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A SAMPLE COMPOSITING STRATEGY FOR CLASSIFYING NON-RADIOACTIVE HAZARDOUS WASTE FOR TRANSPORT (U)

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ABSTRACT

A probabilistic strategy for sample compositing is developed to minimize the radiometric analyses required for classifying non-radioactive waste for transport. Such waste must have concentrations of total radioactivity that are below an acceptance limit of $L = 2 \text{ nCi/g}$. A composite of N different samples must have a radioactive concentration below L/N to assure that no individual sample is unacceptable. Unacceptable samples are eventually identified by analyses of successive splits composited with $N/2$, $N/4$, ... of the original samples. The probable number of such analyses is derived using Gaussian distributions for the composite concentrations, per invoking the Central Limit Theorem. A preliminary compositing strategy, based only on the average concentration μ , uses $N = L/\mu$ to yield a minimum fraction of $\approx 2\mu/L$ analyses per total samples. These approximations are useful for $L/\mu > 4$. Refined strategies, based on both the μ and σ for the concentration distribution, define the optimization more precisely. Experimental data from composites of 880 samples of low-level radiometric waste are consistent with the calculated predictions .

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1. Introduction

Because of the growing emphasis on the environment, the analysis volume for waste samples has increased dramatically in recent years. At the Savannah River Site (SRS), hazardous waste that is classified as non-radioactive must be certified as having less than 2 nCi/g total activity before it can be shipped off-plant. Compliance with this limit is required by U.S. Department of Transportation (DOT) regulations [1]. The demand for establishing acceptable radionuclide levels for off-plant waste shipments has resulted in strategies for compositing samples to help alleviate the analysis load. A strategy based on probabilistic methods is examined and should have broad application in areas of waste certification. This method is a variation of sequential statistical analysis [2-4], which minimizes the required measurements in batch appraisals.

Various composite strategies have evolved over the years. Early work by Dorfman [5] developed a compositing method for screening blood samples, in which the only individual samples tested were those associated with defective composites. This approach continues to be useful in current applications [6], and it is normally optimized relative to the group size of the composite sample [5-9]. The above method may be optimized further if a sequential binary search is applied to identify the defective samples of defective composites [7-8]. The present work develops a compositing strategy based on a binary search.

2. General Theory

For transport of non-radioactive hazardous waste, DOT requires that the radioactive concentration c be below the limit of $L = 2n\text{Ci/g}$. Normally, the contents of waste containers are homogenized, and then individual samples are taken and analyzed. If N waste samples from different containers are equally mixed as a single composite sample, this sample must have $c < L/N$, to assure that no individual component sample of the composite contains the entire contamination at a level exceeding L . If measurements on any composite sample yielded $c > L/N$, then two recomposites would be made, each from a different $N/2$ samples included in the original composite. Each of these recomposites would be tested against the criteria that $c < L/[N/2]$ or $c < 2L/N$. This recompositing process would continue as necessary, until (1) all recomposites have satisfied $c < 2^j L/N = L_j$ at some j th recomposite level and (2) any individual component samples with $c > L$ have been identified. Because each recomposite will split the preceding composite by a factor of 2, it is recommended that $N = 2^n$, where n is an integer. Both N and 2^n notations are included in the development below. Thus, the j th recomposite limit is given by

$$L_j = 2^j L/N = 2^{j-n} L. \quad (1)$$

The above recompositing scheme requires some knowledge of the probabilities of exceeding the L_j limits to project how many sample analyses are needed for a given N . Suppose some large number of samples S must be appraised relative to compliance with the limit L . Then, the initially composited samples will require A_0 analyses, as given by

$$A_0 = S/N = 2^{-n} S. \quad (2)$$

A total of $A_0 P(c_0 > L_0)$ analyses would fail the compliance criteria, requiring analysis of A_1 recomposited samples, which are composed of paired splits from the first composites. (Here, $P(c_0 > L_0)$ is the probability that an initial composite sample has $c_0 > L_0$). More explicitly, A_1 is given by

$$\begin{aligned} A_1 &= 2 A_0 P(c_0 > L_0) \\ &= 2^1 S/N P(c_0 > L_0) \\ &= 2^{1-n} S P(c_0 > L_0) \end{aligned} \quad (3)$$

Continuing this procedure, it is readily observed that the number A_j of j th recomposite analyses will be given by

$$\begin{aligned}
 A_j &= 2 A_{j-1} P(C_{j-1} > L_{j-1}) \\
 &= 2^j S/N \prod_{i=1}^j P(C_{i-1} > L_{i-1}) \\
 &= 2^{j-n} S \prod_{i=1}^j P(C_{i-1} > L_{i-1})
 \end{aligned} \tag{4}$$

Thus, the total number A of expected analyses is obtained by summing the A_j , viz

$$A = \sum_{j=0}^n A_j \tag{5}$$

where in summary

$$\begin{aligned}
 A_j &= 2^j S/N \prod_{i=0}^j P_i = 2^{j-n} S \prod_{i=0}^j P_i \\
 P_i &= 1 \text{ for } i = 0 \\
 &= P(C_{i-1} > L_{i-1}) \text{ for } i > 0 \\
 L_i &= 2^{1/N} L = 2^{i-n} L
 \end{aligned}$$

3. Probability Assignments

3.1 Fundamental Considerations

The preceding treatment defines the general formalism; however, the probabilities P_i must be defined to obtain results. For a composite made of a sufficiently large number N of samples, the Central Limit Theorem predicts that the composite concentrations have a Gaussian distribution. Subcomposites of large $N/2^j$ should also have Gaussian distributions. Thus, for sufficiently large composites, the P_i of P_1, P_2, \dots should approximate Gaussian distributions, but the $\dots P_{n-1}, P_n$ cannot in general be assumed to be Gaussian. Fortunately, the latter P_i normally have only a minor influence on the number of predicted analyses A , and thus the Normal distribution can be used for each P_i . The discussion below details the basis for this choice of P_i and develops the resulting formalism for predicted analyses A .

Most strategies would use a first composite $P_1 \leq 0.5$, to assure that no more than 50% of these composites require recomposites as paired splits. These first recomposites would have probability $P_2 < P_1$ of requiring the next recomposite step, because the contaminant limit L_j is twice as high for successive recomposite steps. (The effective σ_j of the distribution also increases with successive recomposite steps, but by a smaller factor $\approx \sqrt{2}$, which does not cancel the effect of the increased L_j). Thus, in general, $0.5 \geq P_1 > P_2 > \dots > P_n$. Consequently, for the A predicted by eq. (5), each term $A_j = 2P_1 2P_2 \dots 2P_j S/N$ becomes progressively smaller as j increases, so that the terms containing $\dots P_{n-1}, P_n$ have only minor influence on the total sum A. Thus, use of the Gaussian distribution for P_1, P_2, \dots , where the Central Limit Theorem applies, and for $\dots P_{n-1}, P_n$, where the effect on A is minor, should yield reasonable assignments for the P_i .

The probabilities P_i are defined by the Gaussian distribution notation

$$P(x > X) = \int_X^{\infty} \frac{1}{\sqrt{2\pi}} S \exp\left[-\frac{(x - U)^2}{2 S^2}\right] dx = G(X|U, S) \quad (6)$$

From the Central Limit Theorem, composites of N individual samples, which are distributed with mean μ and variance σ^2 , have a Gaussian distribution $G(X|\mu, \sigma/\sqrt{N})$. Applying this to P_1 ,

$$P_1 = P(c_0 > L_0) = G(L_0|\mu, \sigma/\sqrt{N}) = G(L_0|\mu, \sigma_1) \quad (7)$$

and in general we may write

$$P_i = P(c_{i-1} > L_{i-1}) = G(L_{i-1}|\mu, \sigma/\sqrt{N/2^{i-1}}) = G(L_{i-1}|\mu, \sqrt{2^{i-1}}\sigma_1) \quad (8)$$

where σ_1 corresponds to the distribution of the initial composites. The σ_1 may be obtained directly from a series of composite measurements or indirectly from a series of sample measurements. However, if sample measurements are used, caution should be exercised, because the sample measurements are not likely to have a Gaussian distribution. The present study addresses samples that are likely to have log-Normal distributions, as discussed below.

Log-Normal distributions are often applicable to environmental radiation measurements [10]. Because non-radioactive hazardous waste contains primarily natural

radionuclides, with the possibility of contamination by man-made radio-nuclides, it is considered that this waste should have radioactive concentrations that are log-Normally distributed. Consistent with this treatment, waste is expected to be generated from a variety of sources with contamination levels that depend on numerous multiplicative factors which are randomly distributed. Distributions that depend on random multiplicative factors are log-Normally distributed [11].

A log-Normal distribution should be examined if a series of individual sample measurements is to be studied. The log-Normal distribution is given by

$$P(\ln(x) > \ln(X)) = G(\ln(X) | \ln(\mu_g), \delta) , \quad (9)$$

where the log of each measurement yields a Gaussian, with mean equal to the log of the geometric mean μ_g and sigma of δ . Thus, a sigma deviation of $\ln(x) - \ln(\mu_g) = \delta$ corresponds to a one-sigma factor deviation of $F = x/\mu_g = \exp(\delta)$. If this distribution is to be examined in x (as opposed to $\ln(x)$), the resulting mean μ and variance σ^2 are related to the log-Normal parameters as [11]

$$\begin{aligned} \mu &= \mu_g \exp(\delta^2/2) \\ \sigma^2 &= \mu_g^2 \exp(\delta^2) [\exp(\delta^2) - 1] . \end{aligned} \quad (10)$$

These values of μ and σ are the ones that apply to the Gaussian probabilities defined for P_i above.

Although it would be ideal to have values for μ and σ for developing a compositing strategy, this is not always practical in the early phase of a measurement program. Typically, one needs to define a preliminary compositing strategy at the initiation of such a program; then, as the measurements data base increases, refined strategies based on μ and σ values can evolve. Thus, approaches for both preliminary and refined compositing strategies are needed and each is discussed below.

3.2 Preliminary Approach

A few preliminary measurements are usually sufficient to yield an average concentration μ , but the σ and functional nature of the P_i can only be determined from larger data sets. However, the μ alone is a very useful parameter, because it is an approximation of the median for anticipated P_i distributions in many applications, including the present one. In fact, for the Gaussian model of P_i developed above, the mean and median are identical. Because the median of P_i corresponds to $P_i = 0.5$, the preliminary estimate of μ contains some information about the probabilities. This information can be used to develop a preliminary compositing strategy.

The preliminary compositing strategy is developed using eq. (5), and the information for $P_i = P(c_{i-1} > \mu) = 0.5$. Suppose a strategy using $P_1 = 0.5$ is selected, which corresponds to the upper limit for reasonable recompositing, as discussed earlier. Then, μ must equal $L_0 = L/N = L/N_m$, where N_m is the value of N that produces this mean condition. The corresponding P_2 has the value of 0.5 when $L_1 = 2L/N = 2L/2N_m = \mu$, or when N increases to $2N_m$. Continuing this progression, in general

$$P_i = 0.5 \quad \text{when } N = 2^{i-1}N_m = 2^{i-1}L/\mu, \text{ for } i > 0 \quad (11)$$

To estimate P_i at other N , the σ must be known, as indicated by eq. (8). To examine a simplified overview, σ is set equal to zero, yielding δ -function distributions of P_i , viz

$$\begin{aligned} P_i &= 0.0 & \text{when } N < 2^{i-1}N_m \\ &= 0.5 & \text{when } N = 2^{i-1}N_m & \quad i > 0 \\ &= 1.0 & \text{when } N > 2^{i-1}N_m \end{aligned} \quad (12)$$

$$P_i = H(N - 2^{i-1}N_m) = H(N/N_m - 2^{i-1})$$

where $H(\dots)$ is the Heaviside function.

Using these P_i , the number of analyses per total samples is calculated from eq. (5) as

$$\begin{aligned} A &= S/N \left[1 + \sum_{j=1}^n 2^j H(N/N_m - 1)H(N/N_m - 2) \dots H(N/N_m - 2^{j-1}) \right] \\ &= S/N \left[1 + \sum_{j=1}^n 2^j H(N/N_m - 2^{j-1}) \right] \end{aligned} \quad (13)$$

In generalized notations, this may be written

$$(A/S)N_m = N_m/N \left[1 + \sum_{j=1}^n 2^j H(N/N_m - 2^{j-1}) \right] \quad (14)$$

which essentially allows all N_m cases to be given in a single plot as shown in fig. 1. From this plot, it is recognized that for a given N_m , the minimum A/S corresponds to $N = N_m$. If a non-zero σ were used, the plot would smooth out the abrupt changes

shown for the δ -function P_i , but the general behavior would be similar. As a reference, the figure presents a smoothed average of this δ -function model. In general, for a preliminary strategy based only on the average μ , an initial composite of

$$N = N_m = L/\mu \quad (15)$$

should be near the optimum. Furthermore, the figure indicates that the corresponding minimum $A/S N_m$ is between 1 and 3, yielding the estimate of

$$(A/S)_{\min} \approx 2/N_m = 2 \mu/L . \quad (16)$$

It is also noted that as N increases, the asymptotic values of $A/S N_m$ are between 2 and 4, as indicated in the figure.

3.3 Refined Approach

In the refined approach, sufficient data are available to obtain both μ and σ that define the P_i . It is wise to test that the P_i are suitably modeled as Gaussian distributions, before invoking eq. (8); however, given that N is reasonably large, the Central Limit Theorem virtually assures this. Conformance with a Gaussian is confirmed if an integral probability plot of the data yields a straight line.

Upon certifying the above requirements, the probabilities P_i may be directly calculated using eq. (8). Then these P_i are incorporated into eq. (5) to yield the number of analyses.

4. Calculations and Measurements

Preliminary data for waste drum samples indicated a μ somewhat below 0.1 nCi/g. Thus, since $L = 2$ nCi/g, initial composites with $N = 20 \approx L/\mu$ drum samples each were used, per eq. (15). This compositing strategy was used for a total of 880 drums which required $A_0 = 44$ analyses on initial composites, $A_1 = 4$ analyses on 1st recomposites, and $A_2 = 2$ analyses on 2nd recomposites, for a total of $A = 50$ analyses. Thus, the required analyses were only about 6% of those that would have been used if each drum were analyzed individually. This compares favorably with the $\approx 10\%$ projected by eq. (16).

The refined data resulting from the 44 composite analyses were sufficient to yield values of $\mu = 0.056$ nCi/g and $\sigma_1 = 0.026$ nCi/g, as shown in the probability plot of fig. 2. The resulting straight line also confirms that the composite data are

distributed as a Gaussian. Table 1 shows that the A_j predictions of the refined approach are in reasonable agreement with the observed number of analyses. In particular, the refined approach for $N = 20$ yields 48 analyses, which is in good agreement with the 50 observed experimentally.

Table 1 displays a series of refined predictions over the range of $N = 1$ to $N = S = 880$, wherein the μ and σ from the experimental measurements are used. It is interesting to compare the above predictions for $N = 20$ against those for the minimum number of analyses. Specifically, a minimum of 46 analyses results when $N = 24$, which is only slightly lower. Also, had the final average of 0.056 nCi/g been used (instead of 0.1 nCi/g) as a preliminary μ estimate, the corresponding $N = L/\mu = 36$ would have estimated $(2\mu/L)S = 49$ analyses for the preliminary approach, which is very close to the 50 predicted by the refined approach. Overall, Table 1 illustrates that the choice of N is not extremely sensitive to the predicted number of analyses A , provided that N is in the vicinity of the minimum, which is relatively broad.

Calculations were also performed to examine how accurate the preliminary predictions would be for similar cases with different L and σ . For this examination, the L and σ for the experimental results were varied by multiplicative factors. Fig. 3 shows $A/S N_m$ vs N curves for L multiplied by factors ranging from 1/4 to 4. Over this range the absolute minima of $A/S N_m$ deviated by less than $\pm 25\%$. Also, the minimum for each curve is no more than 25% lower than that obtained with the preliminary approach using $N = N_m$. Fig. 4 gives similar data for σ , which is also varied by factors ranging from 1/4 to 4. Here, the absolute minima are within $\pm 40\%$, and the minimum for each curve is no more than 35% lower than that obtained with the preliminary approach using $N = N_m$. Thus, the preliminary approach is useful for a wide range of conditions, and the refined approach can be applied for further optimization as data become available.

Figs. 3 and 4 contain curves that are smoothed relative to the δ -function P_i model of fig. 1. All curves examined lie within the extremes of the δ -function curve. In fig. 3, increasing L causes the curve to approach the δ -function curve, because the correspondingly greater N [per larger $N_m = L/\mu$] yields narrower P_i [per $\sigma_1 = \sigma/\sqrt{N}$ of eqs. (7) and (8)]. In fig. 4, directly decreasing σ_1 , per decreasing σ has a similar effect in causing the curve to approach the δ -function curve of fig. 1.

It should be pointed out that the above ranges of σ can infer corresponding log-Normal distributions for the individual samples. From eq. (10), a log-Normal sigma δ is solved to yield

$$\delta = \sqrt{\ln(1 + \sigma^2/\mu^2)} \quad (17)$$

from which one-sigma factors of $F = \exp(\delta)$ are shown to range from 1.6 to 7.9 for the σ -cases examined in fig. 4. Thus, the log-Normal sample distributions can be quite broad and still permit the preliminary method to be applied.

5. Conclusions

For a binary compositing strategy, the present study illustrates that the choice of $N = L/\mu$ samples per composite yields a near-optimum reduction in the analyses required. Furthermore, an optimally low fraction of $\approx 2\mu/L$ analyses per sample is predicted. This choice of N is most useful for preliminary compositing strategies, where insufficient data are available to model the probability distributions in detail. As data evolve to better describe the probability distributions, the strategy can be modified for further optimization using the refined approach.

Predictions with the above approaches apply quite well for SRS radiation measurements that classify non-radioactive waste for transport, and the relative insensitivity to wide variations from the probability distributions for this waste suggest a broad range of applications. Gaussian probability models were applicable for the present analysis; however, other probability models may also lead to the preliminary choice of $N = L/\mu$, provided that their means are reasonable estimates of their medians.

In figs. 1, 3, and 4, the A/S does not increase dramatically above its minimum as N increases beyond the vicinity of N_m . Although near optimum A/S would result from choosing these higher N , their use is not recommended, as much larger numbers of the initial composites would have to be recomposited, causing the bookwork to become more involved. As mentioned already, a strategy with $P_1 \leq 0.5$ is the more reasonable approach. Also, for $N_m = L/\mu < 4$, relatively little advantage is predicted with these binary compositing strategies.

The general theory of this compositing strategy should also have applications in other areas of regulation and quality control, provided that the acceptance limits L are defined. In addition, should an L be redefined by new requirements, the new sampling strategy is easily deduced, using the μ and σ already developed for the earlier L . Of course, if no prior compositing data exist, the new strategy can be developed according to the general approach, whereby a μ from initial data defines the preliminary approach and both μ and σ from sufficient later data define the refined approach.

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Table 1. Results for Refined Approach

Data is for $S = 880$ paint/solvent waste-drum samples grouped as $A_0 = S/N$ initial composites. All predictions use $\mu = 0.056$ nCi/g and $\sigma = \sqrt{N}\sigma_1 = \sqrt{20} \times 0.026$ nCi/g from the measurements data.

Results	N	A_0	A_1	A_2	A_3	A_4	A
<u>Measurements (a)</u>	20	44	4±2	2±2	0		50±3
<u>Predictions</u>							
Minimum N	1	880	0				880.0
Low N	10	88	0				88.0
$N = 2^{m-1} \approx N_m/2$	16	55	1.0	0			56.0
Measurements N	20	44	4.0	0			48.0
Optimum N	24	36.7	9.2	0			45.9
Intermediate N	28	31.4	15.2	0.1	0		46.7
$N = 2^m \approx N_m$	32	27.5	20.7	0.4	0		48.6
$N = N_m$	36	24.4	24.9	1.1	0		50.4
Practical $N \approx N_m$	40	22	27.6	2.5	0	0	52.1
High N	55	16	28.6	12.9	0.1	0	57.6
Maximum N	880	1	2.0	4.0	8.0	(b)	72.6

a) Experimental uncertainties are approximated from Poisson statistics.

b) $A_4 = 16.0$, $A_5 = 28.6$, $A_6 = 12.9$, $A_7 = 0.1$, and $A_8 = 0$, which is the same sequence exhibited in the High N = 55 case.

Figure Captions

Fig. 1. A/S as a Function of N for δ -Function P_i .

Fig. 2. Integral Probability Plot for Liquid Waste Composites
with $N = 20$.

Fig. 3. A/S as a Function of N for P_i from Liquid Waste Data
for different L values.

Fig. 4. A/S as a Function of N for P_i from Liquid Waste Data
for different σ values.

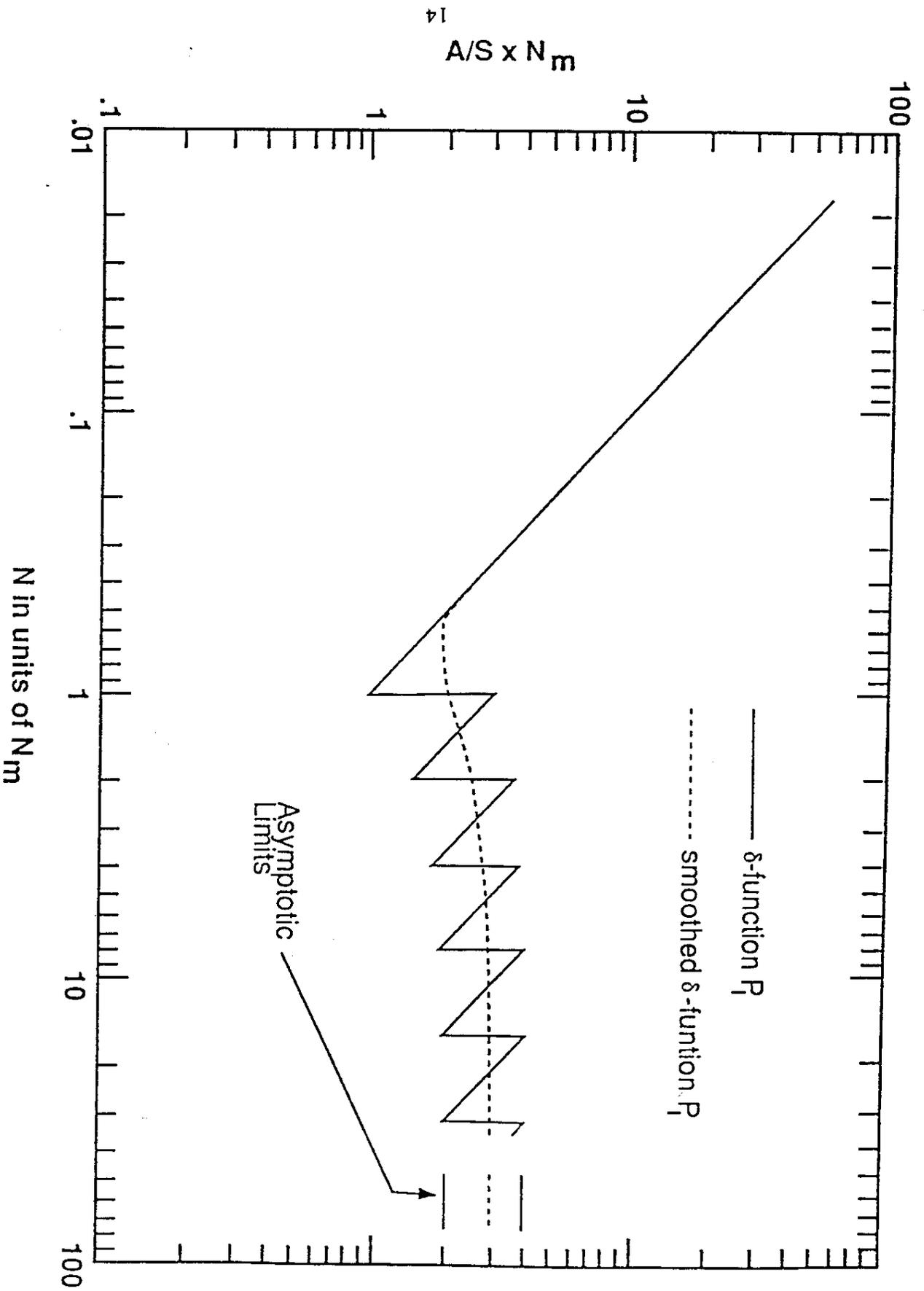


FIGURE 1. A/S as a Function of N for δ -Function P_i .

$$P(c_0 > c) = G(c|\mu, \sigma_1)$$

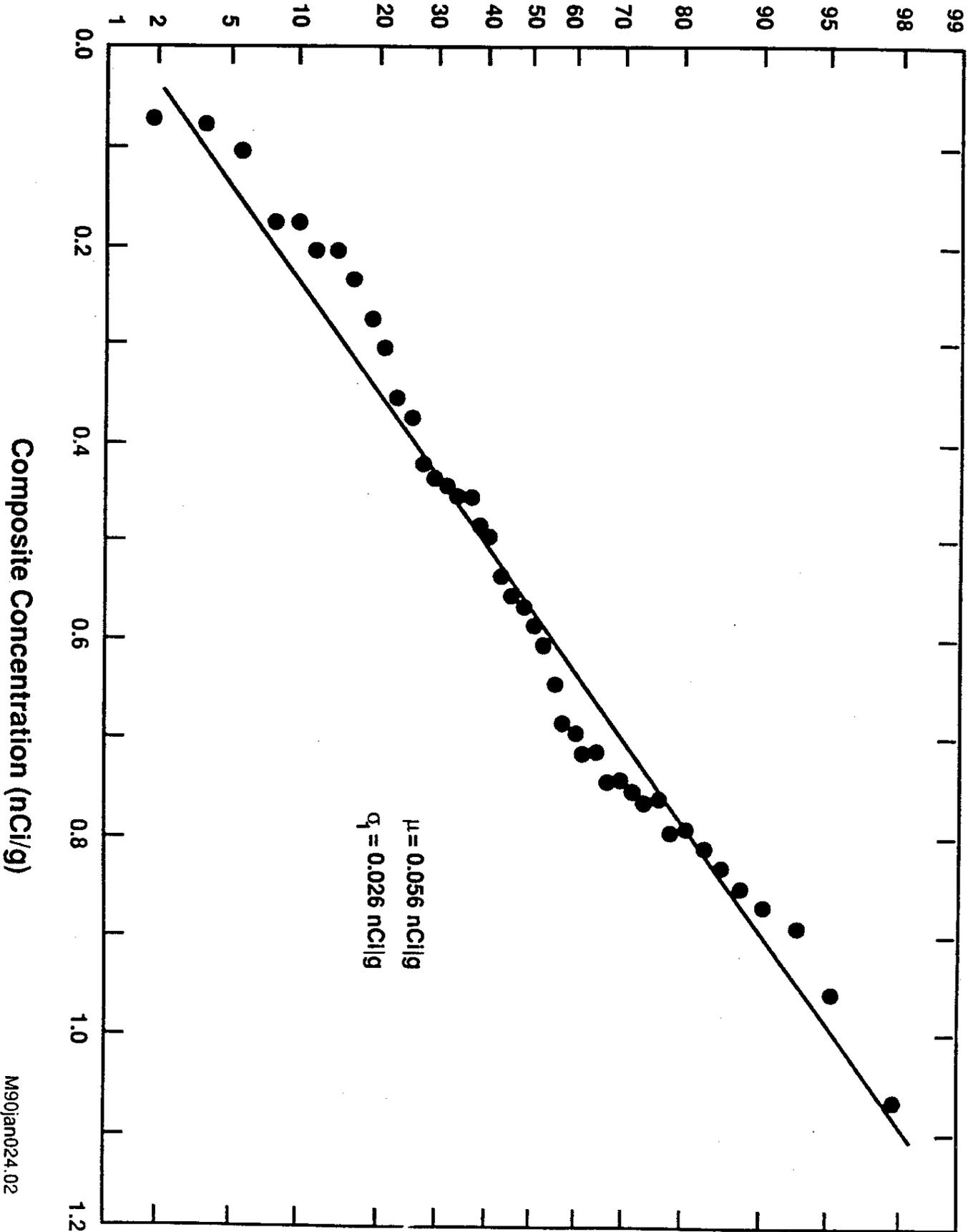


FIGURE 2. Integral Probability Plot for Liquid Waste Composites with N = 20.

M90jan024.02

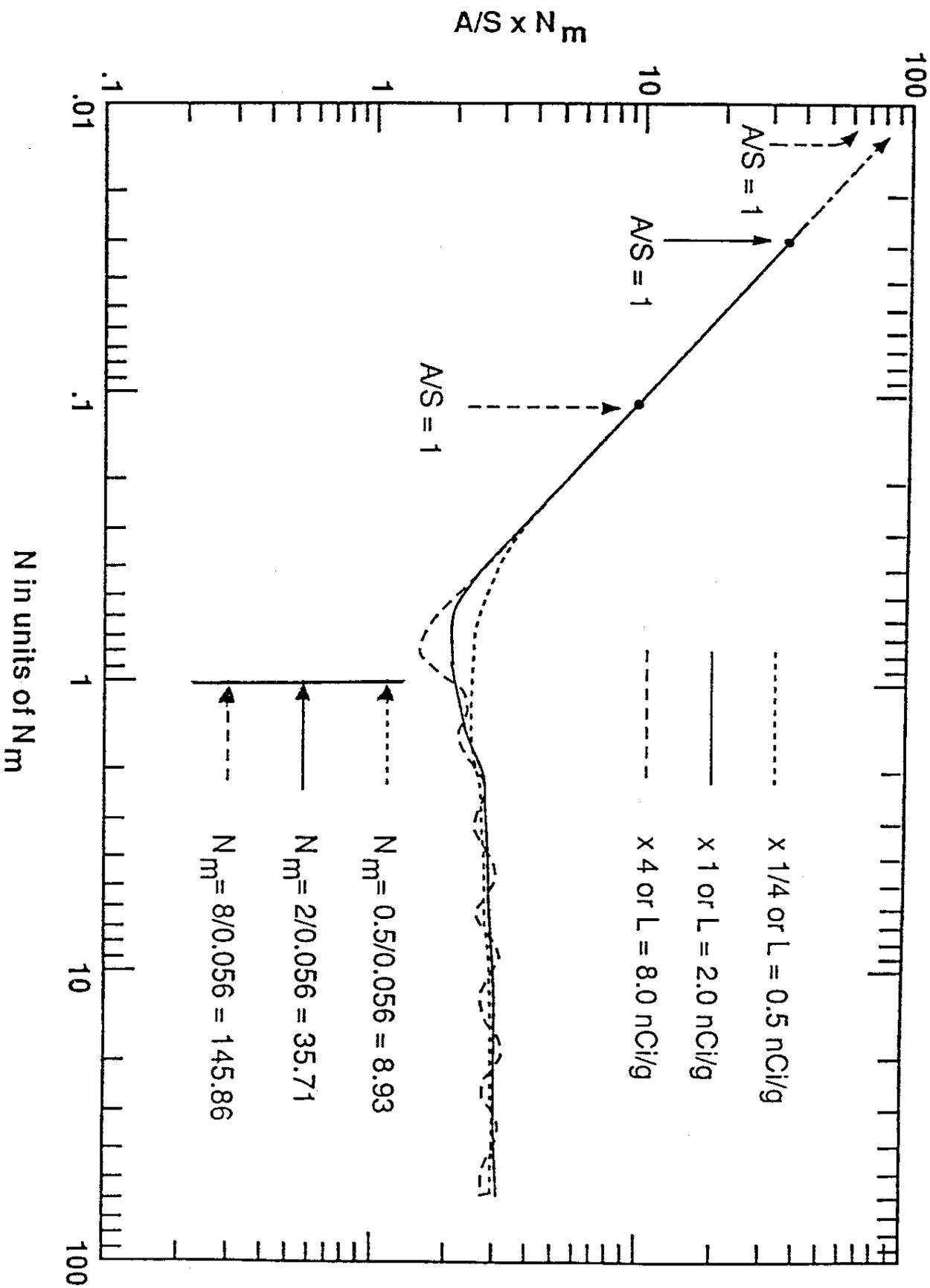


FIGURE 3. A/S as a Function of N for P_i from Liquid Waste Data for different L values.

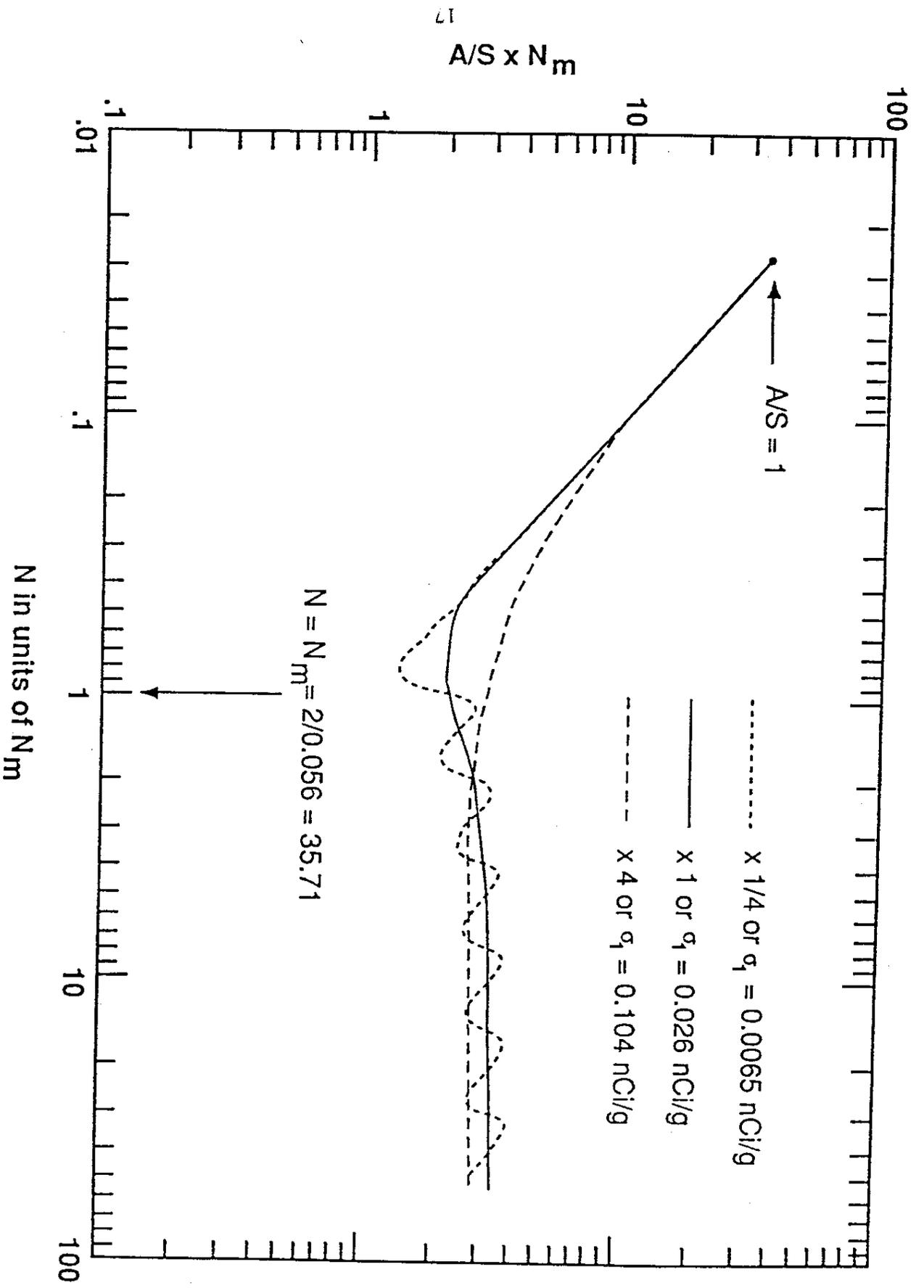


FIGURE 4. A/S as a Function of N for P_i from Liquid Waste Data M90Jan024.04 for different σ values.

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