Estimation of Total Error in DWPF Reported Radionuclide Inventories (U)

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1.0 INTRODUCTION

The Defense Waste Processing Facility (DWPF) at the Savannah River Site is required to determine and report the radionuclide inventory of its glass product. This requirement is given as Specification 1.2.2 in the Waste Acceptance Product Specifications for Vitrified High Level Waste Forms (WAPS) of the U.S. Department of Energy's Office of Environmental Restoration and Waste Management[1]. The DWPF will take the following steps to meet this requirement. For each macro-batch, the DWPF will report both the total amount (in curies) of each reportable radionuclide and the average concentration (in curies/gram of glass) of each reportable radionuclide. The DWPF is to provide the estimated error of these reported values of its radionuclide inventory as well.

The objective of this document is to provide a framework for determining the estimated error in DWPF's reporting of these radionuclide inventories. This report investigates the impact of random errors due to measurement and sampling on the total amount of each reportable radionuclide in a given macro-batch. In addition, the impact of these measurement and sampling errors and process variation are evaluated to determine the uncertainty in the reported average concentrations of radionuclides in DWPF's filled canister inventory resulting from each macro-batch.

2.0 SUMMARY

For each macro-batch, DWPF is to determine estimates of the average concentration and the total curies for each reportable radionuclide. One of two methods is to be used to derive an estimate of the concentration for each of these radionuclides for each batch within the macro-batch. A propagation of variance for each of these two methods is completed to estimate the variance of the total error affecting these derived values. In this analysis, the errors considered reflect only random variation (both within and between batch) and, where appropriate, process variation. Process variation is due to any steps taken during the processing of a batch that influence the concentration of a radionuclide of interest. The processing effects are not necessarily random but are due to the natural variation of the DWPF process (they are assumed to be a representative set of the effects due to such processing).

3.0 ESTIMATING RADIONUCLIDE CONCENTRATIONS

There are two methods for deriving estimates of the radionuclide concentrations for each batch of slurry comprising a DWPF macro-batch: the direct and the indirect methods. Only one of these two methods is to be used for each of the radionuclides of interest.

3.1 Direct Method

Some radionuclides are present in high enough concentrations to be measured directly in the DWPF laboratory. The concentration for each of these radionuclides will be measured in a sample from each Melter Feed Tank (MFT) batch.
3.1.1 Radionuclide Concentrations Per Process Batch

For radionuclide R, measured directly in the DWPF, let $C_{i,R}$ represent the measured concentration of R in the $i$th MFT batch in curies per gram of glass. For a given batch $i$, the measurement process is subject to various sources of random variation which induce uncertainties in the resulting measured concentration. The direct measurement approach assumes that all of these errors are random quantities affecting only the concentration measurement for the given process batch. The uncertainty of each $C_{i,R}$ can be computed from the uncertainties of the appropriate procedures (sampling, which includes inhomogeneity of the MFT contents, and analytical, which includes sample preparation) used for the determination of $C_{i,R}$.

Between any two batches of the macro-batch, the random errors affecting the corresponding concentration measurements are assumed to be independent. For example, there should be no systematic influence from the calibration of the instrument(s) used to make the concentration measurements over two or more batches; that is, these instruments should be recalibrated before being used to make the concentration measurement for each batch.

However, there is variability in the radionuclide concentrations between batches, a batch-to-batch variability, due to random errors or process variation which are independent of the first set of errors, and which are also independent from one batch to the next. These random errors are due to such factors as the inhomogeneity of the feed tank; and the process variation is due to any steps taken during the processing of the batch that influence the concentration of radionuclide R. The processing effects are not necessarily random but are due to the natural variation of the DWPF process (they are assumed to be a representative set of the effects due to such processing).

Consider the population of potential values of the concentrations of radionuclide R over a macro-batch. If the DWPF processing that leads up to the determination of each of the $C_{i,R}$'s for a given macro-batch is a stable, repeatable process, then the concept of such a population makes sense, and the variability of this hypothetical population is of interest. The set of $b$ $C_{i,R}$ values for the macro-batch may be thought of as a random sample from this hypothetical population, and the sample standard deviation of this set of $b$ $C_{i,R}$ values, $s_{C_{i,R}}$, provides an estimate of the standard deviation of this population. This simple statistic, $s_{C_{i,R}}$, appropriately handles the (independent) random errors and process variation influencing the values for the concentrations of radionuclide R for a given macro-batch and provides a meaningful measure of the variability of the population of possible concentrations. It is computed as follows:

$$s_{C_{i,R}} = \sqrt{\frac{\sum_{i=1}^{b} (C_{i,R} - \overline{C_{i,R}})^2}{b - 1}}$$

(1)

where $\overline{C_{i,R}}$ represents the simple arithmetic average of the $b$ $C_{i,R}$ values.
3.1.2 Standard Error of the Average Concentration Per Batch

If the average concentration, \( \overline{C}_{R} \), is reported for a macro-batch, then there is a need to provide an estimate of the uncertainty (including both measurement and process) for this summary statistic. An appropriate estimate of this uncertainty is the standard error (or standard deviation) of this average, denoted by \( S_{\overline{C}_{R}} \), which is computed from the standard deviation of the population given by equation (1) as follows

\[
S_{\overline{C}_{R}} = \frac{S_{C_{R}}}{\sqrt{b}}.
\]

This is the appropriate (1-sigma) uncertainty for the average concentration, \( \overline{C}_{R} \), for a macro-batch.

3.1.3 Total Radionuclide Content Per Macro Batch

The DWPF is also required to report the total content (in curies) of the radionuclide for the macro-batch. The total curies of each reportable radionuclide will be calculated by adding together the curies of this radionuclide in each process batch. The estimated error for this total is discussed in this section.

For radionuclide R, measured directly in the DWPF, let \( C_{i,R} \) represent the concentration of R in the i\(^{th}\) MFT batch, \( W_{i} \) the solids content of the i\(^{th}\) MFT batch, and \( V_{i} \) the volume of the i\(^{th}\) MFT batch sent to the melter. The content in curies of radionuclide R in the i\(^{th}\) batch (denoted by \( C_{i,R} \)) is given by the product

\[
C_{i,R} = C_{i,R}W_{i}V_{i}.
\]

Since there are b batches in the macro-batch, there will be b values of \( C_{i,R} \) for each radionuclide R in a given macro-batch. The sum of these values represents the total curies of radionuclide R in the macro-batch. This sum is expressed by the equation

\[
C_{R} = \sum_{i=1}^{b} C_{i,R} = \sum_{i=1}^{b} C_{i,R}W_{i}V_{i}.
\]

Since the values on the right-hand side of equation (4) are measured quantities, they are subject to various sources of error that plague these measurement processes. The uncertainties for each \( C_{i,R} \) have already been discussed including the errors associated with the sampling (e.g., MFT inhomogeneity) and analytical (including preparation and instrumentation) procedures. For a given batch i, the errors associated with the \( C_{i,R} \) and \( W_{i} \) measurements are assumed to be correlated, and the errors associated with \( V_{i} \) are assumed to be independent of the errors of these other two measurements. These are the errors influencing the measurements for each batch i of the macro-batch.
The errors associated with one MFT batch, i, are expected to be independent of all the other batches making up the DWPF macro-batch. The independence of the errors across batches implies no systematic influence from tank volume calibrations or from instrument calibrations; specifically, the instruments used to measure the $C_{i,R}$ and $W_i$ for batch $i$ are to be recalibrated before being used to measure these values for batch $i+1$.

Since $C_{i,R}$ is the product of random variables (as given by equation (4)), the approach provided by Goodman in [2] and [3] was considered as a possible method for estimating the error variance of $C_{i,R}$ in equation (3). However, the more general method of variance or error propagation (see reference [4]) was selected for use here for consistency since it is appropriate for this situation and handles the more complex relationships derived from the indirect method.

A variance propagation approach, based on a Taylor series expansion, is used for estimating the error variance of the total content of radionuclide R, $C_R$, in equation (4). A covariance term is introduced in the approximation to properly handle the correlated errors for $C_{i,R}$ and $W_i$. Let $Var(C_R)$ represent the estimate of the variance of $C_R$ for the macro-batch; then

$$Var(C_R) = \sum_{i=1}^{b} \left( \left( \frac{\partial C_R}{\partial C_{i,R}} \right)^2 \cdot var(C_{i,R}) + \left( \frac{\partial C_R}{\partial W_i} \right)^2 \cdot var(W_i) \right)$$

$$+ \left( \frac{\partial C_R}{\partial V_i} \right)^2 \cdot var(V_i) + 2 \left( \frac{\partial C_R}{\partial C_{i,R}} \right) \left( \frac{\partial C_R}{\partial W_i} \right) \cdot cov(C_{i,R}, W_i),$$

(5)

where

$$\frac{\partial C_R}{\partial z}$$ represents the partial derivative of $C_R$ in equation (4) with respect to the variable $z$,

$var(C_{i,R})$ represents the random error variance for $C_{i,R}$ for batch $i$,

$var(W_i)$ represents the random error variance for $W_i$ for batch $i$,

$var(V_i)$ represents the random error variance for $V_i$ for batch $i$, and

$cov(C_{i,R}, W_i)$ represents the random error covariance for $C_{i,R}$ and $W_i$ for batch $i$.

Taking the partial derivatives leads to the following estimate of the error variance of the total radionuclide content, $C_R$, for the macro-batch.
To complete the determination of the estimate of the error variance of \( c_i \), the error variances of \( C_i, W_i \), and \( V_i \) must be estimated. Each of these variances are estimated by the appropriate random errors: (sampling, preparation, and instrument). The covariance of the errors for \( C_i, W \) and \( W_i \) must also be estimated to complete the determination of equation (6).

3.2 Indirect Method

The concentrations of some radionuclides, particularly those present in trace amounts, will be determined by characterization of the macro-batch prior to processing in the DWPF. Samples of the waste in the Tank Farm will be taken from each sludge and precipitate batch making up the macro-batch. Each reportable radionuclide whose concentration is to be determined with this indirect approach occurs in the sludge or precipitate (not both). The concentrations of the reportable radionuclides in the individual waste streams (sludge and precipitate) will be determined from these samples. Finally, the concentration of each of these radionuclides in a given MFT batch will be calculated by multiplying the average concentration in the waste streams (over the macro-batch) by a factor reflecting the dilution of that stream in the MFT batch. The dilution factors for a waste stream are based on the dilutions seen in the MFT batch of certain species unique to that waste stream.

3.2.1 Radionuclide Concentrations Per Process Batch

For a radionuclide, \( R \), which is determined indirectly using analyses of SRS Tank Farm samples, let \( C_{i,R} \) represent the concentration of \( R \) in the \( i \)th MFT batch. Then,

\[
C_{i,R} = \frac{1}{X} C_{\text{stream},R} \left[ \sum_{a=1}^{X} \frac{C_{i,a}}{C_{\text{stream},a}} \right],
\]

where \( X \) is a constant representing the number of dilution factors estimated for the unique waste stream that contains radionuclide \( R \) \(^1\),

\( C_{\text{stream},R} \) is the concentration of radionuclide \( R \) in its unique feed stream,

\( C_{\text{stream},a} \) is the concentration of species \( a \) in the feed stream, and

\(^{1}\) There are two waste streams: sludge and precipitate. Iron, aluminum, manganese, and calcium occur only in the sludge, and these species will be used for that waste stream with \( X=4 \). Potassium and titanium occur only in the precipitate hydrolysis aqueous (PHA) stream, and they will be used for that waste stream with \( X=2 \).
\( c_{i,a} \) is the concentration of species \( a \) in the \( i \)th MFT batch.

The concept of a hypothetical population of possible concentrations for a given macro-batch also is to be used, since an estimate of the variance of this population is of interest. As in section 3.1.2 above, there are within-batch and between-batch errors associated with the uncertainty of the set of \( b C_{i,R} \) values computed using equation (7) for a macro-batch.

First, consider the within-batch variance of \( C_{i,R} \), which is to be denoted by \( \text{var}_i(C_{i,R}) \). Some of the errors contributing to the uncertainty of the quantities in equation (7) are expected to be correlated. Specifically, the error for \( C_{\text{stream},R} \) is expected to be correlated with the error for each of the \( C_{\text{stream},a} \)’s, the errors for the \( C_{\text{stream},a} \)’s are expected to be correlated with each other across the \( a \)’s, and the errors for the \( c_{i,a} \)’s are expected to be correlated with each other across the \( i \)’s but not across the \( i \)’s.

Using the variance propagation approach (and its associated notation) leads to the following equation for \( \text{var}_i(C_{i,R}) \):

\[
\text{var}_i(C_{i,R}) = \left( \frac{\partial C_{i,R}}{\partial C_{\text{stream},R}} \right)^2 \text{var}(C_{\text{stream},R}) + \sum_{a=1}^{x} \left( \frac{\partial C_{i,R}}{\partial C_{\text{stream},a}} \right)^2 \text{var}(C_{\text{stream},a}) \\
+ \sum_{a=1}^{x} \left( \frac{\partial C_{i,R}}{\partial c_{i,a}} \right)^2 \text{var}(c_{i,a}) + \sum_{a=1}^{x} \sum_{c=1}^{x} \frac{\partial C_{i,R}}{\partial c_{i,a}} \frac{\partial C_{i,R}}{\partial c_{i,c}} \text{cov}(c_{i,a}, c_{i,c}) \\
+ \sum_{a=1}^{x} \sum_{c=1}^{x} \frac{\partial C_{i,R}}{\partial c_{\text{stream},a}} \frac{\partial C_{i,R}}{\partial c_{\text{stream},c}} \text{cov}(C_{\text{stream},a}, C_{\text{stream},c}) \\
+ 2 \left( \frac{\partial C_{i,R}}{\partial C_{\text{stream},R}} \right) \sum_{a=1}^{x} \frac{\partial C_{i,R}}{\partial C_{\text{stream},a}} \text{cov}(C_{\text{stream},R}, C_{\text{stream},a}).
\]  

(8)

Evaluating the partials of equation (8) leads to.
\[
\text{var}_i(C_{i,R}) \approx \left( \frac{1}{x} \sum_{a=1}^{x} \frac{C_{i,a}}{C_{\text{stream},a}} \right)^2 \text{var}(C_{\text{stream},R}) + \sum_{a=1}^{x} \left( - \frac{C_{\text{stream},R} C_{i,a}}{x C_{\text{stream},a}^2} \right)^2 \text{var}(C_{\text{stream},a}) + \sum_{a=1}^{x} \sum_{c=1}^{x} \left( \frac{C_{\text{stream},R} C_{i,a}}{x C_{\text{stream},a}^2} \right) \text{cov}(C_{i,a}, C_{i,c}) + \sum_{a=1}^{x} \sum_{c=1}^{x} \sum_{a \neq c}^{x} \left( \frac{1}{x} \sum_{a=1}^{x} \frac{C_{i,a}}{C_{\text{stream},a}} \right) \left( - \frac{C_{\text{stream},R} C_{i,c}}{x C_{\text{stream},a}^2} \right) \text{cov}(C_{\text{stream},a}, C_{\text{stream},c}) + 2 \sum_{a=1}^{x} \left( \frac{1}{x} \sum_{c=1}^{x} \frac{C_{i,c}}{C_{\text{stream},c}} \right) \left( - \frac{C_{\text{stream},R} C_{i,a}}{x C_{\text{stream},a}^2} \right) \text{cov}(C_{\text{stream},R}, C_{\text{stream},a}) \]

(9)

Averaging the \( \text{var}_i(C_{i,R}) \) values computed over the \( b \) batches of the macro-batch provides an estimate of the within-batch variance of the underlying population of the \( C_{i,R} \) values for the macro-batch, denoted by \( \text{var}_w(C_{i,R}) \). Thus,

\[
\text{var}_w(C_{i,R}) = \frac{1}{b} \sum_{i=1}^{b} \text{var}_i(C_{i,R})
\]

(10)

The between-batch variability of the concentrations of radionuclide \( R \) is reflected in the variation of the set of \( b \) \( C_{i,R} \) values; that is, the sample variance of these \( b \) values. This sample variance also contains some of the within-batch variation and, thus, overestimates the between-batch variability. However, the contribution of the within-batch variability to this term is not easily determined (if at all), so no attempt is made to separate this portion of the sample variance from that attributed to the between-batch variation. The estimate of the total variance of the \( C_{i,R} \) values of the macro-batch, denoted by \( \text{var}(C_{i,R}) \), is given by the sum of the within- and between-batch variances, or

\[
\text{Var}(C_{i,R}) = \text{Var}_w(C_{i,R}) + \frac{1}{b-1} \sum_{i=1}^{b} (C_{i,R} - \overline{C_{i,R}})^2
\]

(11)

where \( \overline{C_{i,R}} \) represents the average concentration of the \( b \) \( C_{i,R} \) values. Although the estimate given by equation (11) may overestimate the total variance, no problem is anticipated by the over estimation. When the actual data are evaluated this assumption will be checked.
The standard deviation computed from the square root of the variance estimate of equation (11) is an appropriate measure of the standard deviation of the possible population of concentration values or radionuclide R for a given macro-batch.

3.2.2 Standard Error of the Average Concentration Per Batch

As in the direct measurement case of section 3.1.2, if the average concentration, \( \bar{C}_{i,R} \), is reported for a macro-batch, then there is a need to provide an estimate of the uncertainty (both measurement and process) for this summary statistic. An appropriate estimate of this uncertainty is the standard error (or standard deviation) of this average which is the square root of the estimate of the variance of this average, denoted by \( \text{var}(\bar{C}_{i,R}) \). The estimate of this variance is given by

\[
\text{var}(\bar{C}_{i,R}) = \frac{\text{var}(C_{i,R})}{b} \tag{12}
\]

The corresponding estimate of the standard deviation is the appropriate (1-sigma) uncertainty for the average concentration, \( \bar{C}_{i,R} \), for a macro-batch for a radionuclide R computed using the indirect measurement process.

3.2.3 Total Radionuclide Content For a Macro-Batch

Another possible format for reporting the radionuclide inventory for a macro-batch is to report the total content (in curies) of the radionuclide for the macro-batch.

Using the notation developed above and letting \( C_R \) represent the total curies of radionuclide R in the macro-batch, then

\[
C_R = \sum_{i=1}^{b} C_{i,R} W_i V_i = \frac{1}{\chi} C_{\text{stream},R} \sum_{i=1}^{b} \frac{C_{i,a}}{C_{\text{stream},a}} \sum_{i=1}^{b} C_{i,a} W_i V_i \tag{13}
\]

This is the quantity to be reported to represent the total content (in curies) of radionuclide R in a given macro-batch.

In addition to the correlations already described for the model given by equation (7), the errors for the \( C_{i,a} \)'s may be correlated with the error in \( W_i \) for each batch i in equation (13).

An error propagation for equation (13) can be used to generate an appropriate estimate for the uncertainty of this reported total radionuclide content with these correlations taken into consideration. If the error variance of \( C_R \) is denoted by \( \text{var}(C_R) \), then
\[
\text{var}(C_R) \approx \left( \frac{\partial C_R}{\partial C_{\text{stream},R}} \right)^2 \text{var}(C_{\text{stream},R}) + \sum_{a=1}^{x} \left( \frac{\partial C_R}{\partial C_{\text{stream},a}} \right)^2 \text{var}(C_{\text{stream},a}) \\
+ \sum_{i=1}^{b} \left( \frac{\partial C_R}{\partial W_i} \right)^2 \text{var}(W_i) + \sum_{i=1}^{b} \left( \frac{\partial C_R}{\partial V_i} \right)^2 \text{var}(V_i) + \sum_{i=1}^{b} \sum_{a=1}^{x} \left( \frac{\partial C_R}{\partial c_{i,a}} \right)^2 \text{var}(c_{i,a}) \\
+ \sum_{a=1}^{x} \sum_{c=1}^{x} \sum_{c \neq a} \left( \frac{\partial C_R}{\partial C_{\text{stream},a}} \right) \left( \frac{\partial C_R}{\partial C_{\text{stream},c}} \right) \text{cov}(C_{\text{stream},a}, C_{\text{stream},c}) \\
+ \sum_{i=1}^{b} \sum_{a=1}^{x} \sum_{c=1}^{x} \sum_{c \neq a} \left( \frac{\partial C_R}{\partial c_{i,a}} \right) \left( \frac{\partial C_R}{\partial c_{i,c}} \right) \text{cov}(c_{i,a}, c_{i,c}) \\
+ 2 \sum_{i=1}^{b} \left( \frac{\partial C_R}{\partial W_i} \right) \sum_{a=1}^{x} \left( \frac{\partial C_R}{\partial c_{i,a}} \right) \text{cov}(W_i, c_{i,a}) \\
+ 2 \sum_{a=1}^{x} \left( \frac{\partial C_R}{\partial C_{\text{stream},R}} \right) \left( \frac{\partial C_R}{\partial C_{\text{stream},a}} \right) \text{cov}(C_{\text{stream},R}, C_{\text{stream},a})
\]

(14)

Evaluating the partial derivatives of equation (14) yields the following
\[
\text{var}(C_R) \approx \left( \frac{\sum_{i=1}^{b} V_i W_i}{x} \sum_{a=1}^{X} C_{i,a} \right)^2 \text{var}(C_{\text{stream},R}) + \sum_{a=1}^{X} \left( \frac{C_{\text{stream},R}}{x} \sum_{i=1}^{b} \frac{C_{i,a} W_i V_i}{C_{\text{stream},a}^2} \right)^2 \text{var}(C_{\text{stream},a}) + \sum_{i=1}^{b} \left( \frac{C_{\text{stream},R}}{x} \sum_{a=1}^{X} \frac{C_{i,a} V_i}{C_{\text{stream},a}} \right)^2 \text{var}(W_i) + \sum_{i=1}^{b} \left( \frac{C_{\text{stream},R}}{x} \sum_{a=1}^{X} \frac{C_{i,a} W_i V_i}{C_{\text{stream},a}^2} \right)^2 \text{var}(c_{i,a}) + \sum_{a=1}^{X} \sum_{c \neq a}^{X} \left( -\frac{C_{\text{stream},R}}{x} \sum_{i=1}^{b} \frac{C_{i,a} W_i V_i}{C_{\text{stream},a}^2} \right) \left( -\frac{C_{\text{stream},R}}{x} \sum_{i=1}^{b} \frac{C_{i,c} W_i V_i}{C_{\text{stream},c}^2} \right) \text{cov}(C_{\text{stream},a}, C_{\text{stream},c}) + \sum_{i=1}^{b} \sum_{a=1}^{X} \sum_{c \neq a}^{X} \left( \frac{C_{\text{stream},R} W_i V_i}{xC_{\text{stream},a}} \right) \left( \frac{C_{\text{stream},R} W_i V_i}{xC_{\text{stream},c}} \right) \text{cov}(c_{i,a}, c_{i,c}) + 2 \sum_{i=1}^{b} \sum_{a=1}^{X} \left( \frac{C_{\text{stream},R} V_i}{x} \sum_{c=1}^{X} \frac{C_{i,c}}{C_{\text{stream},c}} \right) \left( \frac{C_{\text{stream},R} W_i V_i}{xC_{\text{stream},a}} \right) \text{cov}(W_i, c_{i,a}) + 2 \sum_{a=1}^{X} \left( \frac{b}{x} \sum_{i=1}^{b} \frac{C_{i,a} W_i V_i}{C_{\text{stream},a}^2} \right) \left( -\frac{C_{\text{stream},R}}{x} \sum_{i=1}^{b} \frac{C_{i,a} W_i V_i}{C_{\text{stream},a}^2} \right) \text{cov}(C_{\text{stream},R}, C_{\text{stream},a}) \right). (15)
\]

Simplifying equation (15) leads to
This variance propagation for the indirect approach assumes that all of the errors affecting these measurements are independent (except as indicated by the appropriate covariance terms), random quantities. To complete the determination of the \( \text{var}(C_R) \) in equation (16), the variances and covariances of the errors in the measurements of this equation must be estimated.
4.0 CONCLUDING COMMENTS

As more definitive estimates of the errors affecting these measurements are identified, they may be used in these calculations to provide a representative estimate of the total error in the reported radionuclide inventory for each DWPF macro-batch. Specifically, if some of the errors are relative errors, then estimates of the corresponding variances should be entered appropriately into the equations of this report. For example, if \( y \) is a measurement with a relative standard deviation of \( n\% \), then the estimate of the variance of the error associated with the measurement \( y \) is equal to \( \left( \frac{yn}{100} \right)^2 \).

The approach presented in this memo assumes that all of the measurements will be corrected for known biases associated with vessel uniformity, sampling, and analysis. If not accounted for or if steps are not taken (such as recalibrating instrumentation between MFT process batches), the systematic errors from these sources could be significant contributors to the overall uncertainty. In this analysis, the errors considered reflect only random variation.

5.0 REFERENCES


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