KfK 4721
EUR 11398 EN
Mai 1990

The DoD Method

W. Beyrich, W. Golly, N. Peter, R. Seifert
Institut für Datenverarbeitung in der Technik
Projekt Wiederaufarbeitung und Abfallbehandlung

Kernforschungszentrum Karlsruhe

KERNFORSCHUNGSZENTRUM KARLSRUHE

Institut für Datenverarbeitung in der Technik

Projekt Wiederaufarbeitung und Abfallbehandlung

KfK 4721

EUR 11398 EN

PWA 15/90

THE DOD METHOD

W. Beyrich*), W. Golly, N. Peter, R. Seifert

*) Delegate of the Commission of the European Communities

KERNFORSCHUNGSZENTRUM KARLSRUHE GMBH, KARLSRUHE

Kernforschungszentrum Karlsruhe GmbH
Postfach 3640, 7500 Karlsruhe 1

ISSN 0303-4003

Abstract

The DoD method for robust estimating of variances of measurement series (time series) without the exclusion of outliers is described in detail, including its mathematical-statistical basis. In particular, it is proved that the total of all n(n-1)/2 absolute differences obtainable from n measurement values (realizations of random variables) may be subdivided into groups, each of them consisting of stochastically independent differences only. This is necessary for theoretical foundation of the DoDM estimator. Numerical examples of possible applications are given. Thereby, the evaluation of IDA-80 data presented earlier is completed applying the DoDM estimator.

Die DoD-Methode

Es wird die DoD-Methode zur robusten Schätzung der Varianzen von Meßreihen (Zeitreihen) ohne Ausschluß von Ausreißerwerten einschließlich ihrer mathematisch-statistischen Grundlage umfassend beschrieben. Insbesondere wird bewiesen, daß die Gesamtheit aller n(n-1)/2 absoluten Differenzen, die zwischen n Meßwerten (Realisierungen von Zufallsvariablen) gebildet werden können, derart in Gruppen einteilbar ist, daß jede Gruppe ausschließlich stochastisch unabhängige Größen enthält.Dies ist für die theoretische Fundierung des DoDM-Schätzers notwendig. Zahlenbeispiele für mögliche Anwendungen werden gegeben. Dabei wird die früher beschriebene Auswertung von IDA-80 Daten durch Anwendung des DoDM-Schätzers ergänzt.

CONTENT

LIST OF TABLES AND FIGURES

page

0.1 List of Tables

0.2 List of Figures

1. INTRODUCTION

Estimation of variances or standard deviations of measurement data is an important task in many fields, e.g. in practical safeguards. Conventional evaluation needs homogeneous data material. However, according to the experience, data groups being homogeneous in the statistical sense are the exception.

Therefore, during the last years, an other estimate, the DoD method (*D*istribution *o*f *D*ifferences), was developed at KfK as an - first of all empirical - approach of measurement data evaluation /1, 2, 3/. With this method, the absolute differences of the results of repetitive measurements are used as basis for statistical data treatment. Recent theoretical studies have shown that the DoD method delivers robust estimates for the standard deviation of normally distributed groups of data /4, 5/.

Main advantage of the method is the fact that no application of outlier criteria for the data analysis becomes necessary. Other than in the conventional computing method, the value of the estimate derived for the standard deviation of a group of data is influenced above all by the number and hardly by the quality of existing outliers.

This feature is of particular importance for the evaluation of analytical interlaboratory experiments: It is unsatisfactory to suppress analytical measurement values for merely statistical reasons - however using evaluation techniques like variance analysis, outlier rejection is indispensible in order to obtain sufficiently homogeneous data material.

Furthermore, the selection of the outlier criterion among the many methods described in the literature as well as the arbitrariness in fixing the threshold for data elimination introduce a considerable ambiguity in the statistical elimination of expectations. Being independent from the handling of outliers, estimation of standard deviations by the DoD method always leads to same result, regardless of the statistician who performed the evaluation.

2. MATHEMATICAL-STATISTICAL BACKGROUND

The DoD method (*D*istribution *o*f *D*ifferences) delivers a robust estimate for the standard deviation of a normally distributed random variable X. In the following, the mathematical-statistical basis for the estimation method is presented.

It is assumed that there is a sample of normally distributed random variables $X_1, X_2, \ldots, X_n$ which are independent, identically distributed (i.i.d.) with a given but unknown standard deviation σ. In the following, three possibilities of an estimate for the unknown standard deviation σ are explained.

a) DoDU method

Let n be an even number, i.e. n=2m. Then a new set of random variable is defined as

$$Z_k = | X_{2k} - X_{2k-1} | , \quad k=1,2,\ldots,m \qquad (1)$$

If the sample has an odd number (that means n is odd), the last value is neglected. It is easy to check that $Z_1, Z_2, \ldots, Z_m$ are also independent identically distributed random variables. For the distribution function of Z_k, one gets

$$H(z) = \begin{cases} 2\Phi(z/(\sigma\sqrt{2}))-1, & \text{for } z \geq 0 \\ 0, & \text{otherwise} \end{cases} \qquad (2)$$

where $\Phi(\cdot)$ denotes the normal distribution function with expectation value 0 and standard deviation 1. From Eq.2, one gets at σ

$$q_\sigma = 2\Phi(1/\sqrt{2})-1 \approx 0.52. \qquad (3)$$

Summarily it can be stated that H(z) is a strictly increasing distribution function, see also Fig.1, and that σ is the q_σ-quantile of H(z). According to Fig.1, a suggestion for the estimation of σ is the inverse function of H evaluated at $q_\sigma=0.52$, i.e. $H^{-1}(q_\sigma)$.

Because σ is unknown (it has to be estimated), $H^{-1}(q_\sigma)$ cannot be evaluated. So the empirical distribution $H_m^*(z)$ which is defined as

$$H_m^*(z) = (1/m) \sum_{i=1}^{m} I(Z_i \le z) \tag{4}$$

may be used, where I stands for the indicator variable. If A is an arbitrary boolean expression, the indicator variable I is defined as follows:

$$I(A) = \begin{cases} 1 & \text{if A is true} \\ 0 & \text{if A is false} \end{cases}$$

The inverse function of H_m^* is defined as

$$H_m^{*\,-1}(u) = \inf\{z : H_m^*(z) \ge u\} \tag{5}$$

where $0 \le u \le 1$.

In our case, this line of reasoning leads to the following definition for an estimation function for σ /4/:

$$\text{DoDU} = H_{m=n/2}^{*\,-1}(q_\sigma). \tag{6}$$

In this connection, it is very helpful that $H_m^{*\,-1}(u)$ can be written as order statistics. If one has the simple random sample $Z_1, Z_2, \ldots, Z_m$, then the sample function $Z_{(k)}$, $1 \le k \le m$, denotes the k^{th} position of the ordered sample, i.e.

$$Z_{(1)} \leq Z_{(2)} \leq Z_{(k)} \leq \ldots . Z_{(m)} . \qquad (7)$$

Using this, the estimate DoDU may be written as

$$\text{DoDU} = Z_{([q_\sigma \bullet m]+1)} \qquad (8)$$

where $[q_\sigma \bullet m]$ is the largest integer less than or equal to $q_\sigma \bullet m$. The estimate DoDU is asymptotically normally distributed with mean σ and variance $\{q_\sigma(1-q_\sigma)/(n/2)\}/h(\sigma)$ where $h(z)$ is the density function of $H(z)$. Furthermore, there is

$$\lim_{n\to\infty} P\{|\text{DoDU}-\sigma|>\varepsilon\} = 0, \text{ for all } \varepsilon>0.$$

That means DoDU is a consistent estimator for σ.

b) DoDA method

In Eq.1, page 9, there is a somewhat arbitrary choice for the Z_k's. Also, the random sample is split in half. Another aspect is that for graphical estimation it is more suitable to apply all absolute differences. In sum these considerations lead to the idea of using all differences for the estimation of σ. Defining

$$Y_{ij} = | X_i - X_j | \text{ for all } i<j, \qquad (9)$$

$n(n-1)/2$ absolute differences are obtained which have to be arranged in ascending order $Y_{(1)} \leq Y_{(2)} \leq \ldots . \leq Y_{(n(n-1)/2)}$. Of course, the variables Y_{ij} are not pairwise independent. So not all the theoretical considerations can be applied as for the Z_k variables. Nevertheless, one defines analogous to the estimate DoDU

$$\text{DoDA} = Y_{([q_\sigma \bullet n(n-1)/2]+1)} \qquad (10)$$

as estimate based on all absolute differences.

c) DoDM method

A further possibilty to apply the idea of estimating the standard deviation with a quantile is to devide all n(n-1)/2 absolute differences into (n-1) (in the case of even n) groups of n/2 stochastically independent differences, where in each of the (n-1) groups every original value is considered exactly once. If n is an odd number, n groups of (n-1)/2 stochastically independent differences are formed, where in each of the groups exactly one of the original values is omitted, thus in total each of the n values appears (n-1) times /6/.

The procedure which gives the groups may be defined as follows: For every pair of measurements (X_i,X_j with i<j), the value of function T gives the group to which each of the absolute differences has to be attached; if n is an odd number one has to add a dummy variable and in the following Eq.11a, one has to replace n by n+1. For those even numbers one gets for i<j<n:

$$T(i,j) = \begin{cases} i+j-1, & \text{for } i+j \leq n \\ i+j-n, & \text{for } i+j>n. \end{cases} \qquad (11a)$$

And in the case no dummy variable had to be applied (n was originally even) for i<j=n:

$$T(i,n) = \begin{cases} 2i-1, & \text{for } 2i \leq n \\ 2i-n, & \text{for } 2i>n. \end{cases} \qquad (11b)$$

Tab.I illustrates this procedure for n=10 as well as for n=9. That function T holds for all numbers of n original data, says a lemma which is proved in paragraph 3.

Now the n-1 (or n if n was an odd number) estimates $DoDU_1, DoDU_2, \ldots, DoDU_{(n-1)}$ (or $DoDU_n$) can be calculated and combined by

$$DoDM = 1/(n-1) \sum_{i=1}^{n-1} DoDU_i \quad ; \text{ if n is even} \tag{12a}$$

$$DoDM = 1/n \sum_{i=1}^{n} DoDU_i \quad ; \text{ if n is odd} \tag{12b}$$

to a single estimate of the standard deviation σ. It is obvious that DoDM is a consistent estimate of σ and has all the other statistical features of DoDU. Up to now, nothing can be said about its variance. But MONTE-CARLO experiments (see e.g. Tab.II) show very good results for DoDM.

3. PROOF OF THE LEMMA

Basis of the evaluation are all differences (see Eq.9, page 11)

$$Y_{ij} = | X_i - X_j |$$

with $1 \le i < j \le n$,
(there are $n(n-1)/2$ different differences)

which can be formed from a group of n measurement values.

Task:

The subdivision of those $n(n-1)/2$ differences has to be done into $(n-1)$ groups of $n/2$ stochastically independent differences for even n, or into n groups of $(n-1)/2$ stochastically independent differences for odd n, respectively.

Theorem:

Let

$$Y_{ij} = | X_i - X_j | \quad \forall \ 1 \le i < j \le n$$

be $n(n-1)/2$ differences.

For even n, those differences can be subdivided into $(n-1)$ groups of $n/2$ stochastically independent differences each in such a way that each X_k, $k=1,...,n$, is used in each group exactly once.

For odd n, first a dummy value is added. Then those differences can be subdivided into n groups of $(n+1)/2$ stochastically independent differences each in such a way that each X_k, $k=1,...,n$, is used in each group exactly once. After rejection of those differences formed using the dummy value, each group contains at least (n-1)/2 differences and in each of the n groups one of the n measurement values is omitted.

In both cases, therefore, each X_k, k=1,...,n, appears exactly (n-1) times.

For each difference $|X_i - X_j|$, with $1 \leq i < j \leq n$, the appropriate subdivision group might be given by the value of function T, which is defined as follows:

For even numbered n and $1 \leq i < j < n$ by:

$$T(i,j) = \begin{cases} i+j-1, & \text{for } i+j \leq n \\ i+j-n, & \text{for } i+j > n. \end{cases} \tag{13a}$$

For even numbered n and $1 \leq i < j = n$ by:

$$T(i,j) = \begin{cases} 2i-1, & \text{for } 2i \leq n \\ 2i-n, & \text{for } 2i > n. \end{cases} \tag{13b}$$

And for odd numbered n (in this case a dummy value is added) and for $1 \leq i < j \leq n$:

$$T(i,j) = \begin{cases} i+j-1, & \text{for } i+j \leq n+1 \\ i+j-n-1, & \text{for } i+j > n+1. \end{cases} \tag{13c}$$

Proof:

May be T(i,j)=k, whereby k is free selected but fixed with $1 \leq k \leq n-1$.

Case 1 : n even

Case 1.1 : k even

Case 1.1.1: $i+j \leq n$, $j<n$

→ $k+1 = i+j$

→ i takes on values between 1 and k/2, in addition j takes on values between k/2 and k

→ k/2 differences

Case 1.1.2: $i+j>n$, $j<n$

→ $k+n = i+j$

→ j takes on values between ((k+n)/2)+1 and n-1, in addition i takes on values between k+1 and ((k+n)/2)-1

→ (n/2)-(k/2)-1 differences

Case 1.1.3: $2i \leq n$, $j=n$

→ $k+1 = 2i$ is impossible

Case 1.1.4: $2i>n$, $j=n$

→ $k+n = 2i$

→ $i = (k+n)/2$

→ *one* difference

Intermediate result of case 1.1 →

There are $(k/2)+n-(n/2)-(k/2)-1+1 = n/2$ differences in each group and each X_k, $k=1,\ldots,n$, appears exactly once.

Case 1.2 : k odd

Case 1.2.1: $i+j \leq n$, $j<n$

→ $k+1 = i+j$

→ i takes on values between 1 and ((k+1)/2)-1, in addition j takes on values between ((k+1)/2)+1 and k

→ ((k+1)/2)-1 differences

Case 1.2.2: $i+j>n$, $j<n$

→ $k+n = i+j$

→ j takes on values between $(k+n+1)/2$ and $n-1$, in addition i takes on values between $k+1$ and $((k+n+1)/2)-1$

→ $(n/2)-(k+1)/2$ differences

Case 1.2.3: $2i\leq n$, $j=n$

→ $k+1 = 2i$

→ $i = (k+1)/2$

→ *one* difference

Case 1.2.4: $2i>n$, $j=n$

→ $k+n = 2i$ is impossible

Intermediate result of case 1.2 →

There are $(k+1)/2-1+(n/2)-(k+1)/2+1 = n/2$ differences in each group and each X_k, $k=1,...,n$, appears exactly once

Case 2 : n odd

Case 2.1 : k even

Case 2.1.1: $i+j\leq n+1$

→ $k+1 = i+j$

→ i takes on values between 1 and $k/2$, in addition j takes on values between $(k/2)+1$ and k

→ $k/2$ differences

Case 2.1.2: $i+j>n+1$

→ $k+n+1 = i+j$

→ j takes on values between $((k+n+1)/2)+1$ and n, in addition i takes on values between $k+1$ and $((k+n+1)/2)-1$

→ $n-((k+n+1)/2)$ differences

Intermediate result of case 2.1 →

There are $(k/2)+n-(k/2)-(n/2)-1/2 = (n-1)/2$
differences in each group
and each X_k, $k=1,...,n$, appears exactly once
($(k+n+1)/2$ is attached to a dummy value)

Case 2.2 : k odd

Case 2.2.1: $i+j \leq n+1$

→ $k+1 = i+j$
→ i takes on values between 1 and $((k+1)/2)-1$,
in addition j takes on values between
$((k+1)/2)+1$ and k
→ $((k+1)/2)-1$ differences

Case 2.2.2: $i+j > n+1$

→ $k+n+1 = i+j$
→ j takes on values between $((k+n)/2)+1$ and n,
in addition i takes on values between $k+1$
and $(k+n)/2$
→ $n-(k+n)/2$ differences

Intermediate result of case 2.2 →

There are $(k+1)/2-1+n-(k+n)/2 =$
$= k/2-1/2+n-k/2+n-n/2 = (n-1)/2$
differences in each group
and each X_k, $k=1,...,n$, appears exactly once
($(k+1)/2$ is attached to a dummy value).

4. PRACTICAL EXPERIENCE WITH THE DOD METHOD

Instead of the measurement values themselves, the DoD method uses as basic statistical elements the absolute values of all differences which can be formed between them. This feature makes the DoD method also suitable to estimate the probability of occurrence of a certain deviation between the results of repetitive analyses of the same material.

Although it cannot be analytically proved that the DoDA method is a consistant estimator for the standard deviation σ, the results of Monte-Carlo simulations demonstrate a very good performance which is quite close to that of the DoDM estimate, see Tab.II. Therefore, the DoDA estimate can and should be used if a graphical evaluation is performed as discussed in paragraph 6.

On the other hand, for computerized numerical calculations, the DoDM method should be preferred due to its proved theoretical qualities.

As a rule of thumb, meaningful application of the DoDA method requires at least $n=5$ measurement data from which $N=n(n-1)/2=10$ differences can be established. If several smaller groups of repetition measurements of the same kind but belonging to different expectation values are available, the differences calculated separately for each group can be pooled.

For normally distributed groups of data, the DoDA estimate is identical with that calculated conventionally. If extreme values exist, but not more than 20 to 25% of the data, the value of the estimate derived for the standard deviation is influenced above all by their number, and very little by their quality. Only if the percentage of outliers exceeds this limit, the DoDA estimate clearly increases depending on their quality.

In the many cases where the group contains values which do not belong to a normal distribution but are below the threshold chosen for outlier rejection, the DoDA estimate is lower than the conventionally calculated standard deviation. The reason is that the DoD method has the tendency to suppress data deviating from normal distribution.

5. SAFEGUARDS APPLICATIONS

As mentioned above, the estimation of variances of measurement data is an important task in many fields. One important example is the safeguards application /7/.

At least four fields of DoD application in practical safeguards can be identified:

1. Estimation of the so-called 'interlaboratory spread' in analytical intercomparison programs, which is a measure for the scattering of the analytical results obtained by different laboratories around the true value,

2. Estimation of the average repeatability/reproducibility for a certain analytical procedure derived from the individual repeatabilities/reproducibilities of a number of laboratories,

3. Estimation of the repeatability/reproducibility for a certain laboratory (for a certain analytical procedure) based on observations over a long time period, and

4. Estimation of the probability of occurrence for the difference observed in the measurement results of two laboratories (e.g. shipper-receiver difference or operator-inspector difference) using the DoD curve derived in an interlaboratory measurement program for the appropriate analytical procedure.

6. EXAMPLE OF PRACTICAL APPLICATION

Practical application of the DoD method is illustrated with the following example:

In an analytical interlaboratory experiment /8/ n=9 laboratories determined the Pu-238 isotopic content (weight %) of the same sample material by mass spectrometry with the following results[1] which are also displayed in Fig.2:

$x_1 = 0.2043$	$x_2 = 0.2070$	$x_3 = 0.2061$
$x_4 = 0.1706$	$x_5 = 0.2152$	$x_6 = 0.2062$
$x_7 = 0.2108$	$x_8 = 0.2019$	$x_9 = 0.2175$

The value $x_4 = 0.1706$ is obviously an outlier.

Conventional estimation of the standard deviation of this data group without rejection of x_4 would result in

$$s_9 = \{(1/(9-1)) \sum_{i=1}^{9} (x_i-\bar{x})^2\}^{1/2} = 0.0137 \qquad (14)$$

and after rejection of x_4 in $s_8 = 0.0054$.

Applying the DoDA method, the n(n-1)/2=36 absolute differences of type $Z_{(k)}=Y_{ij}=|X_i-X_j|$ for all i<j are calculated and arranged according to:

---

[1] Data taken from Ref./9/, Evaluation Sheet 55, p.150.

$Z_{(1)}=Y_{36}=0.0001$;	$Z_{(2)}=Y_{26}=0.0008$;	$Z_{(3)}=Y_{23}=0.0009$;
$Z_{(4)}=Y_{13}=0.0018$;	$Z_{(5)}=Y_{16}=0.0019$;	$Z_{(6)}=Y_{59}=0.0023$;
$Z_{(7)}=Y_{18}=0.0024$;	$Z_{(8)}=Y_{12}=0.0027$;	$Z_{(9)}=Y_{27}=0.0038$;
$Z_{(10)}=Y_{38}=0.0042$;	$Z_{(11)}=Y_{68}=0.0043$;	$Z_{(12)}=Y_{57}=0.0044$;
$Z_{(13)}=Y_{67}=0.0046$;	$Z_{(14)}=Y_{37}=0.0047$;	$Z_{(15)}=Y_{28}=0.0051$;
$Z_{(16)}=Y_{17}=0.0065$;	$Z_{(17)}=Y_{79}=0.0067$;	$Z_{(18)}=Y_{25}=0.0082$;
$Z_{(19)}=Y_{78}=0.0089$;	$Z_{(20)}=Y_{56}=0.0090$;	$Z_{(21)}=Y_{35}=0.0091$;
$Z_{(22)}=Y_{29}=0.0105$;	$Z_{(23)}=Y_{15}=0.0109$;	$Z_{(24)}=Y_{69}=0.0113$;
$Z_{(25)}=Y_{39}=0.0114$;	$Z_{(26)}=Y_{19}=0.0132$;	$Z_{(27)}=Y_{58}=0.0133$;
$Z_{(28)}=Y_{89}=0.0156$;	$Z_{(29)}=Y_{48}=0.0313$;	$Z_{(30)}=Y_{14}=0.0337$;
$Z_{(31)}=Y_{34}=0.0355$;	$Z_{(32)}=Y_{46}=0.0356$;	$Z_{(33)}=Y_{24}=0.0364$;
$Z_{(34)}=Y_{47}=0.0402$;	$Z_{(35)}=Y_{45}=0.0446$;	$Z_{(36)}=Y_{49}=0.0469$.

The estimate DoDA for the standard deviation of the above nine measurement values is given by Eq.10, page 11. Because there is always $q_\sigma=0.52$ and in this example $n(n-1)/2=36$ (total number of differences), there is

$$\text{DoDA} = Z_{([0.52\bullet 36]+1)} = Z_{([18.72]+1)} = Z_{(19)}=0.0089.$$

Compared to the conventional estimate for the same population $s_9=0.0137$, this value lies closer to the estimate $s_8=0.0054$, derived by conventional calculation after outlier rejection. In particular, please note that - different from calculation by Eq.14, page 25 - this DoDA estimate is completely independent of the actual value of the outlier x_4: As can be verified by the compilation of differences above, only the last eight values $Z_{(29)}$ to $Z_{(36)}$ are influenced by the outlier x_4, whereas the estimate of the standard deviation is given by $Z_{(19)}$[2].

---

[2] If x_4 is rejected, DoD evaluation results in the differences $Z_{(1)}$ to $Z_{(28)}$, and the DoDA estimate for the standard deviation is given by $Z_{(15)}=0.0051$ in good agreement with the estimate $s_8=0.0054$ obtained by conventional calculation.

In Fig.3, the cumulative distribution of the 36 absolute differences is plotted in increasing order ('DoDA display'). It is helpful for judging a difference observed between the results of two laboratories, e.g. those of plant operator and control authority or shipper and receiver: The corresponding ordinate value gives the probability of occurrence of a difference equal to or smaller than the observed one in the analytical assay in question. After threshold values are fixed for considering observed differences as 'acceptable', 'suspicious' or 'unacceptable', such DoDA displays may be used as a tool for verification of safeguards data /10/.

The estimate of the standard deviation of the nine measurement results is given by the abscissa value corresponding to the 52%-ordinate value as indicated[3]. According to the curve drawn empirically, a value of 0.0087 is found with the graphical method compared to 0.0089 calculated above. This discrepancy is diminishing with increasing number n of measurement data.

The horizontal part of the shape of the curve indicates the existence of the outlier value x_4: The eight data points of right section of the curve originate from differences calculated with this outlier value. Plateaus of this type produced by more than one outlier value may indicate a typical source of error.

---

[3] In earlier publications, the differences were plotted in decreasing order. In such case, the estimate of the standard deviation is given by the abscissa value corresponding to the 48%-ordinate value, the complement to the 52%-quantile.

To illustrate the calculation of the appropriate DoDM estimate, now

- the n=9 subgroups (odd case) of independent differences will be created according to Tab.I. The differences within the subgroups are arranged in increasing order of their amounts in order to determine the DoDU estimates, and

- the n=9 DoDU estimates are picked up according to Eq.8, page 11. As n is an odd number, m is given by m=(n-1)/2=4 and therefore $DoDU=Z_{([q_\sigma \cdot m]+1)}=Z_{([0.52 \cdot 4]+1)}=Z_{([2.08]+1)}=Z_{(2+1)}=Z_{(3)}$ as follows:

Subgroup 1:[4]

$Z_{(11)} = Y_{38} = 0.0042$ $\quad Z_{(12)} = Y_{56} = 0.0090$
$Z_{(13)} = Y_{29} = 0.0105$ $\quad Z_{(14)} = Y_{47} = 0.0402$
$DoDU_1 = Z_{(13)} = 0.0105$

Subgroup 2:

$Z_{(21)} = Y_{12} = 0.0027$ $\quad Z_{(22)} = Y_{57} = 0.0044$
$Z_{(23)} = Y_{39} = 0.0114$ $\quad Z_{(24)} = Y_{48} = 0.0313$
$DoDU_2 = Z_{(23)} = 0.0114$

Subgroup 3:

$Z_{(31)} = Y_{13} = 0.0018$ $\quad Z_{(32)} = Y_{67} = 0.0046$
$Z_{(33)} = Y_{58} = 0.0133$ $\quad Z_{(34)} = Y_{49} = 0.0469$
$DoDU_3 = Z_{(33)} = 0.0133$

---

[4] In $Z_{(ik)}$ index i refers to the subgroup according to Tab.I, and k gives the ordered numbering of the differences within the subgroup.

Subgroup 4:

$Z_{(41)} = Y_{23} = 0.0009$ $\quad Z_{(42)} = Y_{59} = 0.0023$

$Z_{(43)} = Y_{68} = 0.0043$ $\quad Z_{(44)} = Y_{14} = 0.0337$

$DoDU_4 = Z_{(43)} = 0.0043$

Subgroup 5:

$Z_{(51)} = Y_{78} = 0.0089$ $\quad Z_{(52)} = Y_{15} = 0.0109$

$Z_{(53)} = Y_{69} = 0.0113$ $\quad Z_{(54)} = Y_{24} = 0.0364$

$DoDU_5 = Z_{(53)} = 0.0113$

Subgroup 6:

$Z_{(61)} = Y_{16} = 0.0019$ $\quad Z_{(62)} = Y_{79} = 0.0067$

$Z_{(63)} = Y_{25} = 0.0082$ $\quad Z_{(64)} = Y_{34} = 0.0355$

$DoDU_6 = Z_{(63)} = 0.0082$

Subgroup 7:

$Z_{(71)} = Y_{26} = 0.0008$ $\quad Z_{(72)} = Y_{17} = 0.0065$

$Z_{(73)} = Y_{35} = 0.0091$ $\quad Z_{(74)} = Y_{89} = 0.0156$

$DoDU_7 = Z_{(73)} = 0.0091$

Subgroup 8:

$Z_{(81)} = Y_{36} = 0.0001$ $\quad Z_{(82)} = Y_{18} = 0.0024$

$Z_{(83)} = Y_{27} = 0.0038$ $\quad Z_{(84)} = Y_{45} = 0.0446$

$DoDU_8 = Z_{(83)} = 0.0038$

Subgroup 9:

$Z_{(91)} = Y_{37} = 0.0047$ $\quad Z_{(92)} = Y_{28} = 0.0051$

$Z_{(93)} = Y_{19} = 0.0132$ $\quad Z_{(94)} = Y_{46} = 0.0356$

$DoDU_9 = Z_{(93)} = 0.0132$

According to Eq.12b, page 13, the n=9 DoDU estimates now are combined to

$$DoDM = 1/9 \sum_{i=1}^{9} DoDU_i = 0.0095.$$

With respect to the variances given in Tab.II, this shows that the DoDM estimate (0.0095) is in good agreement with both the numerical (0.0089) and the graphical (0.0087) DoDA estimates derived above[5].

The evaluations above relate to the interlaboratory spread. If x_i are repetitive measurement results obtained within one laboratory, the estimate of the standard deviation derived by the DoD method describes the within-laboratory repeatability. If such data exist from different laboratories, the absolute differences may be calculated for each laboratory separately and then pooled to one DoD display. From that, an average value for the within-laboratory repeatability of the analytical method in question can be estimated.

---

[5] The small discrepancy between DoDA and DoDM diminishes with increasing number n of measurement data.

7. EVALUATION OF IDA-80 DATA BY THE DOD METHOD

An extended comparison of DoD evaluation results with those obtained by conventional data treatment was made for the IDA-80 measurement evaluation program /11/[6]. This comparison was of particular interest, since the handling of outliers had played a substantial role in the evaluation performed by conventional methods.

The data material of the IDA-80 program consists of concentration values for uranium and plutonium determined in samples of different solutions and of isotopic compositions determined for these elements. Each assay was made by a group of 24 to 30 laboratories.

In Tab.III, four estimates are given for the (relative) standard deviation of the measurement results of each assay:

- the DoD estimate DoDA (column 4, left part),
- the DoD estimate DoDM (column 4, right part),
- the conventional estimate without outlier rejection (column 5),
- and the conventional estimate after outlier rejection as performed in the official evaluation of the IDA-80 program (extreme mean values were checked using the Bartsch criterion /12/ and exceptionally high repeatability values were checked using the Dixon criterion /13/ with $\alpha \leq 1\%$), see column 6.

---

[6] The analytical measurement program relates to the most recent mass spectrometric isotope dilution analysis of uranium and plutonium in input solutions of a reprocessing plant for spent nuclear fuels.

In Tab.III and Tab.IV, the estimates for the interlaboratory spread and the average values of the within-laboratory uncertainty component obtained by the DoD method (DoDA and DoDM estimates in column 4) have been compared with the results obtained in the IDA-80 evaluation after rejection of 10.3% of measurement data as outliers (columns 6). In addition, those estimates have been entered in the tables which result from the conventional calculation with no outlier criteria applied (columns 5).

The following observations can be made:

1. The conventional estimates without outlier criteria applied are higher in nearly all cases (111 out of 112) than the DoD values (DoDA as well as DoDM estimates) - in many cases higher by several hundred percent, see columns 9 in Tab.III and Tab.IV. This confirms the expectation that the values estimated according to the DoD method are little influenced by (individual) extreme values.

2. For the interlaboratory spread (Tab.III), the deviations of the DoD estimates DoDA and DoDM from the conventional results excluding outliers are about as frequently positive as they are negative, see column 10.

   The estimates DoDA and DoDM occur in more than 80% within the 99%-confidence interval of the estimates determined in the IDA-80 evaluation, see column 11[7].

---

[7] If no outlier criteria had been applied in the IDA-80 evaluation, the DoDA and DoDM estimates would occur within the 99%-confidence intervals in only 5 out of the 28 cases (18%).

For those cases where this does not apply, more detailed studies have revealed that this is very probably attributable to the exclusion of the outliers: If the DoD estimates occur below the confidence interval, the last extreme value considered narrowly missed the condition for exclusion, or vice versa.

The observations indicate a satisfactory agreement of the obtained estimates DoDA and DoDM of the interlaboratory spread with those of the IDA-80 evaluation. Since the IDA-80 estimates scatter around the DoD estimates and do not exhibit any bias, application of the Bartsch-outlier criterion /12/ to the laboratory mean values in the IDA-80 evaluation was obviously meaningful.

3. For the within-laboratory uncertainty component (Tab.IV), the IDA-80 estimate exceeds both the DoDA and DoDM estimates in 24 out of 28 (48 out of 56) cases, see column 10, and the DoDM estimates fall in 43% of the cases (12 out of 28) in the 99%-confidence interval of the IDA-80 value. The DoDA estimates, however, fall in only 32% of the cases (9 out of 28)[8] in the 99%-confidence interval of the IDA-80 value, see column 11. The reason may be that the DoDM estimate is mathematically proved to be a consistent estimator for standard deviations.

---

[8] The reason for this rather poor agreement was investigated in more detail in Ref./11/ on the basis of the DoDA estimate only. The values of 32%, 54%, 64% and 75% given in Table VI of Ref./11/ improve to 43%, 64%, 75% and 79%, respectively, if the DoDM estimate is used.

8. ESTIMATION OF THE LONG-TERM REPEATABILITY/REPRODUCIBILITY WITHIN ONE LABORATORY

Over two years, a total of n=101 determinations of the correction factor for isotope fractionation were performed in the mass spectrometer laboratory of the Central Bureau for Nuclear Measurements using the $^{235}U/^{238}U$ ratio. In irregular sequence, eight different isotopic standards of the National Bureau of Standards had been applied /7/.

Fig.4 shows the values of the correction factors standardized to the median value of 0.99940 in the original time series. The total spread is below 0.3%. Conventional calculation of the relative standard deviation results in the estimate 0.053% and 0.99939 for the arithmetic mean value.

In Fig.5, the DoDA display of the n(n-1)/2=5050 differences is shown. The 52%-quantile delivers 0.052% as DoDA estimate of the relative standard deviation[9].

The excellent agreement of the RSD estimates obtained by the DoDA method and by conventional statistics, as well as the agreement of the median with the arithmetic mean value are due to the high degree of homogeneity of the data group. Application of the Bartsch criterion /12/ indicates no outlier value.

Analogous to this example, RSD estimates can be derived for the various procedures performed in any analytical laboratory.

---

[9] In this case, the RSD estimate was determined directly, because the data used to establish the differences are standardized. Thus, the RSD estimate relates to the median value used for standardization.

In safeguards, they are of interest e.g. for establishing facility attachments.

9. CONCLUSION

The DoD method is a reasonable way to estimate standard deviations in many fields. This method is especially appropriate to cases where the data include outliers. Being free of the arbitrariness associated with the use of outlier criteria, the DoD method always leads to reliable estimates. This is of particular interest for safeguards applications.

This publication proved that DoDM is a consistent estimator for standard deviations. DoDM, therefore, should be preferred for computerized numerical calculations.

In the case a graphical evaluation is preferred the estimate DoDA should be used. This graphical display is helpful for judging measurement discrepancies.

REMARK: In order to facilitate practical application of the DoD method, a computer program is currently developed at Karlsruhe. The authors intend to make it available to any user who is interested in.

10. REFERENCES

/ 1 / W. Beyrich, "The Problem of Analytical Interlaboratory Differences in Practical Safeguards", Safeguarding Nuclear Materials, Vol.2, IAEA, Vienna, (1976) p.175.

/ 2 / W. Beyrich, W. Golly, G. Spannagel, "The DoD-Method: An Empirical Approach to the Treatment of Measurement Data Comprising Extreme Values", 3rd Annual Symposium on Safeguards and Nuclear Material Management, ESARDA-13, Karlsruhe (1981) p.289-294

/ 3 / W. Beyrich, W. Golly, G. Spannagel, "Some Features and Applications of an Empirical Approach to the Treatment of Measurement Data", 4th Sale Program Participants Meeting, Argonne, 1981, Washington, D.C.: U.S.DoE (1981) p.60-78

/ 4 / R. Beedgen, "Robust Estimation of Standard Deviations Using Ordered Samples", LA-9238-MS, Los Alamos (1982)

/ 5 / R. Beedgen, "Statistical Analysis of the DoD-Method", 6rd Annual Symposium on Safeguards and Nuclear Material Management, ESARDA-17, Venice (1984) p.533-538

/ 6 / W. Beyrich, "Study on the DoD Method of Measurement Data Evaluation", KfK 4077/EUR 9616e Karlsruhe (1986)

/ 7 / W. Beyrich, W. Golly, P. De Bievre, "Contribution to the Evaluation of Measurement Data in Practical Safeguards", 11th Annual Symposium on Safeguards and Nuclear Material Management, ESARDA-22, Luxembourg (1989) p.341-344

/ 8 / W. Beyrich, W. Golly, G. Spannagel, P. De Bievre W. Wolters, "The IDA-80 Measurement Evaluation Programme on Mass Spectrometric Isotope Dilution Analysis of Uranium and Plutonium: Volume I: Design and Results", KfK 3760/EUR 7990e, Karlsruhe (1984)

/ 9 / W. Beyrich, W. Golly, G. Spannagel, "The IDA-80 Measurement Evaluation Programme on Mass Spectrometric Isotope Dilution Analysis of Uranium and Plutonium: Volume III: Compilation of Evaluation Data", KfK 3762/EUR 7992e, Karlsruhe (1985)

/ 10 / W. Beyrich, G. Spannagel, "Practical Approach to Procedure of Judging the Results of Analytical Verification Measurements", Nuclear Safeguards Technology, II, IAEA, Vienna (1979) p.347

/ 11 / W. Beyrich, W. Golly, "Evaluation of IDA-80 Data by the DoD Method", KfK 4157/EUR 10533EN, Karlsruhe (1989)

/ 12 / H.J. Bartsch, "Handbook of Mathematical Formulas", Academic Press Inc., New York (1974)

/ 13 / W.J. Dixon, F.J. Massay, "Introduction to Statistical Analysis", McGraw-Hill Inc., New York (1969)

TAB. I: FUNCTION T DEFINING THE NUMBERS OF GROUPS TO WHICH EACH DIFFERENCE Y_{ij} IS ATTACHED;
$Y_{ij}=|X_i-X_j|$, $i<j$, $n=10$ or 9, respectively[1]

↓ i \ j →	1	2	3	4	5	6	7	8	9	10
1		2	3	4	5	6	7	8	9	1
2			4	5	6	7	8	9	1	3
3				6	7	8	9	1	2	5
4					8	9	1	2	3	7
5						1	2	3	4	9
6							3	4	5	2
7								5	6	4
8									7	6
9										8

[1] FOR $n=9$ OMIT THE LAST COLUMN.

TAB. II: MONTE-CARLO SIMULATIONS FOR DoDU, DoDA AND DoDM BASED ON 10,000 RUNS AND STANDARD NORMAL SAMPLES (i.e. $\sigma=1$)

1	2	3	4	5	6	7	8
	DoDU			DoDA		DoDM	
SAMPLE SIZE n	ESTIMATE σ	STANDARD DEVIATION	THEORETICAL VALUE[1]	ESTIMATE σ	STANDARD DEVIATION	ESTIMATE σ	STANDARD DEVIATION
6	1.04	0.58	0.66	1.05	0.39	1.03	0.38
10	1.01	0.47	0.51	1.05	0.28	1.01	0.27
20	1.09	0.36	0.35	1.01	0.19	1.09	0.20
40	1.02	0.25	0.25	1.01	0.13	1.02	0.13
80	1.03	0.16	0.16	1.00	0.08	NOT DETERMINED	

[1] FOR GREAT SAMPLE SIZES, THE THEORETICAL VALUE OF THE STANDARD DEVIATION OF THE DoDU ESTIMATE AMOUNTS TO $[\{q_\sigma(1-q_\sigma)\}/h(\sigma)]^{(1/2)}$.

TAB.III: ESTIMATES OF RELATIVE STANDARD DEVIATIONS OF THE INTERLABORATORY SPREAD IN IDA-80;
DoD METHOD IN COMPARISON WITH VARIANCE ANALYSIS (DIXON WITH ALPHA LESS THAN OR EQUAL TO 1%)

1	2	3	4	5	6	7	8	9	10	11	12
			ESTIMATE OF RSD OF INTERLABORATORY SPREAD								
TYPE OF DETERMINATION	PART OF IDA-80 PROGRAM	NUMBER OF LABS	DoD METHOD DoDA / DoDM sR1/sR4 (%)	VAR. ANALYSIS OF ALL DATA sR2 (%)	VAR. ANALYSIS[1] WITHOUT OUTLIERS sR3 (%)	NUMBER[2] OF LABS EXCLUDED	99%-CONFIDENCE LIMITS OF sR3 (%)	DEVIATION OF sR2 FROM sR1 / sR4 (%)	DEVIATION OF sR3 FROM sR1 / sR4 (%)	sR1 OR sR4 WITHIN CONFIDENCE LIMITS OF sR3?	REF. ON PAGE
CONCENTRATION:											
U-ELEMENT	1.11	30	0.59 / 0.55	1.72	0.72	2 + 0	0.53; 1.09	+ 192/ + 213	+ 22/ + 31	YES	18
U-ELEMENT	1.12	27	0.78 / 0.75	2.70	1.03	2 + 0	0.75; 1.60	+ 246/ + 260	+ 32/ + 37	YES	20
U-ELEMENT	1.2	30	0.73 / 0.65	3.01	0.53	3 + 2	0.38; 0.83	+ 312/ + 363	- 27/ - 18	YES	22
U-ELEMENT	2.1	28	0.82 / 0.86	2.64	0.48	3 + 3	0.34; 0.78	+ 222/ + 207	- 41/ - 44	NO	24
U-ELEMENT	2.2	28	0.47 / 0.51	0.82	0.34	2 + 2	0.25; 0.53	+ 74/ + 61	- 28/ - 33	YES	26
U-ELEMENT	2.3	27	0.58 / 0.53	0.93	0.40	2 + 2	0.29; 0.64	+ 60/ + 75	- 31/ - 25	YES	28
Pu-ELEMENT	1.11	28	0.97 / 1.01	1.90	0.82	2 + 0	0.60; 1.26	+ 96/ + 88	- 15/ - 19	YES	30
Pu-ELEMENT	1.12	26	1.95 / 1.79	3.04	2.56	0 + 2	1.85; 4.03	+ 56/ + 70	+ 31/ + 43	YES[3]	32
Pu-ELEMENT	1.2	29	0.65 / 0.73	3.77	0.35	3 + 3	0.25; 0.56	+ 480/ + 416	- 46/ - 52	NO	34
Pu-ELEMENT	2.1	26	1.34 / 1.36	3.20	1.25	2 + 1	0.90; 1.99	+ 139/ + 135	- 7/ - 8	YES	36
Pu-ELEMENT	2.2	27	0.43 / 0.43	0.66	0.35	2 + 1	0.25; 0.55	+ 53/ + 53	- 19/ - 19	YES	38
Pu-ELEMENT	2.3	26	0.49 / 0.48	0.52	0.50	0 + 3	0.36; 0.80	+ 6/ + 8	+ 2/ + 4	YES	40
ISOTOPE ABUNDANCE:											
U-234 ASSAY	1.11	27	4.60 / 4.56	6.36	4.01	1 + 0	2.93; 6.16	+ 38/ + 39	- 13/ - 12	YES	42
U-234 ASSAY	2.1	25	5.62 / 6.92	9.70	6.08	1 + 0	4.39; 9.56	+ 73/ + 40	+ 8/ - 12	YES	44
U-235 ASSAY	1.11	30	0.59 / 0.55	1.53	0.33	5 + 1	0.24; 0.52	+ 159/ + 178	- 44/ - 40	NO	46
U-235 ASSAY	2.1	28	0.72 / 0.77	2.98	0.51	3 + 3	0.36; 0.82	+ 314/ + 287	- 29/ - 34	YES	48
U-236 ASSAY	1.11	30	1.63 / 1.55	5.52	2.26	1 + 2	1.66; 3.44	+ 239/ + 256	+ 39/ + 46	NO	50
U-236 ASSAY	2.1	25	8.96 / 9.19	14.2	7.42	1 + 1	5.32;11.8	+ 58/ + 55	- 17/ - 19	YES	52
Pu-238 ASSAY	1.11	26	5.22 / 4.95	13.7	7.01	1 + 1	5.06;11.0	+ 162/ + 177	+ 34/ + 42	YES	54
Pu-238 ASSAY	2.1	24	5.29 / 8.05	16.2	6.21	2 + 2	4.36;10.3	+ 206/ + 101	+ 17/ - 23	YES	58
Pu-239 ASSAY	1.11	29	0.11 / 0.12	0.16	0.13	1 + 2	0.095;0.20	+ 45/ + 33	+ 18/ + 8	YES	62
Pu-239 ASSAY	2.1	27	0.079/ 0.074	0.093	0.093	0 + 0	0.068;0.14	+ 18/ + 26	+ 18/ + 26	YES	64
Pu-240 ASSAY	1.11	29	0.15 / 0.16	0.32	0.14	1 + 1	0.10; 0.21	+ 113/ + 100	- 7/ - 13	YES	66
Pu-240 ASSAY	2.1	27	0.11 / 0.11	0.14	0.14	0 + 0	0.10; 0.21	+ 27/ + 27	+ 27/ + 27	YES	68
Pu-241 ASSAY	1.11	29	0.46 / 0.56	0.54	0.56	0 + 4	0.41; 0.87	+ 17/ - 4	+ 22/ ± 0	YES	70
Pu-241 ASSAY	2.1	27	0.57 / 0.53	0.62	0.62	0 + 0	0.45; 0.94	+ 9/ + 17	+ 9/ + 17	YES	72
Pu-242 ASSAY	1.11	29	1.07 / 1.08	1.48	1.17	1 + 0	0.86; 1.77	+ 38/ + 37	+ 9/ + 8	YES	74
Pu-242 ASSAY	2.1	27	1.61 / 1.45	3.18	1.26	2 + 0	0.91; 1.96	+ 98/ + 119	- 22/ - 13	YES	76
TOTAL:		771				43 + 36 = 79 (10.3%)		55+; 1-	25.5+; 30.5-	24 4	

[1] FIGURES CORRESPOND TO THE OFFICIAL IDA-80 EVALUATION.

[2] THE NUMBER OF EXCLUDED LABORATORIES IS SPLIT WITH RESPECT TO THE REASON FOR REJECTION: FIGURES REFER TO LABORATORIES EXCLUDED FOR EXTREME MEAN VALUES (BARTSCH CRITERION) AND FOR EXCEPTIONALLY HIGH REPEATABILITY VALUES (DIXON CRITERION).

[3] ONLY VALID FOR DoDA.

TAB. IV: ESTIMATES OF RELATIVE STANDARD DEVIATIONS OF THE WITHIN-LABORATORY UNCERTAINTY COMPONENT (REPEATABILITY) IN IDA-80;
DoD METHOD IN COMPARISON WITH VARIANCE ANALYSIS (DIXON WITH ALPHA LESS THAN OR EQUAL TO 1%)

1	2	3	4	5	6	7	8	9	10	11	12
			ESTIMATE OF RSD OF REPEATABILITY								
TYPE OF DETERMINATION	PART OF IDA-80 PROGRAM	NUMBER OF LABS	DoD METHOD DoDA / DoDM sR1/sR4 (%)	VAR. ANALYSIS OF ALL DATA sR2 (%)	VAR. ANALYSIS[1] WITHOUT OUTLIERS sR3 (%)	NUMBER[2] OF LABS EXCLUDED	99%-CONFIDENCE LIMITS OF sR3 (%)	DEVIATION OF sR2 FROM sR1 / sR4 (%)	DEVIATION OF sR3 FROM sR1 / sR4 (%)	sR1 OR sR4 WITHIN CONFIDENCE LIMITS OF sR3?	REF. ON PAGE
CONCENTRATION:											
U-ELEMENT	1.11	30	0.20 / 0.20	0.77	0.37	2 + 0	0.30; 0.49	+ 285/ + 285	+ 85/ + 85	NO	18
U-ELEMENT	1.12	27	0.24 / 0.26	2.17	0.43	2 + 0	0.34; 0.57	+ 804/ + 735	+ 79/ + 65	NO	20
U-ELEMENT	1.2	30	0.15 / 0.15	0.64	0.18	3 + 2	0.14; 0.24	+ 327/ + 327	+ 20/ + 20	YES	22
U-ELEMENT	2.1	28	0.17 / 0.16	3.52	0.26	3 + 3	0.20; 0.35	+1971/ +2100	+ 53/ + 63	NO	24
U-ELEMENT	2.2	28	0.12 / 0.14	0.41	0.18	2 + 2	0.14; 0.24	+ 242/ + 193	+ 50/ + 29	YES[3]	26
U-ELEMENT	2.3	27	0.23 / 0.23	0.71	0.37	2 + 2	0.29; 0.50	+ 209/ + 209	+ 61/ + 61	NO	28
Pu-ELEMENT	1.11	28	0.15 / 0.15	0.37	0.38	2 + 0	0.30; 0.50	+ 147/ + 147	+153/ +153	NO	30
Pu-ELEMENT	1.12	26	0.24 / 0.24	1.05	0.50	0 + 2	0.40; 0.67	+ 338/ + 338	+108/ +108	NO	32
Pu-ELEMENT	1.2	29	0.22 / 0.25	1.09	0.31	3 + 3	0.24; 0.42	+ 395/ + 336	+ 41/ + 24	YES[3]	34
Pu-ELEMENT	2.1	26	0.20 / 0.20	3.44	0.28	2 + 1	0.22; 0.38	+1620/ +1620	+ 40/ + 40	NO	36
Pu-ELEMENT	2.2	27	0.15 / 0.17	1.00	0.36	2 + 1	0.28; 0.48	+ 567/ + 488	+140/ +112	NO	38
Pu-ELEMENT	2.3	26	0.20 / 0.24	0.75	0.27	0 + 3	0.21; 0.37	+ 275/ + 213	+ 35/ + 13	YES[3]	40
ISOTOPE ABUNDANCE:											
U-234 ASSAY	1.11	27	2.30 / 1.92	10.0	4.77	1 + 0	3.80; 6.32	+ 335/ + 421	+107/ +148	NO	42
U-234 ASSAY	2.1	25	2.25 / 3.00	5.39	4.33	1 + 0	3.42; 5.81	+ 140/ + 80	+ 92/ + 44	NO	44
U-235 ASSAY	1.11	30	0.21 / 0.22	0.52	0.27	5 + 1	0.21; 0.36	+ 148/ + 136	+ 29/ + 23	YES	46
U-235 ASSAY	2.1	28	0.19 / 0.18	0.88	0.16	3 + 3	0.13; 0.22	+ 363/ + 389	- 16/ - 11	YES	48
U-236 ASSAY	1.11	30	0.45 / 0.43	1.71	0.47	1 + 2	0.38; 0.62	+ 280/ + 298	+ 4/ + 9	YES	50
U-236 ASSAY	2.1	25	4.48 / 3.98	6.61	4.67	1 + 1	3.67; 6.31	+ 48/ + 66	+ 4/ + 17	YES	52
Pu-238 ASSAY	1.11	9	1.59 / 1.45	3.75	1.50	1 + 0	1.02; 2.65	+ 136/ + 159	- 6/ + 3	YES	56
Pu-238 ASSAY	2.1	9	1.56 / 1.47	6.35	1.21	1 + 1	0.81; 2.24	+ 307/ + 332	- 22/ - 18	YES	60
Pu-239 ASSAY	1.11	29	0.032/ 0.036	0.12	0.035	1 + 2	0.028;0.046	+ 275/ + 233	+ 9/ - 3	YES	62
Pu-239 ASSAY	2.1	27	0.024/ 0.024	0.045	0.045	0 + 0	0.036;0.059	+ 88/ + 88	+ 88/ + 88	NO	64
Pu-240 ASSAY	1.11	29	0.068/ 0.072	0.25	0.097	1 + 1	0.078;0.13	+ 268/ + 247	+ 43/ + 35	NO	66
Pu-240 ASSAY	2.1	27	0.060/ 0.060	0.15	0.15	0 + 0	0.12; 0.20	+ 150/ + 150	+150/ +150	NO	68
Pu-241 ASSAY	1.11	29	0.21 / 0.21	0.59	0.18	0 + 4	0.14; 0.24	+ 181/ + 181	- 14/ - 14	YES	70
Pu-241 ASSAY	2.1	27	0.23 / 0.24	0.40	0.40	0 + 0	0.32; 0.53	+ 74/ + 67	+ 74/ + 67	NO	72
Pu-242 ASSAY	1.11	29	0.36 / 0.39	1.75	0.95	1 + 0	0.76; 1.25	+ 386/ + 349	+164/ +144	NO	74
Pu-242 ASSAY	2.1	27	0.32 / 0.36	0.86	0.79	2 + 0	0.63; 1.05	+ 169/ + 139	+147/ +119	NO	76
TOTAL:		739				42 + 34 = 76 (10.3%)		56+; 0-	48+; 8-	12 16	

[1] FIGURES CORRESPOND TO THE OFFICIAL IDA-80 EVALUATION.

[2] THE NUMBER OF EXCLUDED LABORATORIES IS SPLIT WITH RESPECT TO THE REASON FOR REJECTION: FIGURES REFER TO LABORATORIES EXCLUDED FOR EXTREME MEAN VALUES (BARTSCH CRITERION) AND FOR EXCEPTIONALLY HIGH REPEATABILITY VALUES (DIXON CRITERION).

[3] ONLY VALID FOR DoDM.

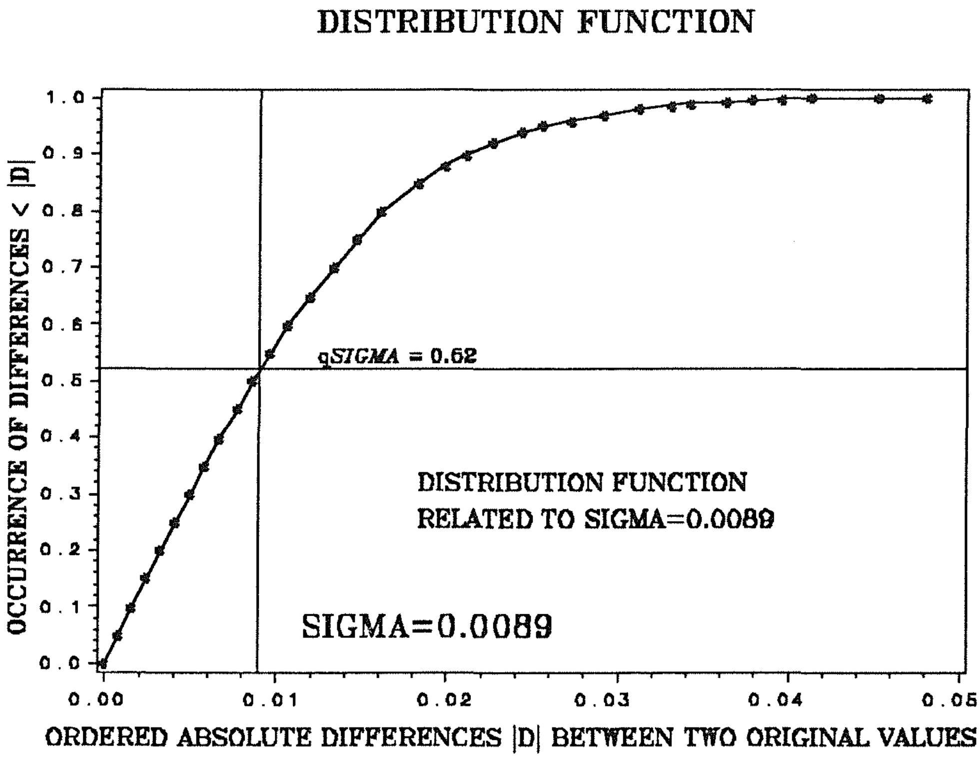

Fig.1: EXAMPLE OF THE DISTRIBUTION FUNCTION H(z) RELATED TO ABSOLUTE DIFFERENCES; σ = 0.0089

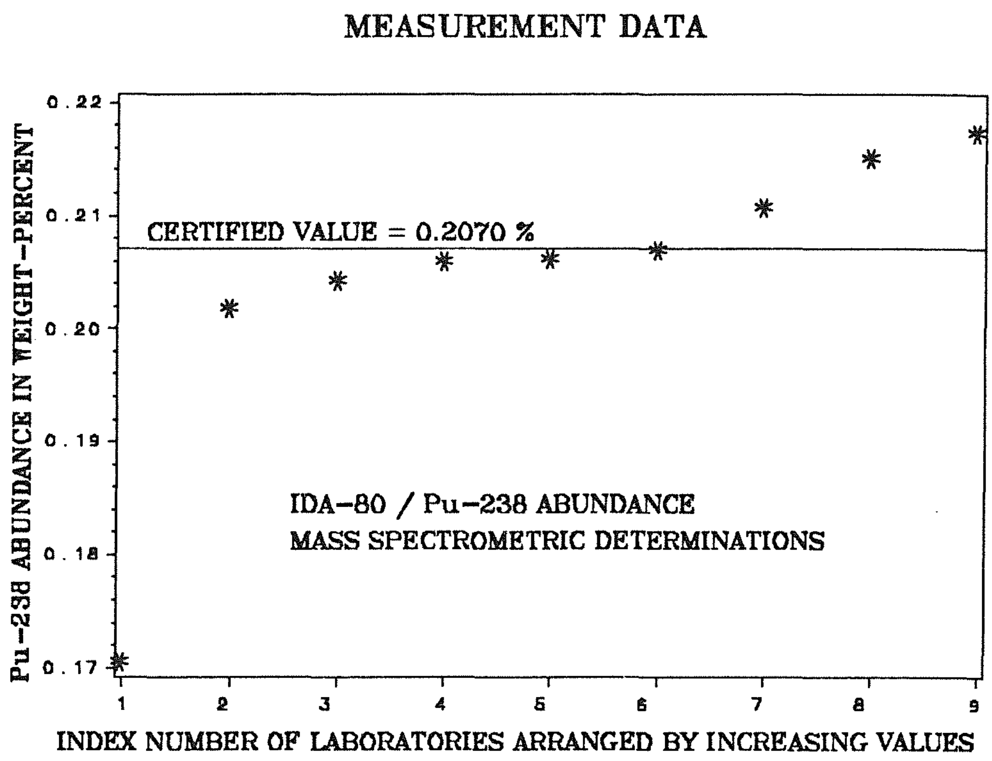

Fig.2: MASS-SPECTROMETRIC DETERMINATION OF ^{238}Pu ABUNDANCE IN IDA-80; MEASUREMENT DATA

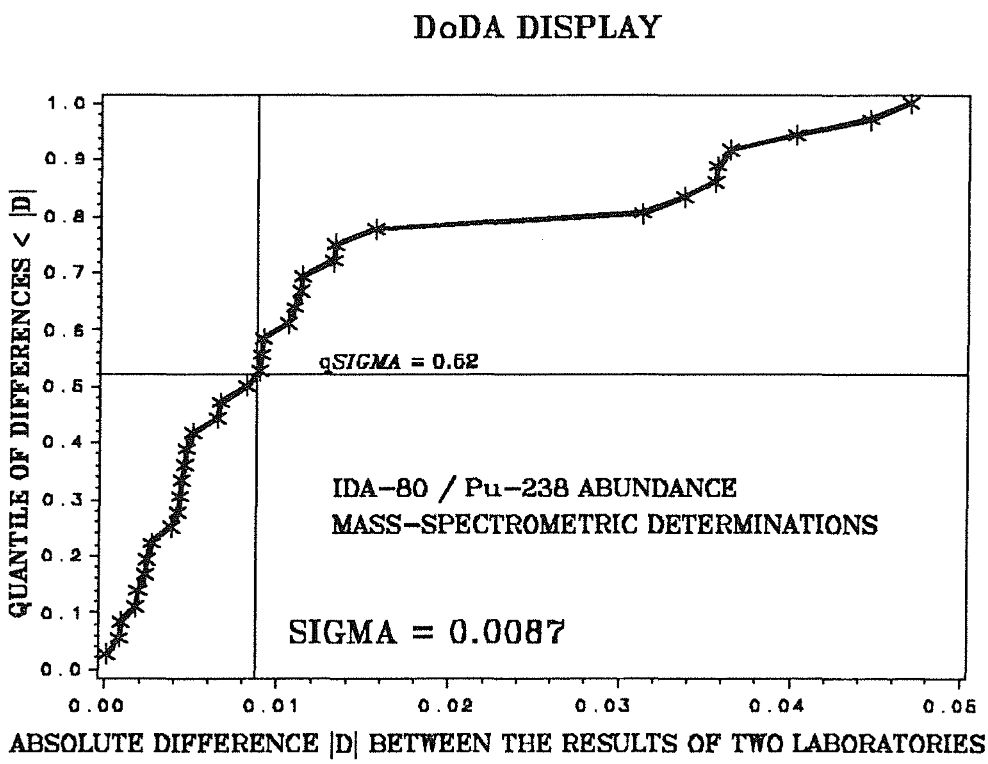

Fig.3: MASS-SPECTROMETRIC DETERMINATION OF ^{238}Pu ABUNDANCE IN IDA-80; DoDA DISPLAY

CALIBRATION DATA

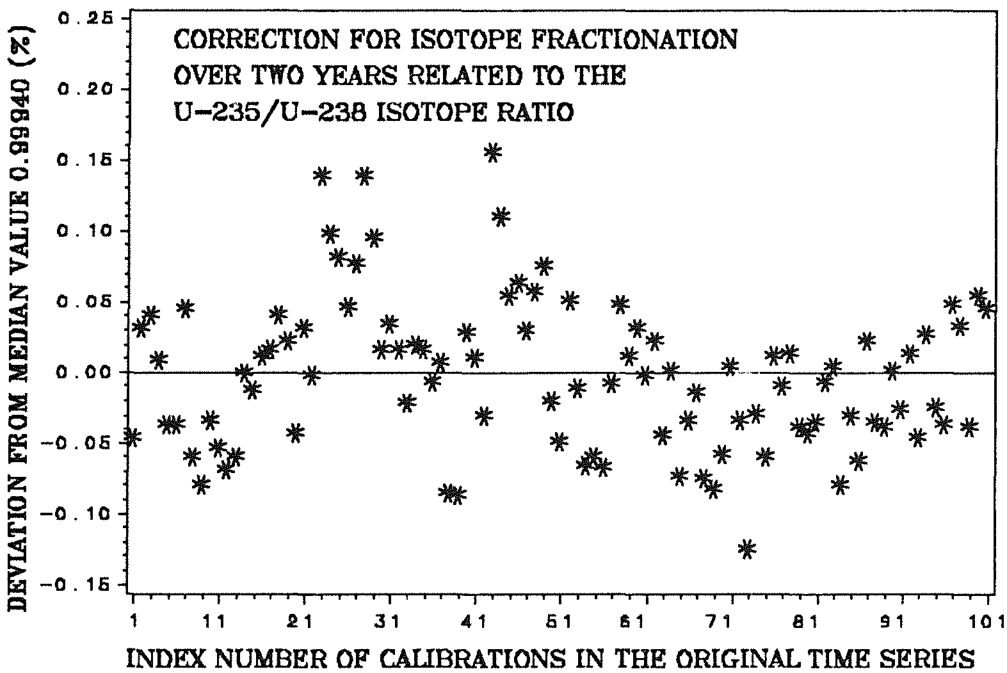

Fig.4: DETERMINATION OF THE CORRECTION FACTOR FOR ISOTOPE FRACTIONATION AT CBNM OVER TWO YEARS RELATED TO THE $^{235}U/^{238}U$ ISOTOPE RATIO IN THE ORIGINAL TIME SERIES; CALIBRATION DATA

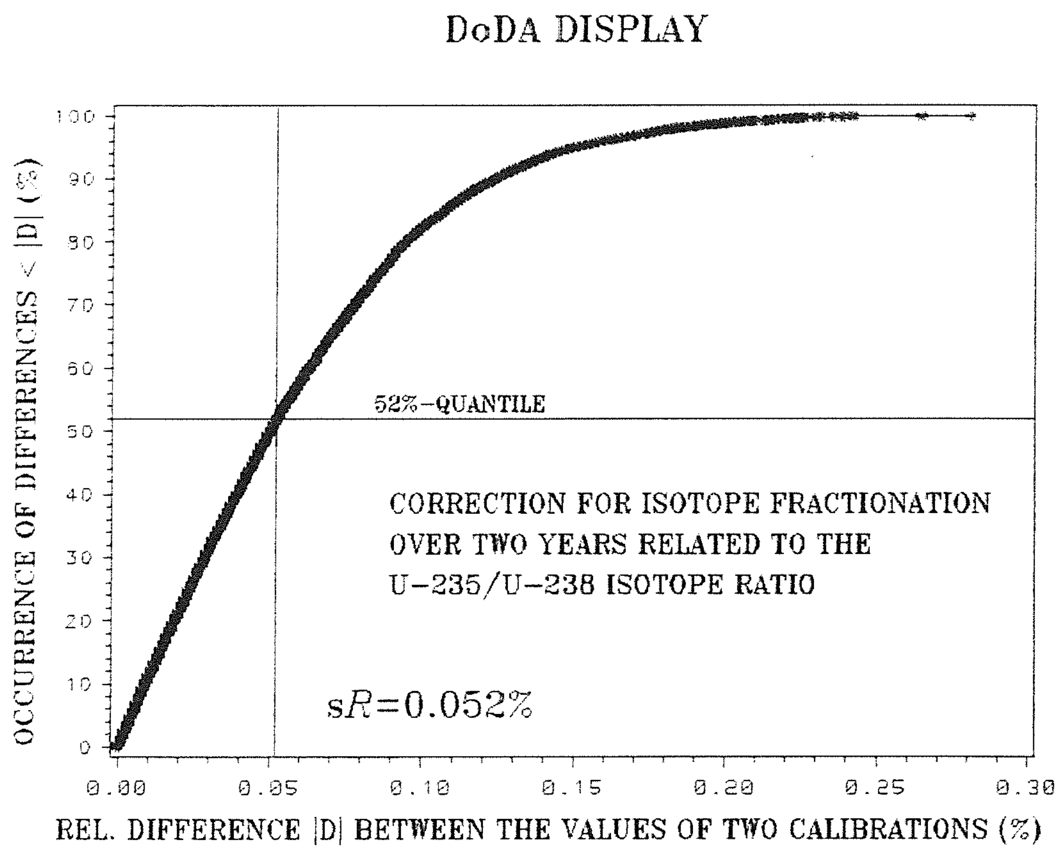

Fig.5: DETERMINATION OF THE CORRECTION FACTOR FOR ISOTOPE FRACTIONATION AT CBNM OVER TWO YEARS RELATED TO THE $^{235}U/^{238}U$ ISOTOPE RATIO IN THE ORIGINAL TIME SERIES OF CALIBRATION; CALIBRATION DATA