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Institut für Angewandte Reaktorphysik

Analysis of Some Available Data on Material Unaccounted For (MUF)

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Zusammenfassung

Mehr als 200 in der Literatur veröffentlichte Werte über MUF (bei der Bilanzierung unerfaßte Mengen an spaltbarem Material) in verschiedenen Kernanlagen wurden darauf untersucht, ob sie a) allein durch systematische Meßfehler erklärbar sind, b) einer statistischen Verteilung zugeordnet werden können und c) ob die normalisierten oder absoluten Werte des MUF für Überwachungszwecke besser geeignet sind.

Die Analyse zeigt, daß unter Berücksichtigung der zugrunde gelegten Bedingungen ein großer Teil der MUF-Daten durch systematische Meßfehler erklärbar ist, jedoch kann auch eine Vielzahl anderer Faktoren zufälliger sowie systematischer Natur zu den MUF-Daten beitragen. Anlagenspezifische Daten können einer Normalverteilung gehorchen. Im Hinblick auf die Gesamteingangsmenge normalisierte MUF-Werte scheinen besser als absolute Werte für die Überwachung geeignet zu sein. Eine ständige Analyse der unter wohldefinierten Bedingungen gewonnenen MUF-Werte ist notwendig, um diese Daten in geeigneter Weise verwenden zu können.

Abstract

More than 200 values for material unaccounted for (MUF) in different nuclear facilities, published in the literature, have been analysed with a view to determine whether a) they can be explained by systematic errors of measurements alone b) they follow any known statistical distribution and c) the absolute or the normalized values of MUF are better safeguards indicators.

The analysis shows that within the restrains stipulated in the paper, a major part of the MUF data can be explained by systematic errors of measurements but a large number of other random and non-random factors contribute also to the MUF values. Facility specific MUF values may show a normal distribution. Normalized MUF values with respect to the total input appear to be a better safeguards indicator than the absolute values. Continuous analysis of MUF values obtained under controlled conditions is essential for the proper use of these values.

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1. Introduction

It is now a well established fact that material balance accountancy complemented by containment and surveillance will be the three basic measures for any safeguards system. They are also of importance for establishing an internal material accountancy system for a facility. Of these three basic measures, the material balance accountancy permits quantitative safeguards statements in case of a probable diversion. In this connection the most important factor which influences such a statement is the MUF (Material Unaccounted For) defined as the difference between book and the physical inventory [13], after a material balance for a material balance area over a given period of time has been established.

In those cases where no measurement errors and no process losses occur in establishing a material balance (e.g. digital accountancy for fuel elements in a light water type reactor), the value of the MUF is zero under diversion free conditions. However, in those nuclear facilities in which fissionable materials are measured by chemical or other methods, the measured amounts are associated with measurement errors so that the difference between the book inventory and the physical inventory is seldom zero. Besides, unknown process losses or hidden inventory in the plant may also cause the MUF to have a different value than zero. Evidently, on top of all these possibilities a

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diversion will make the value of MUF different from zero as well. Therefore, the material balance accountancy provides no doubt the only means of making a quantitative statement in the event of a diversion, for those nuclear facilities in which measurements are directly made on fissionable materials, the inspection organisation has to have an idea on the characteristics of the MUF under diversion free conditions.

As indicated above, the components of the MUF may be regarded to be composed of two factors one being the unmeasured quantities and the others being the measurement errors. The unmeasured quantities may consist of the unmeasured process losses which leave the facility and the hidden process inventories remaining in the facility. Both these quantities may have a systematic and a random component. The measurement errors are also composed of a random part and a systematic part. Therefore the values of MUF are expected to have a mean value and a variance. The variance is expected to be caused mainly by the random fluctuations of the process losses and the systematic errors of measurement as the random part of the measurement errors may be reduced significantly over a large number of measurements.

In the present paper published values on MUF from different facilities have been analysed with regard to the following areas of interest:

a) Can the MUF values be explained by the known systematic errors of measurement methods alone which are normally used in those facilities in which the MUF values are generated.

b) Do the MUF values follow any definite distribution e.g. normal distribution.

c) Are the absolute or the normalized values of MUF better safeguards indicators.

2. Input Data

2.1 Measurement errors

Many published values are available on systematic errors. It is beyond the scope of this report to list all of them. For the purpose of the present investigations two representative sets of values for systematic errors [2, 3] for uranium and plutonium measurement methods in a reprocessing plant have been used and reproduced in Table 1. For facilities other than chemical reprocessing, systematic errors corresponding to the product stream of chemical reprocessing plants have been used for the feed and the product streams. Although this assumption may not always be justified, it appears to be quite correct for the purpose of the present study. Whenever isotopic compositions are involved an additional systematic error of 0.3 % [1] has been considered. The values in [3] have been used for analysis in this report as they appear to be representative of actual present-day plant operating conditions. The values in [2] were obtained in the course of an interlabtest and particularly the values for the input stream of a reprocessing plant may change when large amounts of data become available. The sum of the total systematic errors (Table 1) for a given facility and material is calculated by taking the square root of the sum of the squares of the individual relative standard deviations for the feed and the product streams. Thus the systematic errors are assumed to be independent of each other and are described by variance and not as a bias.

2.2 MUF values

A total of 241 MUF values (M_i) could be obtained from nine different sources [1,4,5,6,7,8,9,10,11]. They are listed in Table 2. For the purpose of analyses these values were grouped in the following manner:

2.2.1 Values of MUF (M_i), normalized with respect to the feed or input (beginning inventory + receipts or one half of the total flow) for four specific facilities [4,5,6,7,10]. They are presented in Tables 3A,3B,3C and 3D. The numbers of M_i values vary between 20 - 30. The absolute amounts and the time sequence of the MUF values are not known.

2.2.2 Values of MUF (M_i) normalized to input for a group of facilities (total number 126). The types of facilities as well as the absolute values and the time sequence of the M_i are not known. The values are summarized in Table 4 [5,11].

2.2.3 Values of MUF (M_{is}) normalized to <u>both</u> the input and the systematic measurement error component. They include all the values from Table 3 except those from [5] and some additional values from Table 2, for which the type of facility in which the MUF values generated was known. The M_{is} values, 89 in number are listed in Table 5. These values were obtained by dividing the M_i values by the corresponding systematic errors (US values) of Table 1. In case of M_i values for isotopes an additional systematic error of 0.3 % was assumed.

2.2.4 30 absolute values of MUF in arbitrary units for a reprocessing facility. The time and feed sequence of these values are known [4]. The relevant data are presented in Tables 7, 8 and 9.

3. Influence of the Systematic Part of the Measurement Errors on MUF

Since the random part of any measurement errors reduces rapidly with increasing number of measurements, it is justifiable to analyse the influence of the systematic part of the measurement errors alone on the MUF Values. The systematic part may consist of a bias and a random part which is described by a relative standard deviation δ. The calibration error in a measurement method is a typical example of the latter. The standard deviations for the systematic errors are normally caused by a fairly large number of complex factors which may have different types of statistical distributions. The resulting distribution of the relative systematic error may therefore, be assumed to be normal. In case about 95 % of the M_i values (i.e. individual MUF values normalized with respect to the input) were to lie within the $\pm$ 1.96 δ range of the corresponding relative standard deviations of the systematic error, they could then be assumed to be explained entirely by the systematic errors only, with the error first kind (α error) of 5 %. For this analysis the results of Table 5 are of interest. Here only those normalized MUF values (M_i) which were known to be generated in a definite type of facility (so that the corresponding values of systematic errors could be allocated to them) could be considered. Each of these values were divided by the corresponding relative s standard deviation value δ for systematic errors as given in [3]. These values are expected to correspond more closely to the plant operating conditions. If the result of the double normalization (i.e. M_{is} values) is found to be greater than $\pm$ 1.96, it can be considered to be an outlier. The resulting values are arranged in the decreasing order of their magnitude. It is to be noted that out of a total of 89 values 19 values, i.e. 21 % of the total are outliers. Besides, the mean value of these M_{is} is positive and not zero.

These results indicate a number of interesting points:

a) Although about 80 % of the MUF values can be explained by the systematic errors alone - and this is a very large number - the relatively large number of outliers seems to indicate that some other components are also contributing to these values.

b) A positive value of the mean of the double normalized M_{is} values show that there is either a bias in the systematic error component or an unknown process loss, or both.

c) Since the unknown process losses may also vary randomly and the variations may be caused by complex contributions of a number of factors having different distributions . These unknown process losses may also have a normal distribution, as has been assumed to be the case with the systematic errors. In that case, the M_i values may be extected to be normally distributed with a standard deviation which may be equal to or larger than that for the systematic errors.

4. Test for Normal Distribution of Normalized MUF (M_i) Values

An analysis of the results of Table 5 shows, as indicated above, that further investigations of the MUF values on whether they are normally distributed or not, are worthwhile. The K-statistics [12] has been used in this report to test the normality of the values. The basic structure of the test is given in Annex 1.

4.1 Restrictions

It is to be noted that statistically significant amount of data (i.e. at least 50 for a given sample) is required to carry out a test for normality with k-statistics. Since the numbers of MUF values particularly for single facilities (Tables 3A, 3B, 3C and 3D) are fairly small, the results of the test should only be considered as indicating trends instead of definite proofs.

Another point to note in this connection is that the g_1 and g_2 values for the test (see Annex 1) are available in the literature [12] for down to a total number of sample values n of 50 and 100 respectively. Therefore, for the normality test in the present report, because of smaller sample numbers, two other ratios namely

$$k_3/\sigma_{k_3} \text{ and } k_4/\sigma_{k_4}$$

have been used. The first ratio is the measure of the skewness (asymmetry) and the second one, the measure of kurtosis. The distribution of a random population of numbers may be considered to be approximately normal if the value of each of these two ratios is found to be less than 2.

4.2 Test results

All the different categories of normalized MUF values as listed under 2.2.1, 2.2.2 and 2.2.3, and presented in Tables 2, 3A-3D, 4 and 5 were tested for normality. The results of the test are summarized in Table 6. A number of points are worth mentioning:

a) All the M_i values (Table 2) when tested together, do not show normality. However, the normality improves significantly if the highest two positive values of MUF (5.86,5.74) are rejected. This rejection may be justified as the values lie beyond the ± 2 σ range of the resulting distribution. By the rejection of only these two values the distribution becomes completely symmetrical (the skewness factor is reduced from 9.24 to + 0.03) and the kurtosis is reduced by a factor of about 3 (from 30.41 to 11.44). However, the distribution cannot be considered as normal because of the high value of the kurtosis. The results are illustrated by the histogram of Fig. 1.

b) The M_i values for all the four individual facilities (Tables 3A, 3B, 3C and 3D) are normal distributed (with the restriction of 4.1). However, when the values are combined together, the resulting distribution can no longer be considered as normal. Here also only the kurtosis causes the abnormality. Fig. 2 gives the histrograms of the results of these four tables as well as the histogram of the combined MUF values.

c) The MUF values for the group of facilities (small number per facility) listed in Table 4 do not show a normal distribution. The data in this table differ from those in Table 2 mainly by the absence of the facility specific M_i values. The distribution shows the same type of trends as for the Table 2 data. The skewness is reduced almost to zero by the elimination of the highest two values, the kurtosis remains.

d) The double normalized M_{is} values in Table 5 (normalized both with respect to input and to the relative standard deviations for the corresponding facilities) do not show a normal distribution. The difference between the values in this table and the sum of the values in Tables 3A, B, C, and D lies mainly in the absence of the 26 values of Table 3B [5] for which the type of facility is not known. A comparison of the test results for these two groups of values (columns 7 and 8 in Table 6) indicates that an additional normalization with respect to systematic errors causes an improvement in the normality of the distribution (skewness improved from - 0.2 to + 0.05 and kurtosis improved from 6.46 to 4.57) for MUF values for groups of facilities.

5. Absolute vs Normalized MUF values

Although most of the MUF values (211 out of 241) considered in this study were available as normalized values i.e. as percentage of feed or input, some 30 values from [4] were also available in absolute units. They provided an oppertunity to test the suitability of the absolute MUF values for safeguards purposes. Here again a world of caution is necessary. The number of MUF values available is small. Therefore the results of the analyses can be regarded as indicating some trends only.

The absolute and the normalized (with respect to feed) values of MUF for this facility were tested for normality. The results are summarized in Table 7A. The histograms of the distributions with absolute and normalized MUF values are given in Fig. 3. Since the time sequence of the reprocessing campaigns, for which the MUF values were available, was also known, the mean values and the corresponding standard deviations for the absolute MUF values for a number of sequentially overlapping accounting periods were estimated and are presented in Table 8. For comparison, the mean value and the standard deviations for the corresponding normalized MUF values (M_i) were also calculated and are presented in Table 8A. A similar set of data for the absolute and the relative MUF values for the same sequentially overlapping accounting periods for increasing values of feed, was also calculated and are presented in Tables 9 and 9A respectively. The results of Tables 8 and 8A and those of 9 and 9A are presented graphically in Fig. 4.

An analysis of these results tends to indicate the following:

a) Both the absolute and the normalized values for the reprocessing facility appear to be normally distributed. However, the normalized values seem to be significantly more normal than the absolute values (see Fig. 3). The distribution for these values show a much better symmetry (g_1 = 0.019 against 0.54; Table 7A).

b) The mean and the standard deviations show a much wider spread for the absolute MUF values for both the time and the feed sequences. Those for the relative values are more stable and show a fairly narrow band of scatter.

c) The numbers of outliers for absolute MUF values for the time and the feed sequences are fairly large. They are reduced for relative MUF values, and are almost independent of the sequential periods chosen.

6. Discussion of Results

At the beginning it should again be emphasized that because of the limited amount of data available, the results may only show some trends in the behavior of the MUF data. Some discussions of the results are attempted in the framework of this limitation.

6.1 Systematic errors and normalized MUF

An analysis of the results in Table 5 showed that about 80 % of the 89 normalized MUF (relative to feed or input) values could be explained by the systematic errors for the corresponding facility. The systematic error has been assumed to vary randomly for the MUF value generated in different campaigns. This appears to be quite often the case. Use of consumable measuring equipment, calibration of measurement units from campaign to campaign, use of different measurement standards, may all cause the systematic error associated with these events to vary randomly from campaign to campaign. The fact that such a large fraction of the analysed relative MUF data can be explained by such systematic errors alone, is in itself fairly important. This would mean firstly, that the systematic errors make a major contribution to the components of MUF and secondly, that the relative MUF values will be a more suitable safeguards indicator than the absolute values of MUF.

Another highly significant fact is that about 80 % of the 241 relative MUF values analysed are positive and the mean value is positive and not zero. This would tend to indicate that there is a continuous unknown loss of material (or hidden inventory) in the facilities in which the MUF data were generated. Although such a bias may also be caused by an unknown bias in the measurement methods, this appears to be less probable since it would mean that all the measurement methods in all these different types of facilities would have a bias in the same direction. The positive mean bias in the MUF values becomes difficult to detect separately as in most of the cases it is considerably smaller than the standard deviation.

It is also important to note that the mean values of the relative MUF tend to remain stable over a wide range of normalized values. This would mean that the bias (caused probably by process losses) is also dependent on feed. This is another reason for using the normalized MUF values for safeguards purposes.

6.2 Distribution and components of normalized MUF values

6.2.1 Distribution

The results of Table 6 indicate that all facility specific normalized MUF values are normally distributed with a positive mean value and a standard deviation which is larger than the mean and than that for the systematic error alone (excepting one case for U-235 facility, Table 3C). For combined values from different groups of facilities, the same trend can be noted i.e. the mean values are less than the standard deviation, but the normality of the distribution is destroyed. The only reason for the non-normal distribution may be the insignificant amount of the data analysed and real presence of contamination. In order to illustrate the sensitivity of the test, the values of k_3, k_4 and their variance are also included in Table 6. At this point some further discussion on the different components of MUF appears to be relevant.

6.2.2 Components of MUF

It was postulated at the beginning that the MUF may consist of two basic components namely, the unknown losses and the measurement errors. The unknown losses have to be specific of the process i.e. of the type of the facility, whereas, the measurement errors should depend on the measurement methods used. Both these components may have a systematic and a random part and may or may not be dependent on the feed or the input. Some typical examples of these possibilities may be considered.

a) The reproducibility of any measurement method is the random part of the measurement error for this method. In a recent interlabtest [2] the reproducibility of the measurement method for uranium concentration in the product stream of a reprocessing plant was found to have a relative standard deviation of 0.11 % By n repetitions of measurements by the same method, the contribution of this part of the error is reduced by 0.11 $\%/\sqrt{n}$. Since in a normal campaign, fairly large number of measurements by the same method are expected to be made, the contribution of the random part of the measuring method to the MUF values becomes small. However, for those small campaigns, this contribution may not be negligible.

b) The calibration errors for a measurement method used in a particular facility is an example of the systematic part of the measurement error. So long as the same calibration curves are used, the error propagation for this type of error follows a linear rule. However, the values of the calibration curves for the same method varies from facility to facility and from campaign to campaign in a random fashion. The variance for such systematic errors may be estimated with the help of results obtained from a suitably constructed interlabtest. They are expected to be normally distributed with a mean value of zero. The variance for the systematic part of the measurement method for uranium concentration mentioned before, was found to be $\pm$ 0.2 % during the same interlabtest [2]. Contribution of this part of the error to the MUF values is not reduced by repeated measurements as it propagates linearly. This part of the error is dependent on the amount of feed.

The systematic component of the errors may also have a bias (e.g. the measurement errors associated with an analytical standard). This can be determined only with difficulty. However, the influence of such errors can be eliminated by using the same measurement methods (with the same instruments) for the input and the output streams. A bias can be independent of the measured amounts e.g. the absolute value of bias in a weighing balance, which remains constant over a wide range of weighed amounts.

c) A part of process losses or hidden inventories may be independent of the feed or input amount, for example, those losses caused by the deposition of fissile materials on the surface of process equipment because of corrosion or unavoidable reactions. The amounts remaining in filters, centrifuges etc. may also be independent of the input. Any accidental losses, retentions of process material in one campaign and reworking and the production of the same material in some other subsequent campaigns, are also examples of process losses which may be independent of the input. Depending on the process conditions, such losses may have a small or a large random component.

Some other types of process losses may depend on the amounts of feed treated in a campaign. Examples of this type are unmeasured process losses which leave the facility during a campaign, write offs which may be made of the amounts estimated only by difference and so on. Since the feed itself may be randomly distributed (e.g. batch-to-batch variations) or the measured values are generated randomly, the feed dependent process losses have also a random component. Besides, the losses themselves may also vary in a random manner.

6.2.3 Relation between distribution and components of MUF

Coming back once more to the distribution it may be remembered that the mean of the normalized MUF values were found to be fairly stable and varied in the range of 0.15-0.5 or in the range of 0.15-0.4 when the highest two values in Tables 2 and 4 were rejected. It is evident that such stability could not be caused by process losses which were independent of feed, since the feed for different campaigns in different facilities considered in this report, varied over a very wide range. Therefore, the contribution must have been from the process losses which are feed dependent and may or may not leave the facility during or after the campaign. It was mentioned earlier under 6.1 that the bias in measurement error could in all probability not contribute to the positive mean values of the normalized MUF.

It is however, difficult to determine the components from which the additional relative standard deviations for the normal distributions have been contributed. The absolute values of these relative standard deviations are in the range of 0.2-1.25 %. It may either be from the throughput-dependent or throughput-independent process losses. A remote possibility of this contribution may be from the random component ofthe measurement errors also. If the total integral amounts of feed during the campaigns considered here were large, the relative standard deviations caused by the feed-independent process losses would be small in comparison and would be completely masked by the relative standard deviations caused by the feed-dependent process losses. The influence of the former would also be reduced over a longer period of plant operations. Both the larger amounts of feed and the longer period of operation would tend to reduce the influence of the random part of the measurement errors also. Only for small amounts of feed over short periods of time can the relative standard deviations of feed-independent process losses make an important contribution to the standard deviation of the MUF values.

6.3 Absolute vs normalized MUF values

The distribution of the absolute and the normalized MUF values for a reprocessing plant [4], as well as the means (μ) and relative standard deviations (σ) of these values for a number of sequentially overlapping accounting periods were analysed in chapter 5. There was a clear indication that for the set of data considered, the normalized values of MUF (M_i) are better suited for safeguards purposes. Since both the μ and the σ values are much more stable for M_i values, they can be used for predicting the M_i values for future campaigns with different amounts of feed, than the ones for which the M_i values are available. The M_i values are much more normally distributed than the absolute MUF values. As discussed earlier, this is explainable because of the fact that both the μ and the σ of the distributions are determined mainly by the feed (or input). This does not mean however, that the actual MUF and the feed values should be correlated in a statistical sense (linear correlation). Because of the randomness of the MUF values no linear correlations can normally be established.

7. Conclusions

The present report is an effort to analyse the published data on MUF for different nuclear facilities. From the point of view of safeguards, a knowledge on the origin and behavior of MUF values is necessary before any quantifiable statement on the MUF values can be made.

241 published values of MUF normalized with respect to feed or to input and 30 absolute MUF values were analysed in respect of three areas of interest namely, a) the influence of systematic errors, b) the probable distribution of the MUF values and c) the suitability of the absolute or the normalized values of MUF for safeguards purposes.

Although in most of the cases the amounts of data analysed were relatively small, a number of generalized conclusions subject to the restrictions indicated in the report appears to be possible. They are summarized below:

7.1 About 80 % of the available MUF data from different facilities could be explained by the corresponding systematic errors of measurement alone which were assumed to be normally distributed. Although this is not the only component contributing to the MUF values, in the absence of any other data, and when the systematic components of errors for measurement methods are known, they can be used as a starting basis for predicting the behavior of MUF values for similar facilities using the same type of material and the same measurement methods. They can also be used as a quantifiable basis for making statements on MUF values.

7.2 The normalized MUF values for a given facility may be expected to have a normal distribution with a positive mean value and a relative standard deviation which may be larger than that assumed for the systematic error. Such a normal distribution may therefore, form a quantifiable basis for the preparation of decision models with the help of which statements on the MUF values may be made by a safeguards authority.

The distribution of MUF values from groups of facilities was not found to be normal.

7.3 The mean values of all the different categories of MUF data were found to be positive varying within a fairly narrow range of 0.15-0.4. They indicate to an unknown feed dependent process loss. The broadening of the relative standard deviation (which in most cases was much larger than the mean) of the resulting normal distribution of MUF may also be caused by the variance of this feed-dependent process loss.

7.4 The normalized values of MUF appear to be better suited for safeguards purposes than the absolute values. For a reprocessing facility, the mean and the relative standard deviations for these values for a number of sequentially overlapping accounting periods, showed a much more stable trend than those for the absolute values.

7.5 It is absolutely essential to collect and analyse MUF values for different nuclear facilities in a systematic manner continuously and on a world wide basis. The data analysed in this report indicate that the normalized values of MUF may be expected to behave in a predictable manner.

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Table 1: Systematic Errors for a Chemical Reprocessing Plant

Error description	Relative standard deviation $\delta^{+)}$ in %			
	Values from an interlabtest /2/		US values /3/	
	Plutonium	Uranium	Plutonium	Uranium
Total systematic error for feed	2.7	1.4	0.44	0.41
Total systematic error for products	0.25	0.2	0.37	0.24

$^{+)}$the component of systematic error for isotopic analysis is taken as 0.3 % /1/

Table 2: A List of All the Normalized MUF+) Values (M_i) Used in this Paper

Serial No.	M_i	Material	Ref.	Serial No.	M_i	Material	Ref.	Serial No.	M_i	Material	Ref.
1	-0.17	-	5	46	0.07	-	5	91	0.11	-	5
2	0.18	-	5	47	0.11	-	5	92	2.44	-	5
3	1.18	-	5	48	0.13	-	5	93	0.05	-	5
4	-0.41	-	5	49	0.21	-	5	94	0.06	-	5
5	0.22	-	5	50	0.31	-	5	95	0.53	-	5
6	0.10	-	5	51	0.78	-	5	96	-0.06	-	5
7	-0.09	-	5	52	1.00	-	5	97	0.35	-	5
8	0.12	-	5	53	0.64	-	5	98	-0.02	-	5
9	-0.02	-	5	54	0.94	-	5	99	2.14	-	5
10	0.09	-	5	55	1.23	-	5	100	0.92	-	5
11	1.16	-	5	56	1.14	-	5	101	-0.03	-	5
12	-0.13	-	5	57	0.07	-	5	102	0.06	-	5
13	0.42	-	5	58	2.44	-	5	103	0.19	-	5
14	1.52	-	5	59	1.24	-	5	104	0.15	-	5
15	1.35	-	5	60	1.18	-	5	105	0.02	-	5
16	0.07	-	5	61	0.01	-	5	106	0.10	-	5
17	0.34	-	5	62	1.78	-	5	107	0.36	-	5
18	0.69	-	5	63	-0.03	-	5	108	-0.62	-	5
19	0.54	-	5	64	0.07	-	5	109	0.23	-	5
20	1.20	-	5	65	0.63	-	5	110	0.08	-	5
21	0.17	-	5	66	0.11	-	5	111	0.08	-	5
22	0.21	-	5	67	0.01	-	5	112	0.08	-	5
23	0.01	-	5	68	0.15	-	5	113	-0.80	-	5
24	0.29	-	5	69	0.52	-	5	114	0.11	-	5
25	0.30	-	5	70	0.67	-	5	115	-0.19	-	5
26	-0.10	-	5	71	0.60	-	5	116	0.12	-	5
27	0.09	-	5	72	-3.22	-	5	117	0.02	-	5
28	0.82	-	5	[73]	5.74	-	5	118	0.27	-	5
29	0.30	-	5	74	0.01	-	5	119	-0.15	-	5
30	0.43	-	5	75	0.12	-	5	120	0.17	-	5
31	2.96	-	5	76	0.10	-	5	121	0.04	-	5
32	1.06	-	5	77	0.09	-	5	122	0.09	-	5
33	2.22	-	5	78	0.09	-	5	123	0.06	-	5
34	0.08	-	5	79	0.31	-	5	124	-0.06	-	5
35	0.39	-	5	80	0.61	-	5	125	-1.12	-	5
36	0.19	-	5	81	0.49	-	5	126	1.00	-	5
37	0.04	-	5	82	0.11	-	5	127	1.94	-	5
38	-0.06	-	5	83	0.03	-	5	128	0.65	-	5
39	0.13	-	5	[84]	5.86	-	5	129	1.30	-	5
40	0.13	-	5	85	1.62	-	5	130	1.38	-	5
41	-0.32	-	5	86	2.05	-	5	131	0.33	-	5
42	-0.36	-	5	87	0.12	-	5	132	0.46	-	5
43	0.35	-	5	88	-0.07	-	5	133	-1.23	-	5
44	-1.96	-	5	89	1.43	-	5	134	0.85	-	5
45	0.21	-	5	90	0.05	-	5	135	1.00	CR	4

Table 2 contd.

Serial No.	M_i	Material	Ref.	Serial No.	M_i	Material	Ref.	Serial No.	M_i	Material	Ref.
136	1.08	CR	4	186	0.39	Pu-239	7	236	1.40	Pu CR	8
137	1.30	CR	4	187	1.11	Pu-239	7	237	1.48	Pu-239+241 CR	8
138	0.45	CR	4	188	2.87	Pu-239	7	238	1.00	U CR	9
139	-2.63	CR	4	189	0.19	Pu-239	7	239	1.40	Pu CR	9
140	-1.08	CR	4	190	0.23	Pu-239	7	240	1.36	Pu-239+241 CR	9
141	-0.73	CR	4	191	0.51	Pu-239	7	241	0.44	U-235	1
142	0.22	CR	4	192	-0.18	Pu-239 CR	7				
143	-0.84	CR	4	193	0.41	Pu-239 CR	7				
144	0.31	CR	4	194	0.07	U-235	11				
145	0.83	CR	4	195	0.09	U-235	11				
146	1.41	CR	4	196	0.27	U-235	11				
147	-2.31	CR	4	197	1.80	U-235	11				
148	-1.04	CR	4	198	0.04	U-235	11				
149	0.74	CR	4	199	0.49	U-235	11				
150	-0.62	CR	4	200	0.49	U-235	11				
151	0.06	CR	4	201	0.94	U-235	11				
152	-0.59	CR	4	202	-0.05	U-235	11				
153	-1.04	CR	4	203	0.40	U-235	11				
154	-0.49	CR	4	204	-0.01	U-235	11				
155	0.32	CR	4	205	0.05	U-235	11				
156	0.62	CR	4	206	0.06	U-235	11				
157	0.90	CR	4	207	0.85	U-235	11				
158	0.93	CR	4	208	0.90	U-235	11				
159	3.34	CR	4	209	-0.38	Pu	11				
160	-0.46	CR	4	210	0.11	Pu	11				
161	0.95	CR	4	211	0.62	Pu	11				
162	-0.78	CR	4	212	0.10	Pu	6				
163	1.27	CR	4	213	0.22	Pu	6				
164	2.15	CR	4	214	-1.28	Pu	6				
165	-0.05	U-235	7	215	0.06	Pu	6				
166	0.21	U-235	7	216	-0.14	Pu	6				
167	0.04	U-235	7	217	-0.10	Pu	6				
168	0.01	U-235	7	218	0.10	Pu	6				
169	0.55	U-235	7	219	-1.36	Pu	6				
170	0.73	U-235	7	220	0.06	Pu	6				
171	0.65	U-235	7	221	1.64	Pu	6				
172	0.67	U-235	7	222	0.18	Pu	6				
173	0.18	U-235	7	223	0.08	Pu	6				
174	0.17	U-235	7	224	1.38	Pu CR	6				
175	0.16	U-235	7	225	-1.34	Pu CR	6				
176	0.02	U-235	7	226	1.34	Pu CR	6				
177	0.06	U-235	7	227	0.06	Pu CR	6				
178	0.06	U-235	7	228	0.16	U-235	10				
179	0.30	U-235	7	229	0.09	U-235	10				
180	0.25	U-235	7	230	0.07	U-235	10				
181	0.44	U-235	7	231	0.06	U-235	10				
182	-0.09	U-235 CR	7	232	0.07	U-235	10				
183	0.37	U-235 CR	7	233	0.25	U-235	10				
184	0.47	Pu-239	7	234	0.24	U-235	10				
185	0.29	Pu-239	7	235	0.12	U CR	8				

+) Numbers from ref 5,7,10 and 11 are normalized with respect to beginning inventory plus receipts, from reference 4 the normalizing factor is feed and from rest of the references it is half of the total flow.

CR represents a chemical reprocessing plant.

The two values in bracket are the highest ones.

Mean value (μ): + 0.38

Standard deviation (σ): $\pm$ 0.95

Table 3A: Normalized MUF (M_i) Values for a Reprocessing Facility [4]. (Normalized with Respect to Feed)

Serial No.	M_i (% of feed)	Serial No.	M_i (% of feed)	Serial No.	M_i (% of feed)
1	3.34	11	0.83	21	- 0.59
2	2.15	12	0.74	22	- 0.62
3	1.41	13	0.62	23	- 0.73
4	1.30	14	0.45	24	- 0.78
5	1.27	15	0.32	25	- 0.84
6	1.08	16	0.31	26	- 1.04
7	1.00	17	0.22	27	- 1.04
8	0.95	18	0.06	28	- 1.08
9	0.93	19	- 0.46	29	- 2.31
10	0.90	20	- 0.49	30	- 2.63

Mean value (μ): + 0.18

Standard deviation (σ): $\pm$ 1.25

Table 3B: Normalized MUF (M_i) Values for a Single Facility [5].
(Normalized with Respect to Beginning Inventory and Receipts.)

Serial No.	M_i (% of Input)	Serial No.	M_i (% of Input)	Serial No.	M_i (% of Input)
1	1.94	10	0.23	19	0.04
2	1.38	11	0.17	20	0.02
3	1.30	12	0.12	21	- 0.06
4	1.00	13	0.11	22	- 0.15
5	0.85	14	0.09	23	- 0.19
6	0.65	15	0.08	24	- 0.80
7	0.46	16	0.08	25	- 1.12
8	0.33	17	0.08	26	- 1.23
9	0.27	18	0.06		

Mean value (μ): + 0.22
Standard deviation (σ): ± 0.70

Table 3C: Normalized MUF (M_i) Values for Facilities Handling U-235, other than Reprocessing Plants [1,7,10]. (Normalized with Respect to Input)

Serial No.	M_i (% of Input)	Serial No.	M_i (% of Input)	Serial No.	M_i (% of Input)
1	0.73	10	0.24	19	0.06
2	0.67	11	0.21	20	0.06
3	0.65	12	0.18	21	0.06
4	0.55	13	0.17	22	0.04
5	0.44	14	0.16	23	0.02
6	0.44	15	0.16	24	0.01
7	0.30	16	0.09	25	- 0.05
8	0.25	17	0.07		
9	0.25	18	0.07		

Mean value (μ): + 0.23

Standard deviation (σ): $\pm$ 0.22

Table 3D: Normalized MUF (M_i) Values for Facilities handling Pu and Pu-239, other than Reprocessing Plants [6,7]. (Normalized with Respect to Input.)

Serial No.	M_i (% of Input)	Serial No.	M_i (% of Input)
1	1.64	11	0.18
2	1.36	12	0.10
3	1.11	13	0.10
4	0.51	14	0.08
5	0.47	15	0.06
6	0.39	16	0.06
7	0.29	17	- 0.10
8	0.23	18	- 0.14
9	0.22	19	- 1.28
10	0.19		

Mean value (μ): + 0.14
Standard deviation (σ): ± 0.67

Table 4: Normalized Values of MUF (M_i) for Groups of Facilities. Types of Facilities and Material Used Unknown. (Small Number of Data/Facility)

Serial No.	M_i	Ref.	Serial	M_i	Ref.	Serial	M_i	Ref.
1	5.86	5	46	0.40	11	91	0.07	5
2	5.74	5	47	0.39	5	92	0.07	11
3	2.96	5	48	0.36	5	93	0.06	11
4	2.44	5	49	0.35	5	94	0.06	5
5	2.44	5	50	0.35	5	95	0.06	5
6	2.22	5	51	0.34	5	96	0.05	5
7	2.14	5	52	0.31	5	97	0.05	5
8	2.05	5	53	0.30	5	98	0.05	11
9	1.80	11	54	0.30	5	99	0.04	11
10	1.78	5	55	0.29	5	100	0.04	5
11	1.62	5	56	0.27	11	101	0.03	5
12	1.52	5	57	0.22	5	102	0.02	5
13	1.43	5	58	0.21	5	103	0.01	5
14	1.35	5	59	0.21	5	104	0.01	5
15	1.24	5	60	0.21	5	105	0.01	5
16	1.23	5	61	0.21	5	106	0.01	5
17	1.20	5	62	0.19	5	107	-0.01	11
18	1.18	5	63	0.19	5	108	-0.02	5
19	1.18	5	64	0.18	5	109	-0.02	5
20	1.16	5	65	0.17	5	110	-0.03	5
21	1.14	5	66	0.15	5	111	-0.03	5
22	1.06	5	67	0.13	5	112	-0.05	11
23	1.00	5	68	0.13	5	113	-0.06	5
24	0.94	5	69	0.13	5	114	-0.06	5
25	0.94	11	70	0.13	5	115	-0.07	5
26	0.92	5	71	0.12	5	116	-0.09	5
27	0.90	11	72	0.12	5	117	-0.10	5
28	0.85	11	73	0.12	5	118	-0.13	5
29	0.82	5	74	0.11	5	119	-0.17	5
30	0.78	5	75	0.11	5	120	-0.32	5
31	0.69	5	76	0.11	5	121	-0.36	5
32	0.67	5	77	0.11	5	122	-0.38	11
33	0.64	5	78	0.11	11	123	-0.41	5
34	0.63	5	79	0.10	5	124	-0.62	5
35	0.62	11	80	0.10	5	125	-1.96	5
36	0.61	5	81	0.10	5	126	-3.22	5
37	0.60	5	82	0.09	5			
38	0.54	5	83	0.09	5			
39	0.53	5	84	0.09	5			
40	0.52	5	85	0.09	5			
41	0.49	5	86	0.09	5			
42	0.49	11	87	0.08	5			
43	0.49	11	88	0.07	5			
44	0.43	5	89	0.07	5			
45	0.42	5	90	0.07	5			

Mean value (μ): 0.48

Standard deviation (σ): $\pm$ 1.01

Table 5: Normalized MUF Values (M_{is}) from a Known Type of Facility and Material.
(Normalized with Respect to Input/Feed and Systematic Errors.)

Serial No.	M_{is}	Reference	Serial No.	M_{is}	Reference
1	5.86	4	46	0.40	7
2	4.78	7	47	0.39	4
3	3.77	4	48	0.38	7
4	3.15	6	49	0.38	7
5	2.47	4	50	0.36	7
6	2.46	8	51	0.35	6
7	2.46	9	52	0.32	7
8	2.42	6	53	0.26	10
9	2.35	6	54	0.25	8
10	2.28	4	55	0.21	10
11	2.28	8	56	0.21	10
12	2.23	4	57	0.19	6
13	2.09	9	58	0.19	6
14	2.08	9	59	0.18	10
15	1.89	4	60	0.15	6
16	1.85	7	61	0.13	7
17	1.75	4	62	0.13	7
18	1.67	4	63	0.12	6
19	1.63	4	64	0.12	6
20	1.62	7	65	0.11	6
21	1.58	4	66	0.11	7
22	1.49	7	67	0.09	7
23	1.46	4	68	0.04	7
24	1.44	7	69	0.02	7
25	1.30	4	70	-0.11	7
26	1.22	7	71	-0.16	7
27	1.09	4	72	-0.19	6
28	0.98	7	73	-0.27	6
29	0.98	1	74	-0.28	7
30	0.85	7	75	-0.81	4
31	0.79	4	76	-0.86	4
32	0.78	7	77	-1.04	4
33	0.74	10	78	-1.09	4
34	0.71	10	79	-1.28	4
35	0.67	7	80	-1.37	4
36	0.66	7	81	-1.47	4
37	0.65	7	82	-1.82	4
38	0.63	7	83	-1.82	4
39	0.56	4	84	-1.89	4
40	0.56	7	85	-2.35	6
41	0.54	4	86	-2.46	6
42	0.48	7	87	-2.62	6
43	0.47	7	88	-4.05	4
44	0.47	10	89	-4.61	4
45	0.42	6			

Mean value (μ): + 0.29
Standard deviation (σ): ± 0.91

Table 6: Results of Test of Normality for Different Categories of Normalized MUF Data

Statistics	Relevant values for different categories of data in table							
	2	3A	3B	3C	3D	3A+3B +3C+3D	4	5
$k_1 = \mu$ = mean	0.38 (0.34)+	0.18	0.22	0.23	0.14	0.20	0.29	0.48 (0.40)
$k_2 = \sigma^2$ = variance	0.90 (0.66)	1.57	0.49	0.050	0.44	0.68	0.83	1.01 (0.56)
Standard deviation σ	0.95 (0.81)	1.25	0.70	0.22	0.67	0.82	0.91	1.01 (0.75)
k_3	1.25 (-0.0026)	0.037	0.089	0.011	-0.12	-0.027	0.010	2.29 (-0.0062)
k_4	7.79 (1.57)	1.68	0.27	-0.000024	0.48	1.49	1.69	13.04 (1.82)
σk_3^2	0.018 (0.0071)	0.86	0.031	0.000034	0.032	0.019	0.040	0.051 (0.0089
σk_4^2	0.066 (0.019)	5.81	0.065	0.0000073	0.064	0.053	0.14	0.21 (0.020)
$k_3/\sigma k_3$ measure of skewness	9.24 (-0.03)	0.040	0.51	1.94	-0.69	-0.20	0.050	10.15 (-0.066)
$k_4/\sigma k_4$ measure of kurtosis	30.41 (11.44)	0.70	1.04	-0.0090	1.88	6.46	4.57	28.44 (12.75)
g_1	1.47 (-0.005)	0.019	0.26	1.01	-0.42	-0.049	0.013	2.24 (-0.015)
g_2	9.70 (3.66)	0.68	1.11	-0.0098	2.44	3.25	2.44	12.66 (5.73)
n	241 (239)	30	26	25	19	100	89	126 (124)
Distribution	Not Normal	Normal	Normal	Normal	Normal	Not Normal	Not Normal	Not Normal

+The numbers in brackets are obtained when the highest two values of MUF are rejected.

Table 7: MUF and the Corresponding Feed Values for a Reprocessing Facility in Absolute Units /4/

Serial No. (Time sequence)	Feed (arbitrary units)	MUF (arbitrary units)
1	173.5	1.73
2	184.9	1.99
3	159.8	2.07
4	161.0	0.73
5	44.5	-1.17
6	103.6	-1.12
7	165.4	-1.21
8	213.8	0.48
9	50.9	-0.43
10	169.7	0.52
11	182.4	1.51
12	153.7	2.16
13	93.7	-2.16
14	233.3	-2.42
15	204.5	1.51
16	271.4	-1.68
17	306.6	0.17
18	510.5	-3.02
19	299.5	-3.11
20	424.7	-2.07
21	400.2	1.30
22	764.6	4.75
23	536.1	4.84
24	585.5	5.44
25	95.9	3.20
26	736.7	-3.37
27	392.9	3.72
28	226.0	-1.77
29	332.5	4.23
30	373.8	8.04

Table 7A: Comparison of Test of Normality on Absolute and Normalized MUF Values for a Reprocessing Facility /4/. (Normalized with respect to feed)

Statistics	Values for Absolute MUF	Values for Normalized MUF
$k_1 = \mu$ = mean	0.83	0.18
$k_2 = \sigma^2$ = variance	8.28	1.57
k_3	12.75	0.037
k_4	-13.75	1.68
$\sigma^2_{k_3}$	125.64	0.86
$\sigma^2_{k_4}$	4467.06	5.81
k_3/σ_{k_3}	1.14	0.040
k_μ/σ_{k_4}	-0.21	0.70
g_1	0.54	0.019
g_2	-0.20	0.68

Table 8: Results of Time Sequence Analysis of Absolute MUF Values from a Reprocessing Facility /4/

Serial No.	Accounting period (both numbers inclusive)	Mean μ	Standard deviation σ	Outliers outside $\mu \pm 2\sigma$ (accounting period)	Outliers outside $\mu \pm 2\sigma_c$ +) (accounting period)
1	1 to 15	0.279	1.584	18,19,22,23, 24,26,27,29, 30	22,23,24,30
2	6 to 20	-0.725	1.716	22,23,24,25, 27,29,30	22,23,24,29,30
3	11 to 25	0.695	2.999	30	-
4	16 to 30	1.378	3.740	-	-
5	overall 1 to 30	0.829	2.877	30	30

+) σ_c = corrected standard deviation

$$= \left[\sigma^2 + 2\sigma^2 \sqrt{\frac{2}{n-1}} \right]^{1/2}$$

$\frac{2\sigma^4}{n-1}$ - being the variance of variance

Table 8A: Results of Time Sequence Analysis of Normalized MUF Values

Serial No.	Accounting period (both numbers inclusive)	Mean μ	Standard deviation σ	Outliers outside $\mu \pm 2\sigma$ (accounting period)	Outliers outside $\mu \pm 2\sigma_c$ (accounting period)
1	1 to 15	-0.086	1.283	25	-
2	6 to 20	-0.345	0.944	5,13,25,30	5,13,25
3	11 to 25	0.204	1.320	5,13,25	5,13,25
4	16 to 30	0.437	1.209	5,13,25,30	5,25
5	overall 1 to 30	0.176	1.254	5,25	25

Table 9: Results of Feed Sequence+) Analysis of Absolute MUF Values from Reprocessing Plant /4/

Serial No.	Feed sequence (both numbers inclusive)	Mean μ	Standard deviation σ	Outliers outside $\mu \pm 2\sigma$ (feed sequence Nos)	Outliers outside $\mu \pm 2\sigma_c$ (feed sequence Nos)
1	1 to 15	0.654	1.564	19,21,22,26 27,28,29,30	22,27,28
2	6 to 20	0.179	1.773	21,22,27,28, 29,30	22,28
3	11 to 25	0.909	2.967	22	-
4	16 to 30	1.003	3.825	-	-
5	overall 1 to 30	0.829	2.877	22	22

+) MUF data was arranged in order of increasing feed and then the sequence was analysed.

Table 9A: Results of Feed Sequence Analysis of Normalized MUF Values

Serial No.	Feed sequence (both numbers inclusive)	Mean μ	Standard deviation σ	Outliers outside $\mu \pm 2\sigma$ (feed sequence Nos)	Outliers outside $\mu \pm 2\sigma_c$ (feed sequence Nos)
1	1 to 15	0.206	1.526	4	-
2	6 to 20	0.213	0.864	1,3,4,22	1,3,4
3	11 to 25	0.310	0.951	1,3,4	1,3,4
4	16 to 30	0.145	0.961	1,3,4,22	1,4
5	overall 1 to 30	0.176	1.254	1,4	4

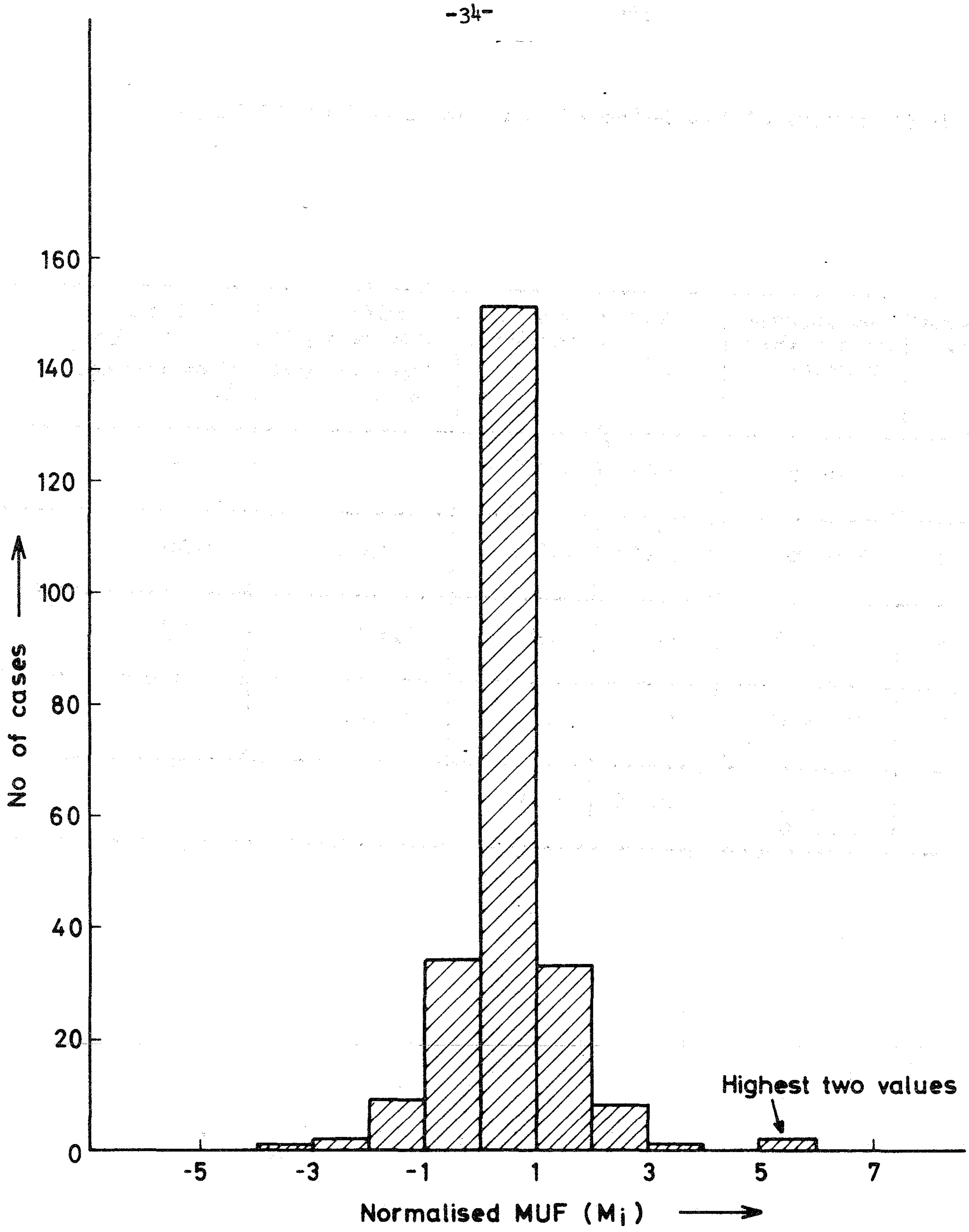


Fig. 1 Histogram of 241 normalised values of MUF (Table 2)

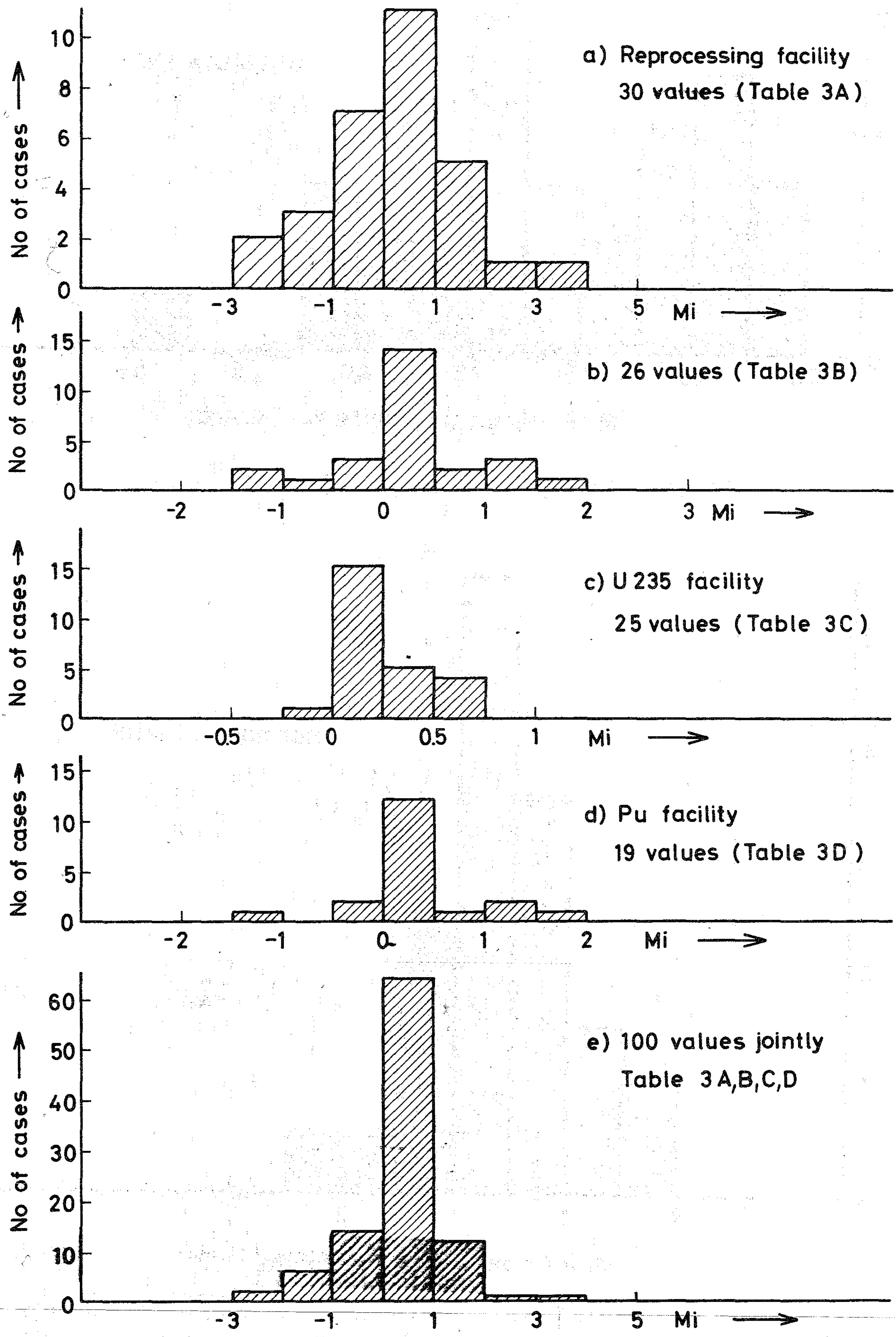


Fig. 2 Histograms a,b,c,d for Mi values for specific facilities. Histogram e for all values jointly from these facilities.

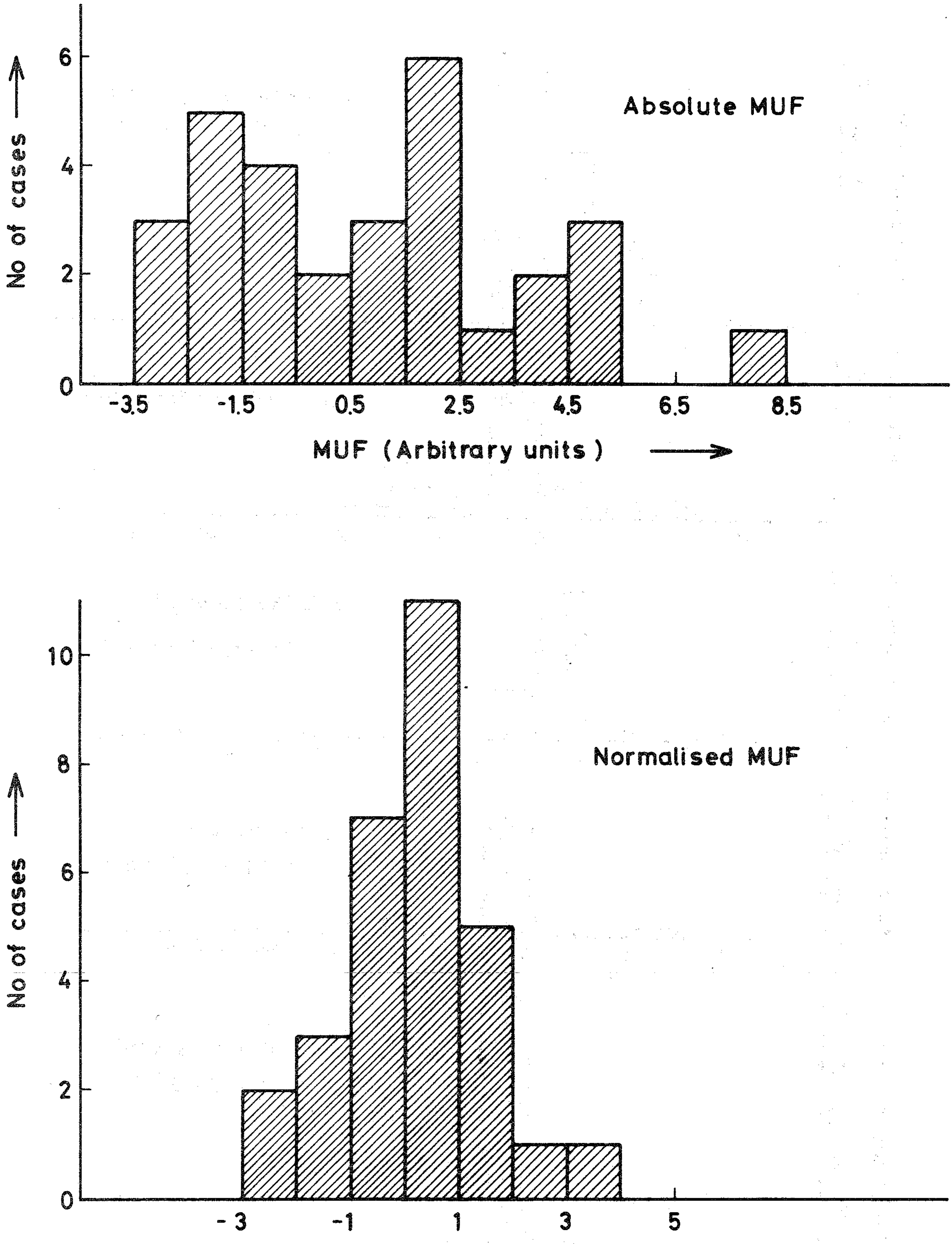
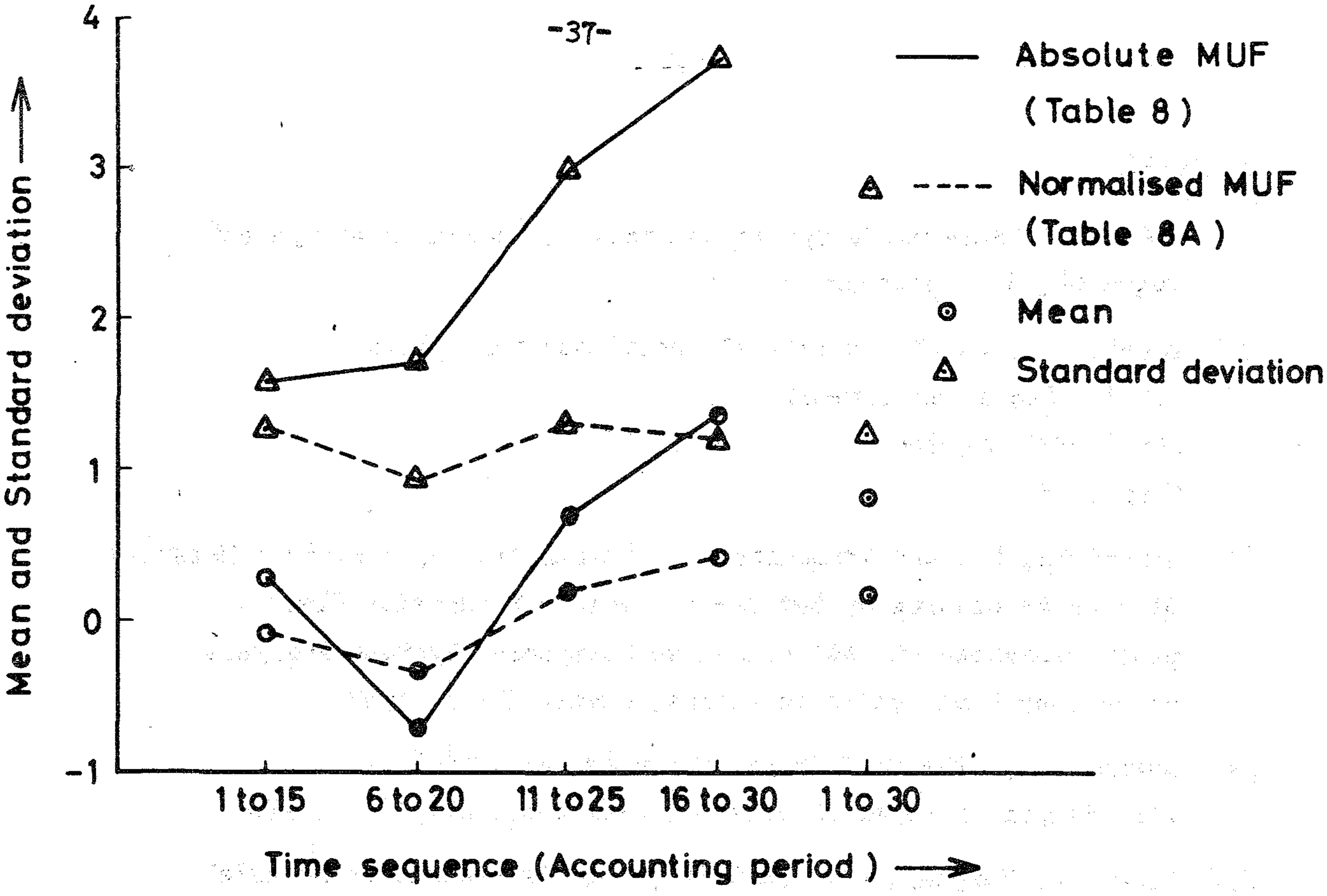


Fig. 3 Histograms of Absolute and Normalised values of MUF in a reprocessing plant (4)



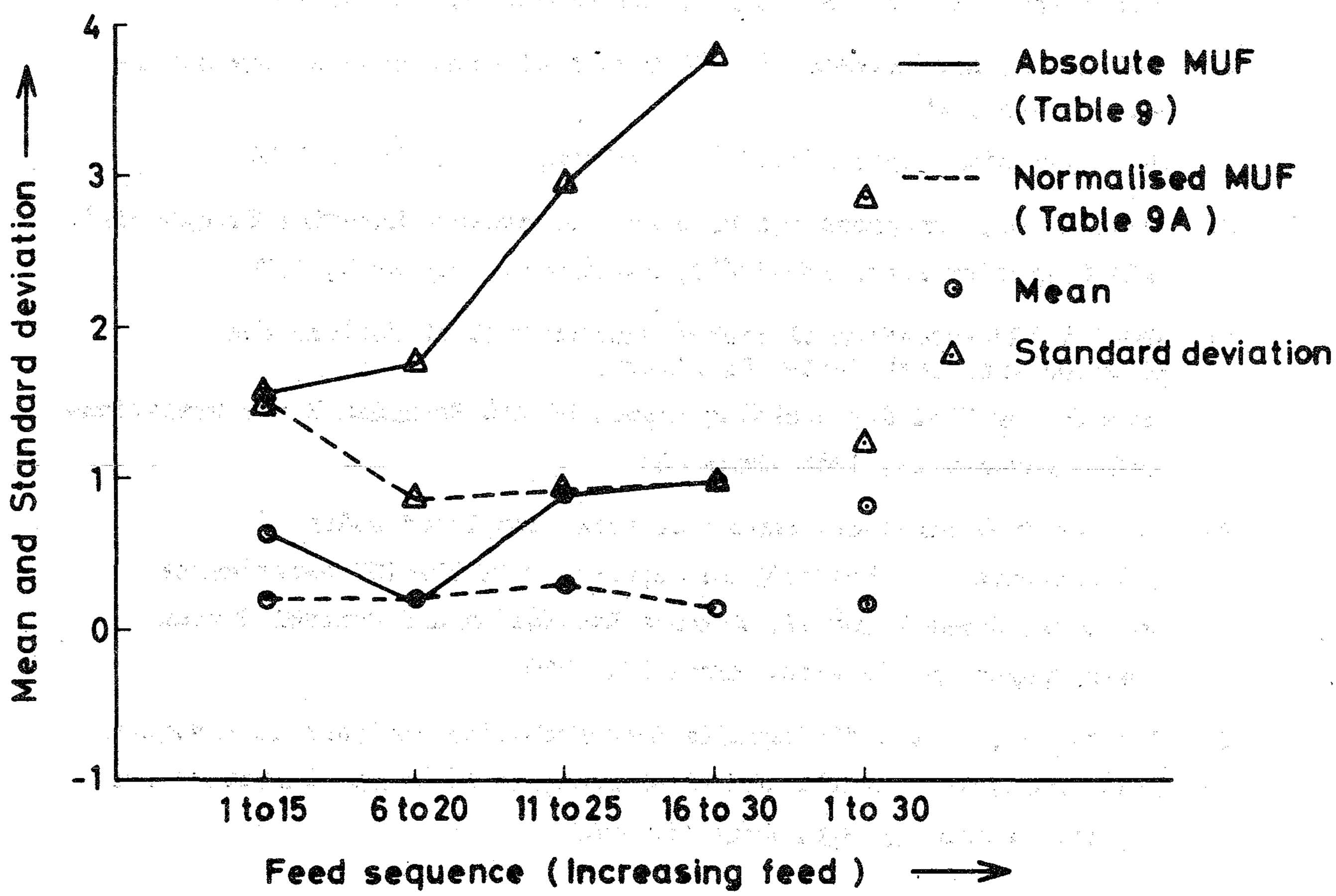


Fig. 4 Mean and Standard deviation of absolute and normalised MUF values for time and feed sequence

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ANNEX 1

The statistical method for testing the normality of a given sample

The statistical method for answering the question if the random variable underlying a given sample is normally distributed or not ('k-statistics') [12] is described in the following:

Let S_r be defined as

$$S_r = \sum_{i=1}^{n} X_i^r \qquad r = 1,2,\ldots$$

where the X_i, i=1,...,n, are the n values of the given sample. Then the quantities k_v, v = 1,...4, are defined in the following way

$$k_1 = \frac{S_1}{n}$$

$$k_2 = \frac{nS_2 - S_1^2}{n(n-1)}$$

$$k_3 = \frac{n^2 S_3 - 3nS_2S_1 + 2S_1^3}{n(n-1)(n-2)}$$

$$k_4 = \frac{(n^3+n^2)S_4 - 4(n^2+n)S_3S_1 - 3(n^2-n)S_2^2 + 12nS_2S_1^2 - 6S_1^4}{n(n-1)(n-2)(n-3)}$$

As one sees immediately, k_1 and k_2 are the sample mean and the sample variance. Since k_3 depends on the cube of the deviations from the expectation value $E(k_1) = \mu$, the expectation values of k_3 will be zero only if the distribution is symmetric. Instead of the value of k_3, which is in units which are the cube of those of the original measurements, the value

$$g_1 = \frac{k_3}{k_2^{3/2}}$$

which is independent of the original units, is usually used as a measure of skewness. The quantity k_4 is particularly sensitive as a measure of 'contamination' in a distribution, i.e. in the presence of a small percentage of widely scattered observations in an otherwise normal distribution. The characteristic of a distribution which is measured by k_4 is called kurtosis. In the case of the normal distribution the expectation value of k_4 is zero. Instead of k_4, normally the dimensionless quantity

$$g_2 = \frac{k_4}{k_2^2}$$

is used.

As in practice even in the case of a normal distribution neither k_3 nor k_4 are zero, it is important to know the possible variations of these quantities. For this reason one determines their variances:

$$\mathrm{var}(k_3) = \frac{6n}{(n-1)(n-2)} \sigma^6$$

$$\mathrm{var}(k_4) = \frac{24n(n-1)}{(n-1)(n-2)(n-3)} \sigma^8$$

However, since they depend on the unknown variance σ^2 of the distribution, they are not directly useful except when n is sufficiently large to assume $\sigma^2 \approx k_2$.

For very large samples, g_1 and g_2 can be considered as being approximately normally distributed with expectation value 0 and variances $\frac{6}{n}$ and $\frac{24}{n}$. Hence, $t_1 = g_1\sqrt{\frac{n}{6}}$ and $t_2 = g_2\sqrt{\frac{n}{24}}$ are approximately standard normal deviates, and one can determine the significance thresholds for a test on normality for a given error probability α. For smaller samples, corrected tables for the distributions of g_1 and g_2 are available and used here.