05}	4.143×10^{-07}	1.685×10^{-07}	4.581×10^{-06}
Terminal Voltage (MV)	1.10	0.46	0.57	1.10	0.50
Charge State	1	1	1	5	3
Transmission	56.300%	41.050%	21.190%	0.002%	Not measured
Blank Measured	9.88×10^{-16}	2.50×10^{-16}	2.12×10^{-15}	1.72×10^{-11}	8.68×10^{-15}
Blank Material	Blank	Phtalic Anhydride	Blank	Blank	Woodward
Standard Material	NIST	NIST SRM 4990C (Ox-II)	KN-4-1	Nishiizumi et al., 2000	n/a
Standard Ratio	2.6775×10^{-12}	1.4202×10^{-12}	6.5727×10^{-11}	9.291×10^{-09}	1.5578×10^{-12}
Standard Deviation (%)	0.535%	0.080%	0.226%	0.705%	0.573%
Rare Isotope (Injected)	^{10}Be+(BeO^-)	^{14}C+(C^-)	^{26}Al+(Al^-)	^{41}Ca+5(CaF_3^-)	^{129}I ($129I^-$)

which makes it possible to tune up the beam in the Y direction. The third BPM in the UAMS makes it possible to monitor and tune the beam after the ESA and steerer magnet. There is also a Quadrupole located just before the 45° Degree Magnet.

2.6. 45° Degree Magnet

A 45° double focusing magnet is crucial to measure $^{10}Be/^{9}Be$ ratios precisely. In addition to a 45° magnet before the detector, a thin, removable, stripper foil is placed before the ESA. Some species traveling through this foil lose one more electron resulting in double charged $^{10}Be^{2+}$ ions to have a slightly higher energy than the interfering $^{10}B^{2+}$ ions (Figure 4). Finally, it is possible to separate these isobars in the gas ionization detector.

2.7. Rare Isotope Detector

The final BPM is placed just before the detector and serves to monitor and tune the final abundant beam trajectory before the detector.

The TÜBİTAK 1MV UAMS system employs a compact GIC (gas ionization counter) with two plates to separate the rare isotopes from remaining interferences, which can be “gated” out from the counts.

The detector connects to preamplifiers which connect to NIM amplifiers for pulse shaping and amplification. The output is sent to a XIA Pixie-4 4 channel analog-digital converter to minimize background from interference ions. Timing signals from the jumping beam system are typically used to gate the output signal along with the coincidence signal. This eliminates the possibility of counting artifacts during the short periods when the abundant isotopes are being accelerated and measured.

2.8. Control Deck

Like all NEC AMS systems, the AccelNET control system PCs are running on Scientific Linux. A second computer on Scientific Linux is running the offline analyzing software ABC software for normalization and evaluation of the results.

3. System Performance

Operation parameters of TÜBİTAK 1MV AMS for routine Carbon measurement are given in Table 4. Systematic testing of all five isotopes was performed in site acceptance tests. All acceptance requirements for the five isotopes were met. Since the system will be used predominantly for carbon measurements several IAEA standards were measured and compared to their consensus value (Table 5). Plotting the measured pMC values over the IAEA consensus values has a 0.99587 agreement.

Radiocarbon measurements have routinely continued since the laboratory was established. The five years of data for $^{14}C/^{12}C$ ratio measurement quality control are represented in Figures 5, 6, 7, 8 and 9 which illustrates times series graph for standard reference materials SRM 4990C (Ox-II), IAEA-C7, IAEA-C8 as well as processed (Phthalic anhydride $C_8H_4O_3$, Sigma Aldrich 320064) and unprocessed blank (Alfa Aesar graphite powder natural, briquetting grade 100 mesh, 99,9995%). It is an attempt to measure standard reference materials with a less than 1 % uncertainty. Providing a low uncertainty level has been the most important parameter for the TÜBİTAK measurement condition. Chemistry of graphite

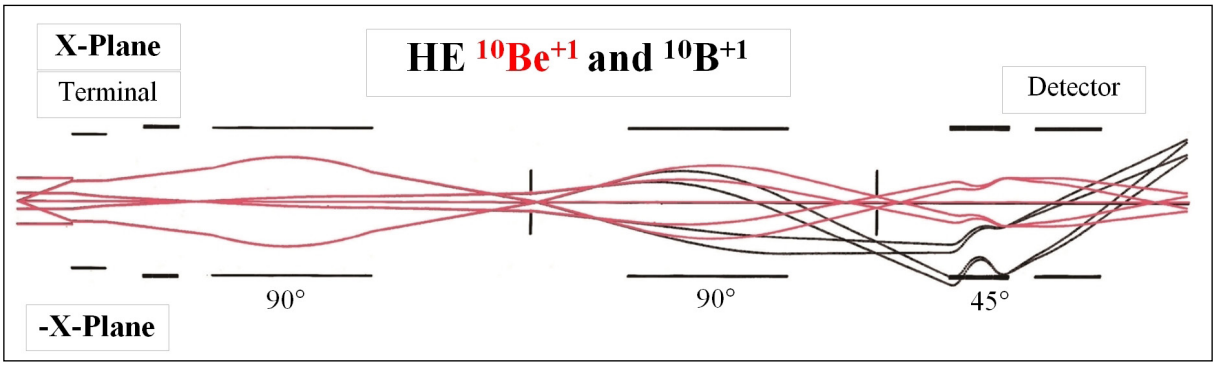

Figure 4- The ion beam profile in double focusing magnet.

Table 4- Operation parameters of TÜBİTAK 1MV AMS for routine Carbon measurement.

Section of AMS	Parameter	Value
Ion Source	Beam Current Energy of the Beam	50-60 µA 61 keV
Injection Magnet	Field MBS	0.29T 10.683 kV 5.090 kV 0.200 kV
Accelerator	Terminal Voltage Argon Stripper Gas Pressure	0.460 MV 0.06 mbar
Analyzing Magnet	Field	0.72T
Faraday Cups	^{12}C Beam ^{13}C Beam	2.149x10-05 A 2.341x10-07 A
Electrostatic Analyzer	Plate Voltage	39.78 kV, -39.78 kV
^{14}C Detector	OXII count rate	~ 10000 cpm

Table 5- Carbon Agreement with IAEA Standards are given for the preliminary measurements done during acceptance tests in 2015. *It is improved to closer to consensus value as 0.00-0.05pMC with eliminating contamination from graphite production in the last five years. **It is measured as 0.22±0.03 pMC when treated with acid base acid (ABA) before measurement (Doğan et al., 2023).

Standard Reference Material	Consensus Value (pMC)	Measured Value (pMC)
IAEA C1	0±0.02	0.136*±0.08
IAEA C2	41.14	41.61±0.22
IAEA C3	129.41	128.95±0.51
IAEA C4	0.20 - 0.44	0.30±0.07
IAEA C5	23.05	23.05±0.12
IAEA C7	49.53	49.51±0.17
IAEA C8	15.03	15.01±0.07
IAEA C9	0.12 - 0.21	0.39**±0.03

production process and keeping ion source clean have been the most important impacts in uncertainty values. After operating for a total of 15 days (360 hours), the ion source has to be cleaned. Ionizer, immersion lens, and other components in this part of the AMS must be disassembled in order to clean the ion source. Typically, it is cleaned with Kimwipe™ and pure water and ethanol. Depending on the amount of crust developed on the surface of the ionizer, a sandblaster can be used with an ionizer and an immersion lens.

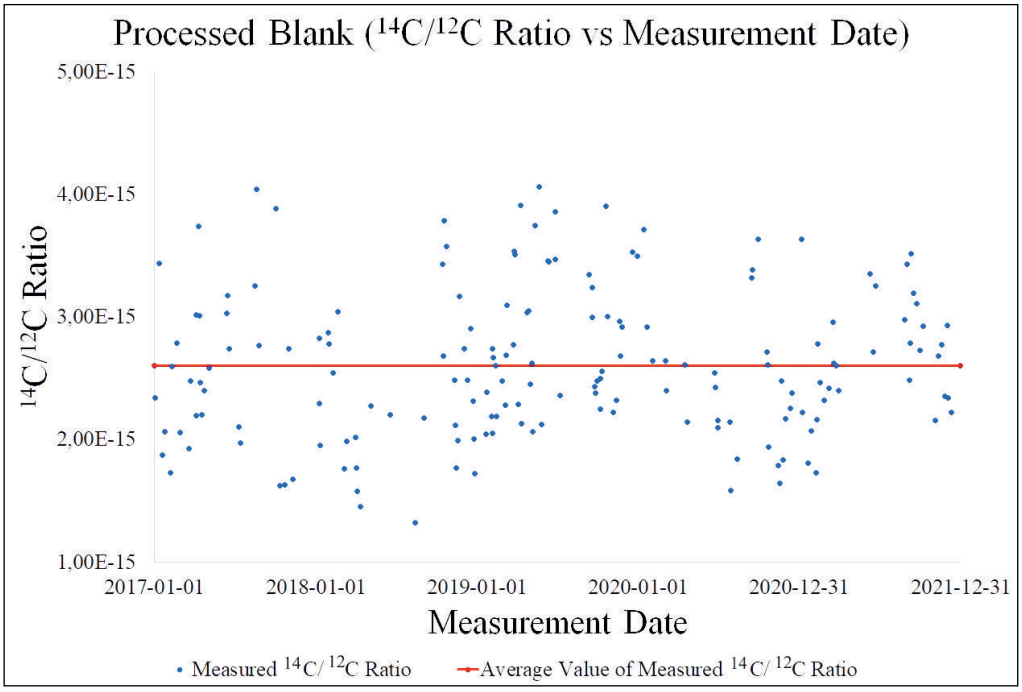

Figure 5- The five years data for ^{14}C/^{12}C ratio measurement quality control are represented for processed blank (Phthalic anhydride $C_8H_4O_3$, Sigma Aldrich 320064). The average value of ^{14}C/^{12}C ratio is measured to be 2.60x10^{-15}. The increase in this value is considered a contamination from the process in the chemistry laboratory.

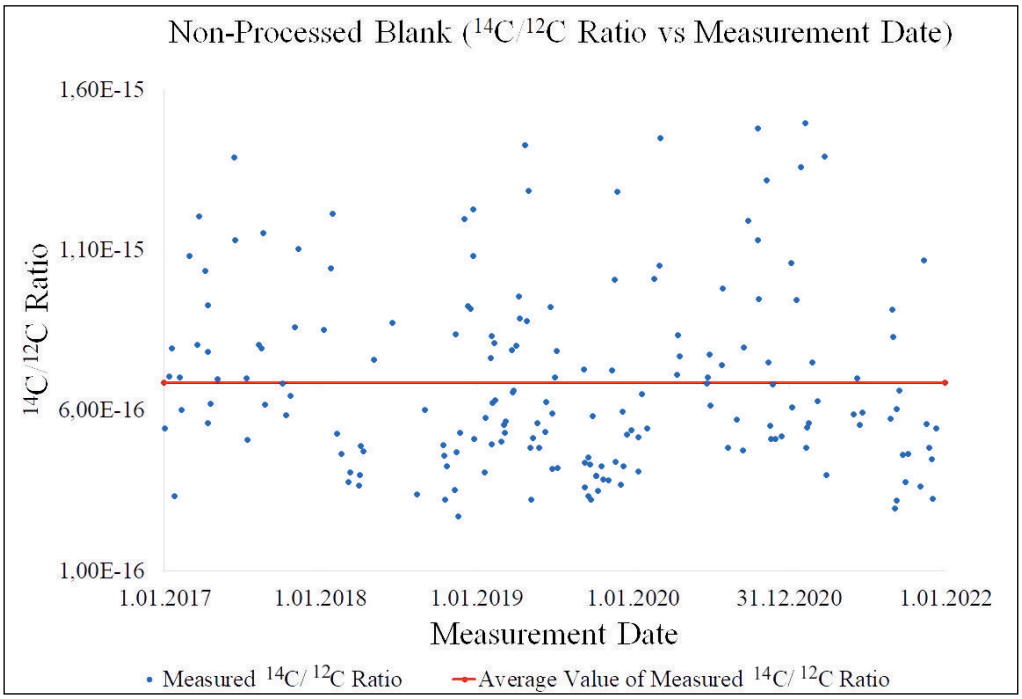

Figure 6- The five years data for $^{14}C/^{12}C$ ratio measurement quality control are unprocessed blank (Alfa Aesar graphite powder natural). The average value of $^{14}C/^{12}C$ ratio is measured to be $6.85x10^{-16}$. The increase in this value is considered rise in the background ^{14}C in the AMS usually because of contaminated ion source.

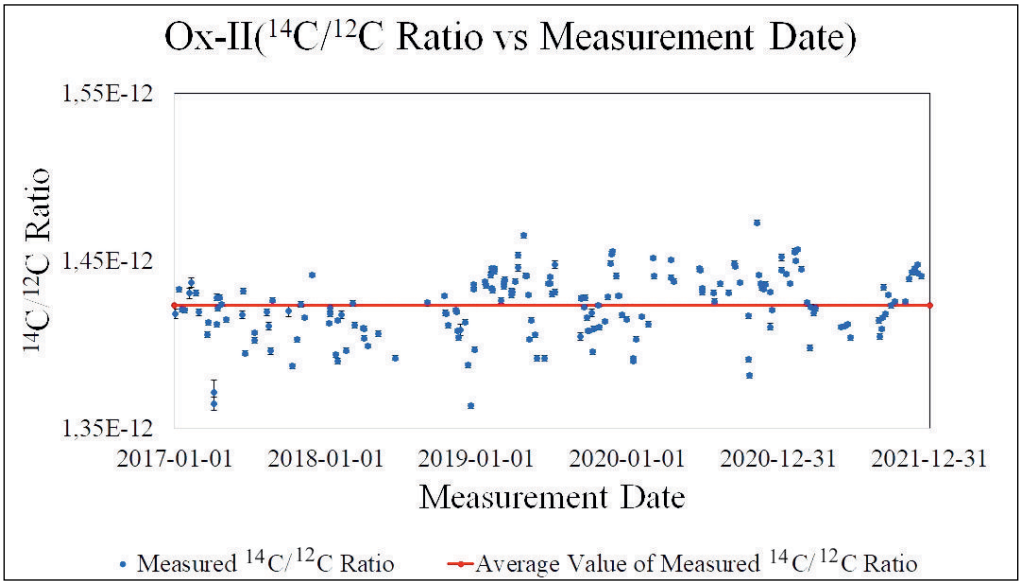

Figure 7- The five years data for $^{14}C/^{12}C$ ratio measurement quality control are represented for the standard reference material SRM 4990C (Ox-II),). The average value of $^{14}C/^{12}C$ ratio is measured to be $1.42x10^{-15}$. The cause change in this value is observed to be associated with replacement of ion source parts.

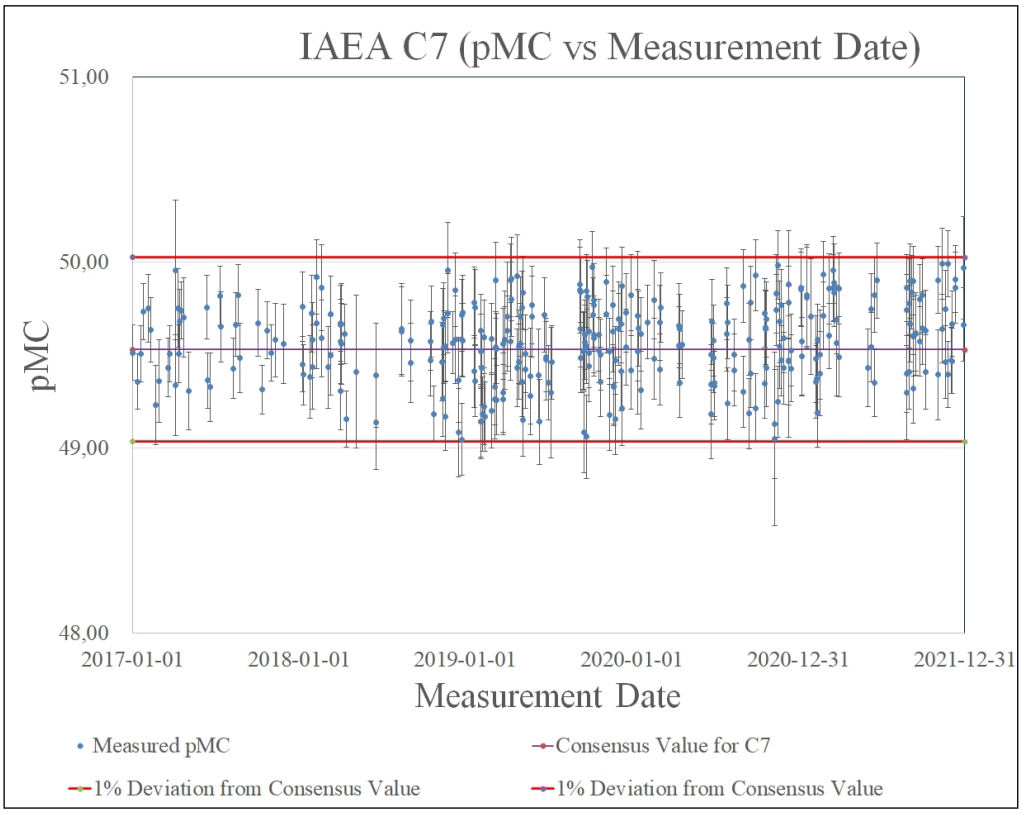

Figure 8- The five years data for $^{14}C/^{12}C$ ratio measurement quality control are represented for the standard reference material IAEA-C7. The average value of $^{14}C/^{12}C$ ratio is measured to be 49.57 pMC which is very close to consensus value 49.53 pMC. IAEA-C7 is measured to be maximum 1% deviation to be accepted; the overall measurement pass the quality control.

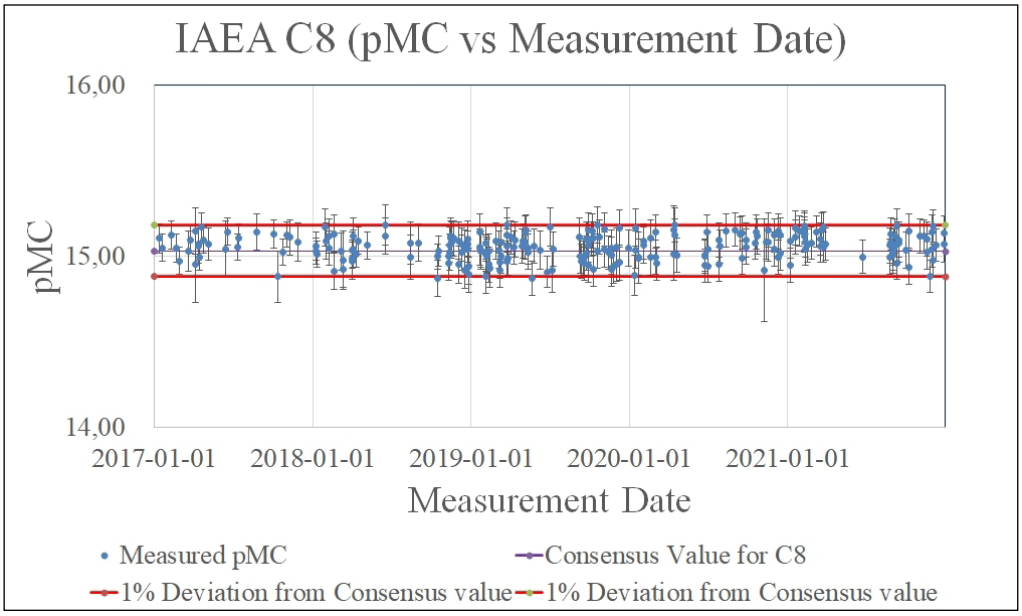

Figure 9- The five years data for $^{14}C/^{12}C$ ratio measurement quality control are represented for the standard reference material IAEA-C8. The average value of $^{14}C/^{12}C$ ratio is measured to be 15.05 pMC which is very close to consensus value 15.03 pMC. IAEA-C8 is measured to be the maximum 1% deviation to be accepted; the overall measurement passes the quality control.

4. Conclusion

A new AMS system based on a 1 MV tandem Pelletron was installed and accepted in December 2015. This UAMS was designed and built to measure five different isotopes. All standard and background isotopic ratios agreed with consensus values. After the system was validated and approved, regular operation procedures were implemented. The laboratory is now performing routine measurements on samples that have been brought in from across the world. The TÜBTAK AMS Laboratory is very significant for its work and support for scientific study in this region because it is one of the only three AMS laboratories in the entire Middle East and the Balkans (Doğan et al., 2023).

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