iPlasmaProQuad: A Computer System Based on a Relational DBMS for Processing and Monitoring the Results of Routine Analysis by the ICP-MS Method

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1 Introduction

Obtaining qualitatively new results in geochemistry, biology, medicine, and other disciplines is determined by how accurate and complete information the researcher has about the chemical composition of the studied object. Today Quadrupole Mass Spectrometry with Inductively Coupled Plasma (ICP-QMS) is the leading method for the determination of ultra-low concentrations of elements in various samples [1, 2]. Improving the accuracy of quadrupole inductively coupled plasma mass spectrometry depends on several steps in the analysis procedure, of which acid mineralization of samples, measurement of mass spectra, and processing mass spectrometric information are decisive.

Sample preparation is a critical step in ICP-QMS analysis that determines its success. Many approaches have been developed and used [3–5]. To improve the reproducibility of acid mineralization of many geological samples simultaneously, an ingeniously designed apparatus has been developed [6]. It allows the acid decomposition of geological samples of different classes (from mafic to ultramafic rocks).

Hardware in several designs (including acid distillation in a gas stream) is intended for simultaneous processing of up to 24 samples at full control of decomposition conditions. The developed hardware is based on the principle of maximum formalization of the description of operations that make up the decomposition procedure for samples of a different type. This principle improves the reproducibility of the decomposition procedure (and, under certain conditions, the accuracy of the analysis).

A significant point is that data processing is an analysis bottleneck that significantly reduces productivity. For example, measuring a batch of 20–30 samples takes less than one working day, while data processing can take two or three days, and sometimes more. It is due to the introduction of various kinds of corrections for matrix effects, spectral interference, spectrometer sensitivity drift over time, integration of results for individual isotopes, and many other factors. In many laboratories, spreadsheets are used for this purpose. The manipulation of spreadsheets requires a clear understanding of the algorithm of action and is usually a sequence of several (usually manual due to variable size) operations on data sets. This is where subjective errors can arise, due to the so-called human factor. In addition, neither the in-house mass spectrometer software nor the subsequent additional calculations in a spreadsheet make it possible to assess the accumulation of error in the calculations and evaluate the most critical stage of the mass spectrometric measurement and calculation of concentration. It is also difficult to document all input corrections, which breaks the chain of traceability of the formation of analysis results.

That is why developing a program for mass spectrometric information processing remains an important task. There are a few publications in the literature devoted to the development of software for such tasks. For example, the article by Tharaud et al. [7] presents the uFREASI software for the processing of elemental analysis data from ICP-MS spectrometers. The software provides concentration calculations, various data correction options, error accumulation evaluation, etc. The software is written in C++ and is freely distributable. Another freeware package is Iolite [8]. It is based on the Igor Pro package with its programming language (Wavemetrics Incorporated). The package includes advanced graphics and many useful options. The Iolite program works with primary spectra of any origin with time-resolved output. The program provides in particular laser ablation (LA ICP-MS) data processing and is aimed at advanced users who develop analysis methodology. The user-oriented package ADE [9] is positioned by the authors for routine ICP-MS analyses. The IsotopeMaker [10] builds on the well-known MATLAB package and is aimed

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primarily at isotopic analysis, including the LA ICP-MS version. Information is available on programs focused exclusively on LA ICP-MS data processing [11, 12].

2 Experimental Part

2.1 Equipment

An X Series II quadrupole inductively coupled plasma mass spectrometer (Thermo Scientific) equipped with a concentric atomizer and a quartz cyclone atomization chamber cooled by a Peltier element (2 °C) was used. Parameters of the measurement: generator output power 1400 W, plasma gas (Ar) flow rate 13 l/min, auxiliary 0.95 l/min, Ar flow rate through the nebulizer 0.87 l/min, sample flow rate 0.8 ml/min. The mass spectra were measured using two scanning modes: Survey Scan with 5 passes from 5 to 240 m/z and Peak Jumping with 1 channel per mass, integration time 20 ms, and 25 passes. All measurements were performed with PlasmaScreen software. If all device settings are observed, the level of oxide ions CeO^+/Ce^+ is not more than 2%, the level of double-charged ions (Ba^{2+}/Ba^+) is not over 3%.

2.2 Reagents

To prepare calibration solutions and for decomposition, we used the listed chemical substances: deionized water distilled without boiling in PTFE Subboiler ECO IR Maassen (Germany) water and acid treatment system; acids produced by Merck (Germany): HF (40% for analysis), HNO_3 (65% for analysis, max 0.005 ppm Hg), $HClO_4$ (70–72% for analysis), H_3BO_3 (for analysis), as well as HCl (GOST 3118-77), distilled (27%) without boiling in the PTFE Subboiler ECO IR system. ICP-MS-68 multi-element standard calibration solutions (Solution A and Solution B) produced by High purity standards (USA), single element solutions of REE with a concentration of each element 1000 µg/ml (Merk, Germany).

Working calibration solutions for ICP-MS were prepared by successive dilution of initial solutions of multi-element standard solutions ICP-MS-68 to concentrations of 10 µg/L for solution A and 5 and 10 µg/L for solution B. A multi-element standard solution for REE group with a concentration of each element of 10 µg/L was prepared by mixing solutions of individual REE standards. In all cases, 2% solution of HNO_3 was used as diluent. A solution of In (10 µg/L in 2% HNO3) was used as an internal standard. Solutions were diluted in 10 ml disposable polyethylene tubes (Litaplast-Med, Republic of Belarus) or 50 ml (Cititest

Labware Manufacturing Co., Ltd., China) before measurement. The gravimetric method was used to control the dilution degree.

3 Results and Discussion

To automate the processing of the mass spectra, a computer software iPlasmaProQuad has been developed. The mass spectrometer is used only as a detector for the mass spectra, providing the corresponding peak areas and the meta-information about the measurement conditions. All other necessary calculations are performed by the computer program itself. The architecture of the iPlasmaProQuad software is based on an entirely different concept compared to the programs mentioned in the Introduction. All calculation operations are based on the system of consecutive queries, the results of which can be seen immediately using MS Access. Query compilation takes place in an interactive environment, so knowledge of the SQL language is usually not required. Learning how to create queries in MS Access using the available constructor is not difficult. This means that users can easily modify and extend the functionality of the program. Another advantage of the developed architecture is that the management of a query sequence can be entrusted to an external program written in a high-level programming language, such as C#. This is exactly how iPlasmaProQuad is implemented, where the interface with the user is written in this language. The task of the interface shell is to request the name of a file containing the results of a batch of samples measured on a mass spectrometer, import the information into database tables, check the integrity of the received information (query calls) and then run a sequence of queries that perform the calculations.

The approach implemented relies, for the most part, on the experience of using relational databases to process information in neutron activation analysis [13, 14]. At that time, desktop DBMSs had relatively limited capabilities. Therefore, they were mainly tasked with storing structured information and generating various types of reports. The progress of the development of desktop database management systems and their convergence in functionality with industrial SQL systems allow today to solve significantly more complex tasks. The first version of the iPlasmaProQuad software was presented at the Euroanalysis 2019 conference [15].

The database currently includes over 50 tables and more than 110 queries. The tables store different data. The tables are prefixed, depending on their purpose, for ease of use:

c_—service user's control data; sample/calibration solution measurement data, batch processing algorithm control

parameters, batch data for each sample (mass, dilution, etc.). The parameters controlling the processing can be changed via the software interface;

h_—handbook data: isotope abundances, recommended contents of elements in standard samples (solutions and solids), statistical tables, relationships of different units, constants, etc.;

t_—temporary/imported data: temporary tables for importing mass spectra measurement results, checking their integrity;

x_—experimental data: copying of validated data from temporary import tables of measurement results and converting them to a standard format;

r_—results data: results of calibrations, calculation of isotope concentrations, isotope consolidation, results of introducing various corrections (matrix effect, calibration drift, interference, etc.);

aux_—auxiliary tables: auxiliary tables for intermediate calculations/data.

All working tables (t_, x_, r_, aux_) are initially present in the database (without records) for their inclusion in the data relational scheme and for correct query execution.

The main window is a system of 5 menu items, the functionality of which follows the name: File (select the file with the measurement data to be processed), Import (enter the measurement results of mass spectra into the database), ControlPanel (control panel for data processing, sets the control table data), Tools (set of functions controlling the database: clearing temporary tables, compacting the database, etc.). In addition, the main screen of the program has 5 tabs, which are accessed according to the current context of the program. The tabs are used to display various reports, manage the list of isotopes involved in the calculation of element concentrations and other tasks. The lower part of the window has a log-file output panel (indicating the status of the program at different stages of its operation). The windows look and what they do is described in the Operator's Manual (F1). Here are two examples. Figure 1 shows a fragment of a window with a list of measured batch samples received after data import into the database and a pop-up window for setting some parameters to control further data processing.

3.1 Data Integrity Check

Once the primary data from the mass spectrometer have been entered into the t_tables, they are supplemented with several reference data and checked for consistency using a query system. The result is a working x_tables, which does not change during processing to ensure that repeated processing cycles can be called with different parameters.

3.2 Codification of Samples

Codification is an important part of determining the correctness and unambiguity of possible mathematical operations in sample lot processing. A set of codification rules has been developed to identify the sample batch and the customer, parallel samples and parallel measurements of the same sample, the presence of an internal standard in the sample, the sample dilution factor, and other data to process them properly.

3.3 Calibration Procedure

It is common practice in ICP-MS analysis to measure in blocks: first, the calibration solutions are measured, then several samples are analysed (10–15, depending on sample type, variation in sensitivity, etc.), then calibration is performed again, and so on. The instrument's in-house software provides a calibration that opens a block of samples. The iPlasmaProQuad software is more powerful in this area: the concentration can be calculated by both opening and closing calibrations or by interpolating calibrations (between the preceding and closing calibrations, depending on the time of measurement) and by combined calibrations (for all blocks).

During the calibration process, an auxiliary table is first generated, containing the data for the standard solutions necessary to calculate the linear regression parameters for each isotope. The sums Σx^2, Σy^2, Σxy, and linear regression parameters for each isotope are calculated for each calibration block separately, including for the union of all blocks ($Y = b * x$). These parameters also include the values of standard deviations of the linear coefficient (b) and the calibration model (Y) to allow for the uncertainty introduced by the calibration in the concentration results. Two calibration tables are generated in which the concentrations of the elements are expressed in both ppb and nanomoles.

There are two models of calibration available to the user. One generally accepted mathematical model of calibration is based on the assumption of homoscedasticity (relative stability, homogeneity of the variance of the random error) of the observed Y values (model 1). This is a prerequisite for the application of the least squares method. Statistical analysis shows, however, that the variance of observed Y values is not constant but is approximately proportional to the square of the measurand. Then the logarithm of Y values will satisfy the homoscedasticity condition (model 2). In this

Fig. 1 Fragment of the main program window after entering data from the mass spectrometer and an example of the dialog box (framed) opened by calling the ControlPanel menu item
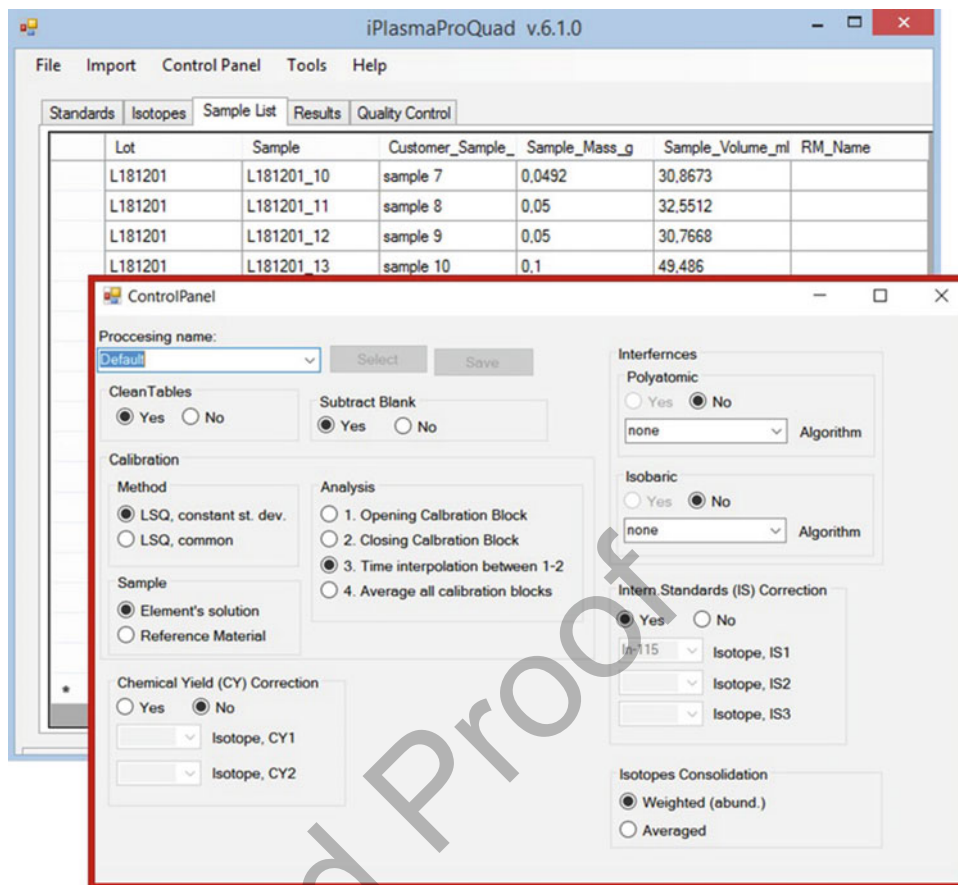
case, the error of the logarithm will correspond to the relative error of the measured quantity Y. By default, the program uses the latest calibration model (model 1 can be plugged in if desired). An important feature of the program is that it implements two branches using both homo- and heteroscedastic calibration curves to calculate the concentration.

3.4 Computation of Concentrations in a Series of Analyzing Solutions

Once the linear regression parameters $Y=b*x$ are calculated for each isotope for each calibration block, they can calculate concentrations in the series of the analyzed solutions. The iPlasmaProQuad software defaults to the sensitivity coefficient b obtained by linear time interpolation between the opening and closing calibration block. Error estimates (variance) are calculated from the combined calibration as relative standard deviation value (1) and absolute standard deviation value (2).

$$dC = Sqr([dY0]^2 + [db0]^2) \qquad (1)$$

where dY0 is the relative standard deviation of Y, db0 is the relative standard deviation of the coefficient b (when all calibration blocks are combined).

$$sC = dC * [Conc\_ppb] \qquad (2)$$

where [Conc_ppb] is the field containing the concentration of the isotope.

3.5 Correction for the Internal Standard

The concentration values calculated in the previous step can be refined by multiplying by a correction factor to account for changes in the sensitivity of the registration system. For this purpose, an internal standard (usually indium) of known concentration (e.g. 10 ppb) is added to the analysed solutions. The composition of the internal standard used is stored in table h_InternalStandards. Similarly, the standard deviation of the concentration of the internal standard is estimated, which is used to estimate the total uncertainty of the results when making sensitivity corrections to the registration system.

3.6 Accounting for Spectral Interferences

The problem of spectral interference in quadrupole mass spectrometry is solved either by using reaction (collision) cells or by a computation method. The latter method somehow relies on experimental results of determining the apparent concentrations of isotopes-analytes and mathematical methods for solving a system of equations.

The database includes a table of potential polyatomic interferences containing about 1 thousand records. The record contains the analysed isotope and the isotope fields composing the polyatomic components. The record includes a field with the apparent concentration of the analysed isotope. The value of the apparent concentration is determined during the analysis, based on the measurement results of various type samples. Among them are blank samples, solutions of standard samples, or solutions of model mixtures. When assessing the correction value for polyatomic interferences involving main elements (which are usually not determined by the ICP-MS method), it is necessary to have information about their concentration (for example, according to ICP-AES data or others).

3.7 Estimation of Detection Limits and Subtraction of Blank Test Results

The isotope concentrations are corrected for the content of analysed elements detected in the blank samples. For this purpose, the mean values and errors of blank samples for different dilutions are computed and entered in an auxiliary table by the correspondent queries. Then the software calculates the detection limits for each isotope for blank samples (DL_Lb) at different dilutions using the Student's t-distribution (table h_t_Student).

$$DL\_Lb = sC\_Lb * t\_95(n\_Lb), \quad (3)$$

where sC_Lb is the standard deviation of the element concentration (ppb) in the blank sample (Lb), t_95(n_Lb)- data from the Student's table.

Calculate the lower (Lci) and upper (Uci) limits of the confidence interval for the element concentration after background subtraction:

$$Lci = [C] - Sqr(([C\_corr\_In] \\ * (1 - 1/[Err\_Factor\_corr\_In]))^2 + [CI\_lb]^2) \quad (4)$$

$$Uci = [C] + Sqr(([C\_corr\_In] * ([Err\_Factor\_corr\_In] \\ - 1))^2 + [CI\_lb]^2) \quad (5)$$

where CI_Lb= CI_Lb = sC_Lb * t_95(n_Lb) / sqrt(n_Lb) the confidence interval for determining the mean value of the element concentration.

Next, the significance of the concentration found compared to the detection limit value is determined. At this stage, three logical fields are generated for the analyst to characterize the three levels of reliability of the found concentration values of the isotope of the element being determined (Table 1).

This produces three qualitative attributes characterizing the analysis result: "confident" when criterion 3 is met; "acceptable" when criterion 2 is met, but criterion 3 is not; "semi-quantitative" when the analysis result is close to the detection limit, i.e., criterion 1 is met, but criterion 2 is not.

3.8 Determining Elemental Concentrations

The iPlasmaProQuad software has sufficient functionality related to the processing of analysis data by individual isotopes to determine element content. The user can select weight functions (isotope abundance, the uncertainty of analysis results, etc.) to calculate element concentrations in samples considering all dilutions and sample mass. It is also possible to exclude certain isotopes (e.g. in the case of spectral interference) when calculating the concentration of an element.

Furthermore, the software identifies parallel samples from the sample code and can consolidate the data in a statistically sound manner. This also applies to parallel measurements of the same sample, e.g. at different dilutions.

3.9 Estimating the Uncertainty of the Concentration Measurement

Ultimately, the software calculates the overall uncertainty of the element concentration measurement in the sample using the partial uncertainty values estimated at each step. They are sample (peak) measurement, calibration, the introduction of correction for changes in sensitivity of the measuring system over time, consideration of matrix effects, the introduction of correction by spectral interference (not yet fully implemented), determination of element concentration by isotope (for multi-isotopic elements), and some other factors.

3.10 Quality Control of the Analysis Results

A special feature of the software is the possibility to carry out quality control of the obtained data. The database contains information for over 30 geological reference materials

Table 1 Reliability criteria for the analysis results

Case	Field name	Status	Criterion
1	Over DL	"Yes"	(1) if $C > DL$, i.e., the element concentration found is above the detection limit
2	Sign	"Yes"	(2) if $Lci > 0$, i.e., the lower limit of the 95% confidence interval of the concentration is above zero
3	Sure	"Yes"	(3) if $Lci > DL$, i.e., confident determination

of different origins used in the lab. The user can extend this list by editing the data in the table h_RMs_SolidList.

If a reference material(s) has been included in the batch of samples analysed, the software automatically provides a report comparing the analysis results obtained with the certified values, including the confidence interval and the relative deviation from the certificate. The results are placed on the Quality Control tab.

3.11 Reporting and Quality Control of Analysis Results

iPlasmaProQuad software is arranged in such a way that initially, the database contains results *of all* (!) calculation variants. The formation of reports is determined by the customer depending on the tasks of the investigation, for which the analysis was carried out. This choice is determined by the parameters set in the control panel of the program. The results are accessible for viewing in a window of the program, tab Results. Simultaneously, the software creates an export file in MS Excel spreadsheet format.

A series of tabs representing similar data sets (Sample element) is placed in the file, which can be seen as a metaphor for a multidimensional array (Fig. 2).

The arrays contain the following data:

- results of the analysis (no data is available if the element is below the detection limit);
- the uncertainty of the measurement results, the detection limit for each element in each sample;
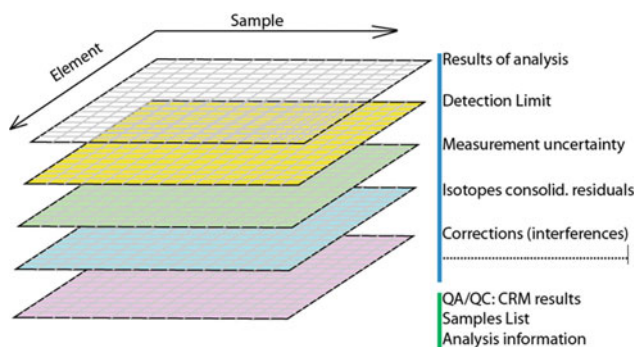
- three tabs containing logical information about the reliability of the measurement results according to the criteria given in Table 1;
- the standard deviation of the results of the analysis of the parallel samples (if they were presented in a batch);
- is the standard deviation of the results of parallel measurements of the same sample, including at different dilutions;
- quality control of the analysis (degree of deviation from certificate data) of the reference sample composition (if it/they were present in the sample lot);
- convergence of isotope analysis data when determining an element (for multi-isotopic elements);
- normalized concentrations (e.g. relative to chondrites, basalts-MORB) and several other data.

In this way, the array index (i, j) can operate with related data when performing intelligent data processing, constructing dependencies concerning the uncertainty of the analysis results when solving problems in the subject area.

Moreover, macros have been prepared in the template of the MS Excel file where the above information is uploaded. The macros can mark the degree of reliability of the data by highlighting them with the background color of the cell on the tab with the analysis results (red, yellow for cases 1 and 2 indicated in Table 1), as well as indicate by font type the results of the analysis of parallel samples (bold) and parallel measurements (italics).

3.12 Use of Concentration Measurement Uncertainty Data When Analysing a Single Sample

It is common practice for geological samples to be collected in the field, in the study area, with reference to the sampling location (coordinates, petrological/mineralogical characteristics, etc.). The laboratory then determines a geochemically representative batch of samples based on the results of the preliminary study, requiring detailed elemental analysis, including the determination of trace elements. At this stage, each sample is represented by a single sample, assuming that the analytical methodology used is well established and its metrological characteristics have been determined. Usually, one or more reference materials of a similar type are added

Fig. 2 Diagram of a multidimensional array of analysis results that are exported to MS Excel tabs

Fig. 3 Results of the analysis of the BIR-1 standard sample. Seven parallel samples, the elements are plotted in the coordinates mean concentration measurement uncertainty (%) and relative standard deviation of experimental results and certificate (%). The designation nDL (next to the element designation) indicates the excess of the concentration found over the detection limit (DL)
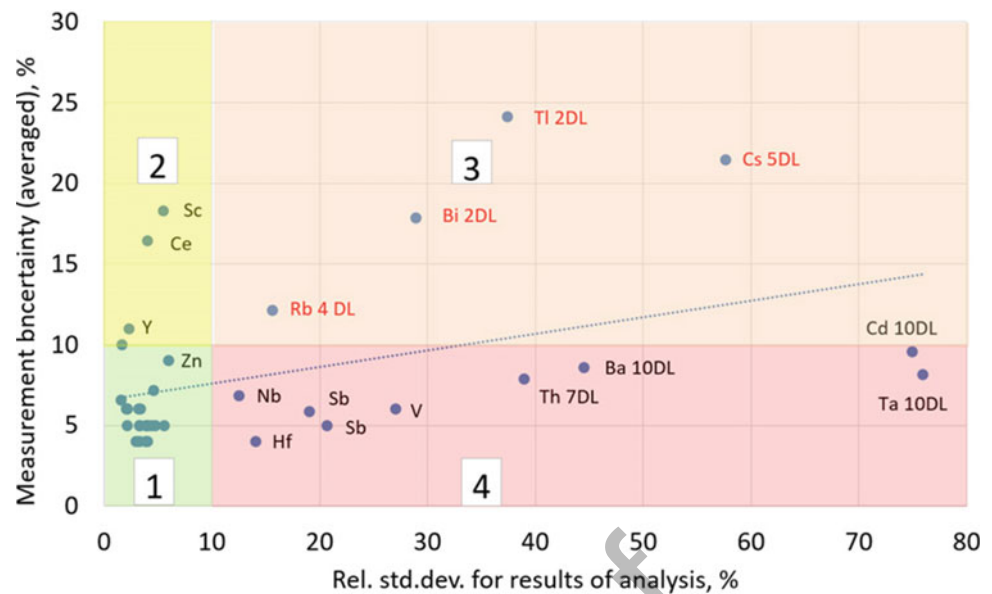
to the batch of samples to control the quality of sample preparation. If reliable and highly reproducible decomposition methods are relied upon, then the uncertainty values obtained in sample measurement can provide useful information in interpreting the data and reveal some problems.

A series of experiments were carried out to illustrate this point. In one of them, a set of seven parallel samples of a well-certified basalt reference material BIR-1 (USGS) was analysed. The decomposition methodology is presented in Ref. [5]. Using iPlasmaProQuad software, processing of samples, including estimation of uncertainty of concentration measurements and calculation of deviation of mean values of certain concentrations from the certificate, were performed. The results of comparison of the relative standard deviation of the analysis results from the certificate and the mean value of uncertainty of concentration measurement for all samples are presented in Fig.3. Assuming that the uncertainty of acceptable quality results should not exceed a threshold of 10%, several zones can then be distinguished.

Zone 1—no problems were found with the results. Interestingly, the uncertainty of the single measurement corresponds to the relative standard deviation of the analysis results from the certificate;

Zone 2—exceeds the threshold of 10% for the uncertainty value of a single sample concentration measurement due to calibration problems;

Zone 3—high uncertainty in measuring sample concentrations correlates with the relative standard deviation data with the certificate. This is explained by the fact that the content of the elements in this zone is close to the detection limit;

Zone 4—problems with the determination of elements in this zone are either due to problems with sample preparation

(sample decomposition) or (less likely) due to the inaccuracy of the certificate.

4 Conclusion

When carrying out mass spectrometric analysis of a series of samples, the device generates large arrays of homogeneous data. A relational database management system (MS Access) is an off-the-shelf solution for handling arrays of homogeneous data. It is shown that the mathematical capabilities of modern relational data management systems (MS Access) are sufficient to process data in a quadrupole ICP-MS. The processing algorithm can be transferred to a network database environment MS SQL, which is easily scalable and is now an industry standard. Users of the developed program receive analysis results in a qualitatively new form (a multidimensional array of structured information), which, in turn, provides an increase in the level of research in the subject area.

References

1. Linge, K.L., Jarvis, K.E.: Quadrupole ICP-MS: introduction to instrumentation, measurement techniques and analytical capabilities. Geostand. Geoanal. Res. **33**(4), 445–467 (2009). https://doi.org/10.1111/j.1751-908X.2009.00039.x

2. Balaram, V.: Current and emerging analytical techniques for geochemical and geochronological studies. Geol. J. **56**(5), 2300–2359 (2021). https://doi.org/10.1002/gj.4005

3. Cotta, A.J.B., Enzweiler, J.: Classical and new procedures of whole rock dissolution for trace element determination by ICP-MS. Geostand. Geoanal. Res. **36**(1), 27–50 (2012). https://doi.org/10.1111/j.1751-908X.2011.00115.x

4. Matusiewicz, H.: Sample preparation for inorganic trace element analysis. Phys. Sci. Rev. **2**(5) (2017). https://doi.org/10.1515/psr-2017-8001

5. Karandashev, V.K., Khvostikov, V.A., Nosenko, S.V., Burmii, Z. P.: Stable highly enriched isotopes in routine analysis of rocks, soils, grounds, and sediments by ICP-MS. Inorg. Mater. **53**(14), 1432–1441 (2017). https://doi.org/10.1134/s0020168517140084

6. Kolotov, V.P., Zhilkina, A.V., Shirokova, V.I., Dogadkin, N.N., Gromyak, I.N., Dogadkin, D.N., Zybinsky, A.M., Tyurin, D.A.: A new approach to sample mineralization in an open system for the analysis of geological samples by inductively coupled plasma mass spectrometry with improved performance characteristics. J. Anal. Chem. **75**(5), 569–581 (2020). https://doi.org/10.1134/S1061934820050081

7. Tharaud, M., Gardoll, S., Khelifi, O., Benedetti, M.F., Sivry, Y.: uFREASI: user-FRiendly Elemental dAta procesSIng. A free and easy-to-use tool for elemental data treatment. Microchem. J. **121**, 32–40 (2015). https://doi.org/10.1016/j.microc.2015.01.011

8. Paton, C., Hellstrom, J., Paul, B., Woodhead, J., Hergt, J.: Iolite: freeware for the visualisation and processing of mass spectrometric data. J. Anal. At. Spectrom. **26**(12), 2508–2518 (2011). https://doi.org/10.1039/c1ja10172b

9. Fleischer, H., Adam, M., Thurow, K., IEEE: Flexible Software solution for rapid manual and automated data evaluation in ICP-MS. In: 2015 IEEE International Instrumentation and Measurement Technology Conference (I2mtc), pp. 1602–1607 (2015)

10. Zhang, L., Ren, Z.Y., Xia, X.P., Li, J., Zhang, Z.F.: IsotopeMaker: a Matlab program for isotopic data reduction. Int. J. Mass Spectrom. **392**, 118–124 (2015). https://doi.org/10.1016/j.ijms.2015.09.019

11. Lopez-Fernandez, H., Pessoa, G.D., Arruda, M.A.Z., Capelo-Martinez, J.L., Fdez-Riverola, F., Glez-Pena, D., Reboiro-Jato, M.: LA-iMageS: a software for elemental distribution bioimaging using LA-ICP-MS data. J. Cheminform. **8**,(2016). https://doi.org/10.1186/s13321-016-0178-7

12. Lugli, F., Weber, M., Giovanardi, T., Arrighi, S., Bortolini, E., Figus, C., Marciani, G., Oxilia, G., Romandini, M., Silvestrini, S., Jochum, K.P., Benazzi, S., Cipriani, A.: Fast offline data reduction of laser ablation MC-ICP-MS Sr isotope measurements via an interactive Excel-based spreadsheet "SrDR." J. Anal. At. Spectrom. **35**(5), 852–862 (2020). https://doi.org/10.1039/c9ja00424f

13. Kolotov, V.P., Atrashkevich, V.V.: Software Aspro-Nuc— gamma-ray spectrometry, routine Naa, isotope identification and data management. J. Radioan. Nucl. Ch. Ar. **193**(2), 195–206 (1995). https://doi.org/10.1007/Bf02039875

14. Kolotov, V.P., Atrashkevich, V.V.: Data-processing in routine neutron-activation analysis using a data-base management-system. J. Anal. Chem. **49**(1), 95–99 (1994)

15. Kolotov, V.P., Zybinsky, A.M., Zhilkina, A.V: Methodology and software for comprehensive ICP-QMS data processing using advanced capabilities of relational data base management system. In: XX Euroanalysis-2019 Conference, Istanbul University, Turkey, September 1–5, pp. 145–147 (2019)