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Southern California Edison Company

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May 5, 1993

Mr. Stewart Schneider, Project Manager
Radiation Protection and Health Effects Branch
Division of Regulatory Applications
Office of Nuclear Regulatory Research
United States Nuclear Regulatory Commission
Washington, DC 20556-0001

Dear Mr. Schneider:

The following are my comments with respect to the proposed continued studies of hot particle exposure effects and their thresholds:

1. Our experience at San Onofre has been quite consistent through the years. While the number of hot particles encountered has declined considerably, the vast majority of particles encountered has been irradiated fuel as opposed to cobalt. As noted in the attachment, only one out of the thirty-eight particles active enough to evaluate since January, 1992 was a cobalt source. As detailed in the attachment, well over half of all the beta particles emitted by the irradiated fuel have energies below 0.5 MeV (many of them well below this energy). Accordingly, I recommend that future work place emphasis on lower energy beta exposures.

2. In view of the fact that much of the material is reported to be available, histopathological studies of selected lesions should be carried out where they might shed light on unexpected outcomes.

3. The suggestion that extended periods of observation lead to a lower threshold dose should be followed up. As was noted at the meeting, there was little mention of controls in the material submitted, and this aspect of the work should be included in future reports. If possible, the design should also seek to answer such questions as whether the same lesion has recurred or a new, delayed lesion is involved and the cause.

D[illegible]



PDL per. S. Schneider

4. I agree with the proposal that another workshop be held to permit simultaneous observations.

Although it does not bear directly on this research, I feel obliged to note that the present regulatory position requires workers to spend at least 20 percent time (a very conservative estimate) in areas where hot particles may be present. These are frequently areas with measurable to significant gamma dose rates.

I hope the table on the first page of the attachment answers Ms. Raddatzx's question regarding recent hot particle experience. I can provide further information if required.

Sincerely,

Peter Knapp

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Enclosures

Attachment to Letter of May 5, 1993

As described in the enclosed paper, San Onofre has had extensive experience in dealing both with the control and dosimetry of hot particles. The purpose of this attachment is to: 1) Transmit a copy of the paper, together with additional material; and, 2) Provide an update of the types and circumstances of personnel contamination incidents at San Onofre. It is hoped that this information may be useful in determining which areas of additional study would be most useful to power plants.

The paper was presented at the ANS Winter Meeting held in San Francisco in December, 1989. It was initially prepared as a result of many questions from other utilities regarding dose evaluation techniques for hot particle contamination incidents. Most useful may be Table I on page 4. That Table lists the radionuclides of interest expected to be present at various times after reactor shutdown. That same relative inventory is mirrored in numerous particle analyses we've conducted.

Six pages, each headed, "Standard Fuel Fragment Content and Energies," are also included. These provide information on beta particle energy distributions in fuel fragments with ages ranging from 30 to 1825 days. Review of these distributions show that in fuel fragments up to two years old, 47 to 50 per cent of the activity is represented by nuclides whose maximum beta energies are less than 0.5 MeV.

By far, the majority of incidents observed at San Onofre involve fuel fragment contaminants rather than cobalt particles. Our procedures call for a conservative field evaluation of possible skin dose when hot particles are found on personnel. When this evaluation suggests that a dose in excess of 100 mrad may have been received, the incident is referred to the Health Physics Engineering Group for full evaluation.

During the relatively slack period from January, 1992 to the present, approximately 38 such incidents were referred for full evaluation. None of these involved a distributed source. That is, all of these were particle contaminants. One involved a Co-60 particle. The rest were all fuel fragments. Analysis by the method outlined in the paper indicated that almost all of them were one year old. Thirty-one of the 38 resulted in dose assignment of 100 mrad or above. The following table presents a summary of the skin dose and MicroCurie-hours associated with these particles.

| SKIN DOSE (rad) | | | *EXPOSURE (uCi-hrs) | | | NUMBER OF PARTICLES |
|---|---|---|---|---|---|---|
| | < | 0.1 | | < | 0.02 | 9 |
| 0.1 | - | 0.5 | 0.02 | - | 0.1 | 20 |
| 0.5 | - | 1.0 | 0.1 | - | 0.2 | 5 |
| 1.0 | - | 2.0 | 0.2 | - | 0.4 | 3 |
| | | 2.250 | | | 0.45 | 1 |
| | | | | | | -- |
| | | | | | | 38 |

*Assumes 5 rad/uCi-hr

Typically, the contaminant is discovered on an item of clothing rather than directly on the skin surface. Only seven of the 38 contamination incidents (or <20%) had the contaminant directly on the skin surface. That circumstance causes cobalt particles to be less of a problem since the Co-60 beta is essentially eliminated by a layer of clothing. As described in the attached paper, 28 mg/cm^2 is sufficient to make the gamma and beta contributions to skin dose from Co-60 equal.

PJK9305.02:ft

# *SKIN DOSE CALCULATIONS FOR IRRADIATED FUEL FRAGMENTS*

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**Abstract** -- The San Onofre Unit 3 nuclear plant experienced fuel cladding failures during its first fuel cycle. When primary systems were opened for maintenance and refueling, some areas in the plant were contaminated with "fleas" -- microscopic, highly radioactive fuel fragments. Despite extensive controls, personnel occasionally become contaminated with a fragment. This paper discusses methods which may be employed for both field estimation of potential skin dose and for subsequent analytical calculation of skin dose from those contamination incidents.

## INTRODUCTION

Now permanently shut down, was

The Southern California Edison Company operates three pressurized water reactors at their San Onofre site. Unit 1 is a 436 MWe plant that went commercial in January 1968. Units 2 and 3 are identical 1100 MWe plants that share a common radioactive waste system but have separate fuel handling buildings. Unit 2 began commercial operation in August 1983, and Unit 3 went commercial in April 1984.

Indications of fuel cladding failures were observed early in the first fuel cycle for Unit 3. The principal radiological evidence of that fuel integrity problem came in the form of high dissolved noble gas levels (up to 25 $\mu$Ci/g) and high Dose Equivalent Iodine-131 values (0.5 $\mu$Ci/g) in the primary coolant. Subsequent inspection revealed that several fuel assemblies had suffered from hydriding leading to serious cladding failure.

During the first Unit 3 refueling outage when primary systems were opened for maintenance and fuel shuffle, some areas on site were contaminated with "fleas" -- tiny irradiated fuel fragments. Since that time, fuel cladding integrity problems at San Onofre's two other units have introduced fuel fragment contaminants there as well.

A comprehensive contamination control program was developed (Reference 1) aimed at confining the contaminants to their source and keeping them off personnel. Despite those extensive controls, particle contamination incidents involving worker's skin and personal clothing occurred. That situation necessitated the development of a program to adequately evaluate the resultant potential dose to the skin.

This paper examines the methods that have been employed to provide an estimate of skin dose via field measurements and the methods to arrive at the final dose of record for contamination incidents involving fuel fragments.

This paper was presented at the 1989 ANS Winter Meeting in San Francisco.

## METHOD

Initial estimate of potential skin dose from fuel fragment contamination incidents is based on measurements made with standard survey instruments (e.g., friskers and ion chambers) which have been corrected for source size and energy effects.

Final determination of record skin dose consists of first establishing the radionuclide content of the contaminant and then calculating potential dose by applying an evaluation method such as Loevinger's Equation or the VARSKIN computer code, described in References 2 and 3, respectively.

Since the determination of instrument correction factors requires a priori knowledge of the dose rate, skin dose calculation will be discussed before survey instrument methods.

## RADIONUCLIDE CONTENT

The capability of a point source contaminant to deliver dose to the skin is a function of its proximity to the skin, activity, energy, and the type of radiation it emits. Before any evaluation of potential dose may begin, those variables must be established. Although more involved than other possible approaches, it is desirable to establish those variables by determining the radionuclide content of the contaminant.

Accurate assignment of potential skin dose requires a complete knowledge of the particle's nuclide content. Of cours , determining the particle's content requires that the actual contaminant be captured for analysis. That has not proved to be a problem. In practice, our technicians are successful in promptly removing and recovering nearly all such contaminants. The actual analysis, however, is not a simple matter. That analysis is complicated by the known or suspected presence of radionuclides that decay with little or no gamma emission. For that reason, gamma spectrometry alone is not sufficient. Beta spectrometry is an option but few utilities can justify such equipment. Extensive chemical analyses would be necessary to identify and quantify all of the radionuclides present. That is not practical for each skin dose evaluation.

Instead, San Onofre uses an indirect approach to characterize the nuclide content of fuel fragments which begins with a GeLi gamma spectrographic analysis. Those radionuclides which cannot be identified in the gamma based analysis fall into two categories: those in a parent-daughter relationship with an identified species; and those which decay without photon emission.

### Parent-Daughter Relationships

Two frequently encountered parent-daughter relationships are Ru-106/Rh-106 and Ce-144/Pr-144. When either Ru-106, Ce-144, or any other radionuclide in that category is identified, its daughter is assumed to be in secular equilibrium and therefore to exist in the same amount as the parent species.

### Pure Beta Emitters

The activities of fission products whose presence is suspected but not identified due to the absence of photon emission are determined by using a table that lists the

inventory and activities of fission products at specified times after shutdown of a reactor that has operated at 1000 kW for 1 year (Reference 4.) While it is understood that commercial reactors are far larger than 1000 kW, the relative amounts of nuclides should be representative. That table has been reproduced in a modified form and included as Table I. (Noble gases, iodines and nuclides contributing less than 0.1% to the total mix have been excluded.)

In addition to the daughter nuclides described above, the following radionuclides are also expected to be present under the conditions and in the quantities described below: Sr-89, Sr-90, Y-90, Y-91, Pr-143, Pm-147. All other nuclides contained in Table I are identifiable by gamma spectroscopy and are included in the dose calculation if detected in the sample.

Fuel Fragment Age

Because of the differing decay rates present, it is both necessary and possible to estimate the "age" of the fuel fragment. That is, how long it has been removed from the neutron flux of the reactor core. That can be accomplished by examining the ratio of identified radionuclides with differing half-lives. To ensure that the difference between the observed ratio and the expected "T=0" ratio is due solely to the difference in decay rates, it is necessary to eliminate other potential removal mechanisms which would invalidate the method. For instance, the water solubility of some elements such as cesium make them undesirable choices. Due to zirconium in some fuel cladding, a ratio of Nb-95 and Zr-95 would produce an invalid result due to the added source of those nuclides. The ideal choice is two nuclides whose presence is strictly from fission (at least in that environment) with the same chemical behavior and different half-lives.

The approximate age of the fuel fragment can be satisfactorily determined by calculating the Ce-141/Ce-144 sample activity ratio and using the plot included as Figure 1. That linear graph is obtained by plotting the cerium activity ratio versus time on semi-log paper. If Ce-141 is not identified by gamma spectroscopy, the MDA value for Ce-141 is used (if available) or, the mixture is decay corrected to the last reactor shut down date. Either method provides a conservative estimate of age.

Referring to Table I, the theoretical yields (T=0 column values) of the added nuclides are divided by the theoretical yield of Ce-144. Those ratios are then normalized by multiplying by the actual activity of Ce-144 present in the sample decay corrected to T=0. That "T=0 mix" of radionuclides is then decay corrected to the estimated age of the fuel fragment.

The identification of Ce-144 in a sample is considered an indication that unidentified pure beta emitting species are also present. If Ce-144 is not identified, no additional radionuclides are added.

Ruthenium-106 is a noteworthy nuclide. It is observed that Ru-106 and its daughter product Rh-106 can exist without other fission products. When $UO_2$ fuel undergoes fission, oxygen becomes available inside the fuel structure. Since the temperature in the center of the fuel is hotter than that toward the edges, the oxygen will migrate out, following the temperature gradient. As it migrates it forms compounds (oxidizes) with fission products. Ruthenium is one element that forms those compounds easily and

Table I: Expected Percentage of Selected Fission Products After Shutdown

| Radionuclide | T=0 | 30 Days | 60 Days | 100 Days | 200 Days | 1 Year | 5 Years |
|---|---|---|---|---|---|---|---|
| Sr-89 | 7.6 | 9.0 | 8.6 | 7.1 | 3.5 | 0.7 | --- |
| Sr-90 | 0.3 | 0.5 | 0.7 | 1.0 | 2.0 | 3.8 | 27.1 |
| Y-90 | 0.3 | 0.5 | 0.7 | 1.0 | 2.0 | 3.8 | 27.1 |
| Y-91 | 9.7 | 12.1 | 12.3 | 12.2 | 6.5 | 1.7 | --- |
| Zr-95 | 9.8 | 12.6 | 13.1 | 12.2 | 8.0 | 2.5 | --- |
| Nb-95 | 9.6 | 16.0 | 19.6 | 20.8 | 16.0 | 6.3 | --- |
| Ru-103 | 6.2 | 6.5 | 5.5 | 3.9 | 1.3 | 0.1 | --- |
| Ru-106 | 0.4 | 0.7 | 1.0 | 1.3 | 2.1 | 2.9 | 1.5 |
| Rh-106 | 0.4 | 0.7 | 1.0 | 1.3 | 2.1 | 2.9 | 1.5 |
| Cs-137 | 0.2 | 0.4 | 0.6 | 0.8 | 1.5 | 2.8 | 1.3 |
| Ba-137 | 0.2 | 0.4 | 0.5 | 0.7 | 1.4 | 2.7 | 0.2 |
| Ba-140 | 10.3 | 3.6 | 1.0 | 0.2 | --- | | |
| La-140 | 10.3 | 4.1 | 1.2 | 0.2 | --- | | |
| Ce-141 | 9.5 | 8.9 | 6.8 | 4.1 | 0.9 | --- | |
| Pr-143 | 9.0 | 3.5 | 1.1 | 0.2 | --- | | |
| Ce-144 | 5.3 | 8.8 | 11.8 | 15.3 | 23.2 | 29.5 | 6.6 |
| Pr-144 | 5.3 | 8.8 | 11.8 | 15.3 | 23.2 | 29.5 | 6.6 |
| Nd-147 | 4.3 | 1.2 | 0.3 | --- | | | |
| Pm-147 | 1.0 | 1.8 | 2.5 | 3.5 | 6.3 | 10.6 | 28.1 |
| % Remaining Activity: | 100 | 56 | 39 | 27 | 14 | 7.5 | 0.9 |

becomes $RuO_4$. $RuO_4$ is very volatile (melting point of 25.5 degrees celsius) and may escape into the coolant through cladding defects.

Promethium-147 is another noteworthy radionuclide. Initially, its activity increases with time to account for ingrowth due to the decay of Nd-147.

This dating method has been demonstrated to provide an acceptable estimate of fuel fragment age. Several fuel fragment contaminants have been sent off-site for vendor analysis. Reference 5 describes the excellent agreement between the mixture of radionuclides predicted by this method and the actual contents determined by those off-site laboratories.

## SKIN DOSE CALCULATION

With an acceptable estimate of the particle's activity and the radionuclides present, potential dose to the skin can be calculated. Current regulation requires that the dose from the presence of a point source contaminant be calculated at a skin depth of 7 mg/cm$^2$ and averaged over a 1

$cm^2$ area. Several dose calculation methods are available to perform that task. The most widely used method is the VARSKIN computer code.

Before VARSKIN was available for general use, skin dose calculation options were more limited. Loevinger's Equation was the only analytical method available and San Onofre used it extensively. Though never intended for use in such situations, proper application of that equation with appropriate variable and parameter values produces acceptable skin dose results. Table II shows the reasonable agreement between Loevinger's approach and VARSKIN. Notice that a third column of values is also included. Those values were calculated using a preliminary version of an EPRI developed skin dose calculation program. Those values are supplied for comparison only.

San Onofre now uses VARSKIN for calculating the skin dose associated with all contamination incidents.

Exposure Variables

Beyond exposure time and particle activity, two other variables are important when the contaminant is located on an item of clothing. Those factors are movement and attenuation.

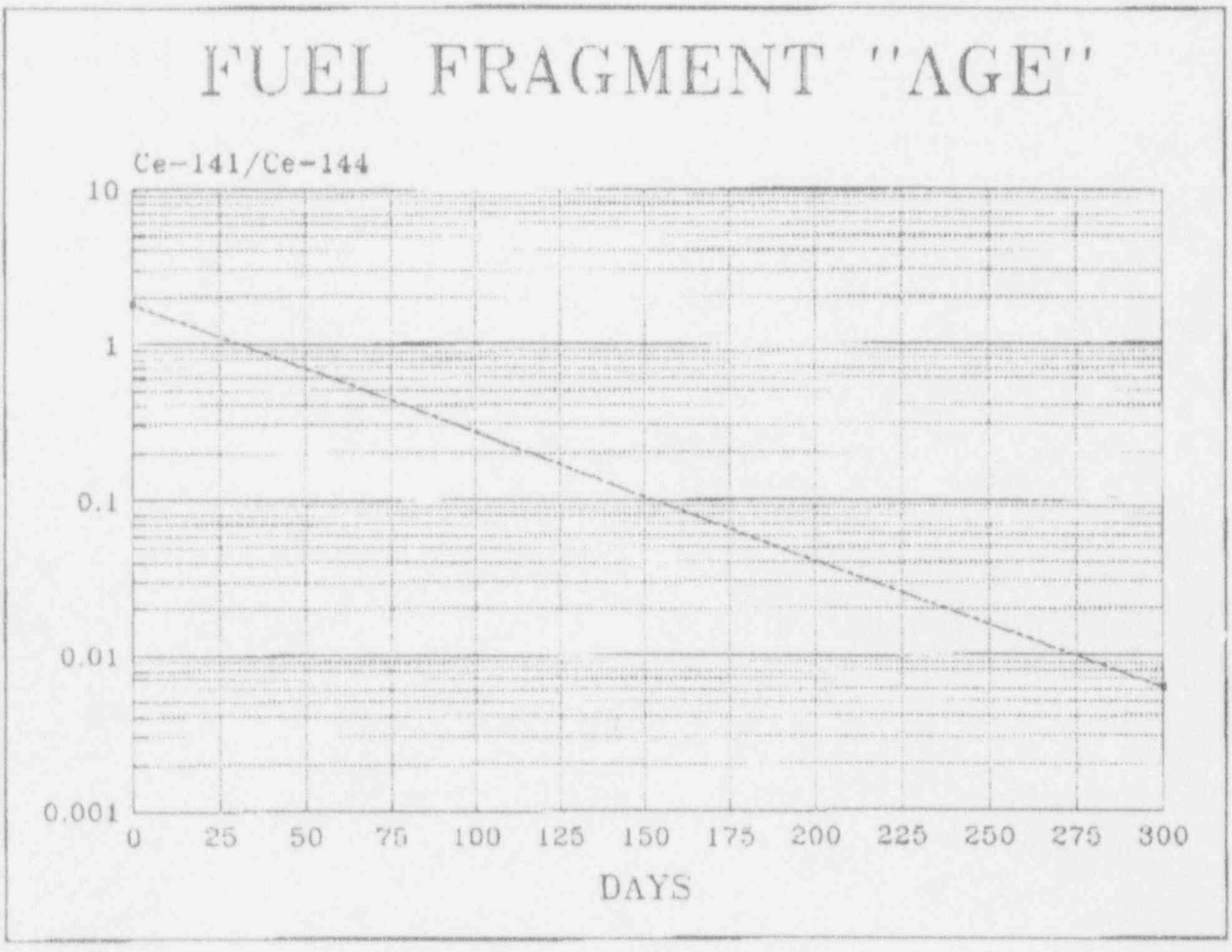


Figure 1: Fuel Fragment "Age"

The typical calculation assumes that the contaminant was stationary during the exposure period. Naturally, in such cases the most highly exposed one square centimeter of tissue is that centered directly beneath the particle. However, when the contaminant is located on clothing, normal movement will result in "spreading" the dose over a larger area. The effect is analogous to a stationary non-point source contaminant with equal total activity that physically covers an area of skin. When VARSKIN prompts the user for source dimensions, one simply enters the area over which the particle moved. (Reference 5 suggests a method for determining the appropriate area to use in such cases.)

The attenuation provided by typical clothing is relatively insignificant with respect to the high energy beta rays emitted by fuel fragment contaminants. However, the physical distance between the particle and the skin provided by the clothing item is a significant factor. VARSKIN prompts the user to enter the depth at which the dose is to be calculated. One can simply enter the sum of the clothing item's density thickness and the fixed 7 mg/cm$^2$ skin depth. Notice, however, that a conservative result is obtained since VARSKIN assumes a constant tissue density and most clothing is less dense than that.

Gamma Exposure

It should be noted that VARSKIN and Loevinger's Equation consider only beta exposure and do not include possible dose due to the gamma component. A separate calculation can be performed to evaluate gamma dose, however, that calculation is normally not necessary. In the case of a typical fuel fragment, gamma dose contributes less than 1 percent to the total dose.

There are instances, however, when gamma exposure is limiting. In cases involving rather weak beta emitters and significant attenuation, dose from the gamma component must be considered. For instance, gamma radiation accounts for 50 % of the skin dose from a cobalt particle separated from the skin by about 28 mg/cm$^2$; that thickness is less than the combination of cotton coveralls over an under garment.

Table II: Comparison of Calculation Methods

| Nuclide | (mrad/μCi-hr) VARSKIN | Loevinger | % Difference |
|---|---|---|---|
| Co-58 | 1190 | 1190 | 1.03 |
| Co-60 | 4130 | 4130 | 1.04 |
| Sr-89 | 8710 | 7270 | 1.00 |
| Sr-90 | 6760 | 6770 | 1.10 |
| Ru-106 | 0 | 0 | 1.00 |
| Rh-106 | 9420 | 6450 | 0.76 |
| Ce-144 | 3240 | 3250 | 1.13 |
| Pr-144 | 9410 | 6700 | 0.78 |
| Totals: | 42860 | 40090 | 0.94 |

## FIELD MEASUREMENT

When a point source contaminant is encountered in the work place, the technician should be equipped to characterize it as either a fuel

fragment or as a lower energy "crud" or cobalt particle. When found during area or job coverage survey, that characterization is necessary to establish appropriate radiological controls. When found on a worker, that characterization allows an upper bound estimate of potential skin dose.

Particle Typing

Upon discovery of a suspected hot particle, the technician must first ensure that it is a true point source. That can be accomplished by observing the count rate while interposing a metal shield with a small hole in it between the contamination and detector.

Once the contaminant is established as a point source, it is necessary to determine whether or not it is a fuel fragment. Since typical PWR contamination has an effective maximum energy that is about one tenth that of a fuel fragment, selective discrimination by beta energy provides the best means to distinguish the two. An 80 to 100 mg/cm$^2$ shield stops essentially all of the beta radiation from ordinary contamination but allows the transmission of nearly two thirds of the beta radiation from fuel fragments.

Laminated cards which approximate 80 mg/cm$^2$, and incidently have hot particle "thumb rules" printed on them, have been supplied to the technicians. By interposing that card between the particle and detector, fuel fragments are readily distinguished from "crud." Specifically, if the count rate when using a frisker is reduced by 50% or less when the card is inserted, the contaminant is a fuel fragment. If it is reduced by more than 90%, the contaminant is likely a cobalt particle. Those same "rules" hold for ion chamber instruments like the Eberline RO-2.

An additional method makes use of an energy compensated GM probe such as the Eberline HP-270. That probe has an open window thickness of 30 mg/cm$^2$ and with the window closed it is 1000 mg/cm$^2$.

Open/closed window ratios greater than 10:1 are obtained with fuel fragments. Ratios less than 5:1 indicate the presence of cobalt or crud contamination.

Activity and Dose Rate

Unfortunately, it is not possible to make a survey instrument measurement in contact with a hot particle and directly read the dose rate. The "contact reading" is not the same as the "true surface dose rate." Those instrument indications must be corrected for geometry and energy effects.

The reading obtained with the instrument on contact with a surface actually gives the "dose rate" at the center of the chamber, not at the surface of the source. The center of the chamber on an RO-2, for instance is more than 2 centimeters from the surface. The "geometry correction factor" for point sources at relatively short distances is significant at approximately 50.

The high energy of the beta radiation from fuel fragments is probably their most noteworthy characteristic. However, that spectrum still contains a low energy beta particle component. Those lower energy beta particles can be significantly attenuated before reaching the chamber center, requiring an "energy correction factor" of approximately 4.

The overall correction factor is the product of the two correction factors, or about 200.