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COMPARISON OF OBSERVED AND PREDICTED KR-85 AIR CONCENTRATIONS*

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ABSTRACT

A computer code, ANEMOS has been written to estimate concentrations in air and ground deposition rates for Atmospheric Nuclides Emitted from Multiple Operation Sources. This code uses a modified Gaussian plume equation. Output from ANEMOS includes annual-average air concentrations and ground deposition rates of dispersed radionuclides and daughters. To use the environmental transport model properly, some estimate of the models predictive accuracy must be obtained. To validate the ANEMOS model, one year of weekly average Kr-85 concentrations observed at 13 stations located 28 to 144 km distant from continuous point source at the Savannah River Plant (SRP), Aiken, South Carolina, have been used. There was a general tendency for the model to underpredict the observed air concentrations slightly. Pearson's correlation between pairs of logarithms of observed and predicted annual-average values was r=0.84. The monthly results tend to show more scatter than do either the seasonal or the annual comparisons.

1. INTRODUCTION

The ANEMOS [1] computer code has been developed at Oak Ridge National Laboratory to estimate concentrations in air and ground deposition rates for Atmospheric Nuclides Emitted from Multiple Operation Sources. ANEMOS is one component of an Integrated Computerized Radiological Risk Investigation System [2] (CRRIS) developed

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for the U.S. Environmental Protection Agency (EPA) for use in determining compliance with the Clean Air Act for radionuclides released from U.S. Nuclear Regulatory Commission-licensed and U.S. Department of Energy facilities.

To use any environmental transport model properly, some estimate of the model's predictive accuracy must be obtained. The best way to determine the accuracy of calculational procedures such as ANEMOS is to compare predictions from the procedure with field measurements taken under release conditions similar to those assumed by the model, a process commonly referred to as model validation. A data set has been made available by the Savannah River Plant (SRP) at Aiken, South Carolina, that makes it possible to perform a validation study of ANEMOS out to a downwind distance off 144 km [3,4]. The purpose of this paper is to discuss the ANEMOS code and to present the results of a validation study using the SRP data base.

2. ANEMOS CODE

ANEMOS estimates concentrations in air and ground deposition rates for Atmospheric Nuclides Emitted from Multiple Operating Sources. ANEMOS is intended to calculate the average concentrations around a source for releases that occur over an extended period of time such as a year. A schematic representation of the ANEMOS code is shown in Figure 1.

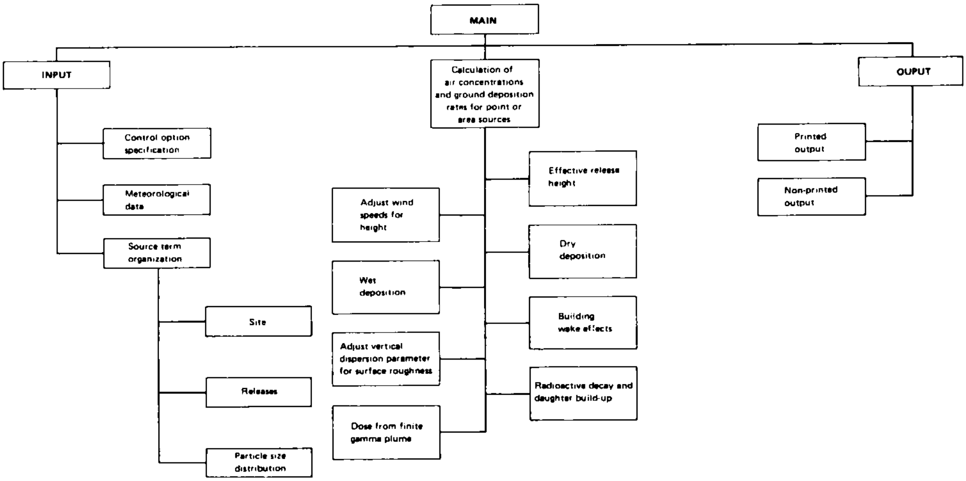

Fig. 1. A schematic representation of the ANEMOS computer code.

Discharges of radionuclides to the atmosphere result in doses to man from inhalation of an immersion in contaminated air, exposure to contaminated ground sources, and ingestion of food-stuffs contaminated by deposited radionuclides.

The calculations made in ANEMOS are based on the use of a modified straight-line Gaussian plume atmospheric dispersion model [5]. Based on the type of long-term meteorological data available for many Unites States cities, a 22.5° sector-averaged air concentration is calculated:

$$C_{is}(x,\theta) = \left[\sum_{p=1}^{N_s} \sum_{r=1}^{N_w} \frac{2.55\ fpr(0)\ Q'_{iprs}(x)}{x\ \bar{u}_r} \right] G(z,\ h_e,\ L_p,\ x) \qquad (1)$$

where

$C_{is}(x,\theta)$ = ground-level air concentration of radionuclide i with particle size s in wind direction θ ($1 \leq \theta \leq 16$) at downwind distance x (activity/m^3);

$Q'_{iprs}(x)$ = effective emission rate for radionuclide i with particle size s in stability class p, wind speed class r, and wind direction θ at downwind distance x (activity/s);

$f_{pr}(\theta)$ = fraction of total release time that wind blows toward direction θ in wind speed class r and stability class p (unitless);

$\bar{u}_r$ = mean wind speed associated with wind speed category r (m/s);

N_s = number of atmospheric stability classes (≤ 8; unitless);

N_w = number of wind speed classes (≤ 8; unitless);

L_p = depth of the atmospheric mixing layer associated with stability class p (m);

h_e = effective height of the plume (m); and

z = the height of the receptor above ground (m).

The value of G (z,h_e,L_p,x) used in Eq. (1) depends on the relationship between z, h_e, and L_p. In the most common case of $z \leq h_e \leq L_p$, and accounting for multiple reflections of the plume between the ground and the top of the mixing layer.

$$G(z,h_e,L_p,x) = \frac{1}{\sqrt{2\pi}\,\sigma_z(p,\,x)} \sum_{m=0}^{n} \left\{ \exp - \frac{1}{2}\left[\frac{2mL_p + h_e - z}{\sigma_z\,(p,\,x)}\right]^2 \right.$$
$$+ \exp - \frac{1}{2}\left[\frac{2mL_p + h_e + z}{\sigma_z\,(p,\,x)}\right]^2$$
$$+ \exp - \frac{1}{2}\left[\frac{2(m+1)\,L_p - h_e - z}{\sigma_z\,(p,x)}\right]^2 \qquad (2)$$
$$\left. + \exp - \frac{1}{2}\left[\frac{2(m+1)\,L_p - h_e + z}{\sigma_z\,(p,\,x)}\right]^2 \right\},$$

where

$m = 0, 1, 2, \ldots n;$

$\sigma_z(p,x)$ = vertical dispersion parameter for stability category p and downwind distance x (m).

When the value of $\sigma_z(p,x)$ becomes greater than one or two times L_p, the radionuclide plume can be assumed to be effectively distributed uniformly in the vertical throughout the mixing layer. Under these circumstances

$$G(z,h_e,L_p,x) = \frac{1}{L_p} \qquad (3)$$

There are times when the relationship $z \leq h_e \leq L_p$ may not hold in the atmosphere for a given assessment problem. If $z < L_p < h_e$ or $h_e < L_p < z$, then

$$G(z,h_e,L_p,x) = 0 \qquad (4)$$

Also, if $L_p \leq z$ and $L_p \leq h_e$, the plume reflects from only a lower face and

$$G(z,h_e,L_p,x) = \frac{1}{\sqrt{2\pi}\sigma_z(p,x)}\left[-\frac{1}{2}\left[\frac{h_e-z}{\sigma_z(p,x)}\right]^2\right]$$
$$+\exp -\frac{1}{2}\left[\frac{-2l_p + h_e + z}{\sigma_z(p,x)}\right]^2 \qquad (5)$$

ANEMOS automatically selects the $G(z,h_e,L_p,x)$ that is most appropriate for a given calculation.

ANEMOS requires as basic meteorological input a joint frequency distribution of wind direction, wind speed class, and atmospheric stability class over the time period of interest in each run. The wind speeds associated with each wind speed class are often measured at 10 m height. These wind speeds can be automatically adjusted by ANEMOS for effective stack heights above 10 m on the basis of a wind profile power law as a function of atmospheric stability class and surface roughness length [6].

The basic set of vertical dispersion coefficient (σ) values used in ANEMOS is that developed by Smith [7] as modified by Hosker [8]. Values of σ are specified as a function of atmospheric stability, downwind distance, and surface roughness length.

The height of the top of the tropospheric mixing layer above ground is considered to be a function of atmospheric stability class. ANEMOS results can be very sensitive to the values chosen for mixing height, especially at downwind distances beyond a few kilometers and when Eqs. (2) and (2) are being used.

Daughter radionuclides may form during downwind travel from released parent radionuclides due to decay processes. In addition, both parent and daughters may be removed from the plume during transport by dry and wet deposition processes. All removal and input processes (including dry and wet deposition and decay and buildup) for all parent and daughter radionuclides are summed and integrated over the time that the plume travels to a given downwind location.

A user of CRRIS may calculate external dose at a location due to a finite cloud containing a gamma- or x-ray-emitting radionuclide passing overhead. This calculation is made on the basis of the methodology presented by Healy and Baker [9] and incorporated into Nuclear Regulatory Commission (NRC) Regulatory Guide 1.109 [10] for use with noble gas releases from stacks greater than 80 m in height. Once the finite plume dose has been calculated, it is divided by the appropriate dose conversion factor to get an "effective" air concentration. This "effective" air concentration is transmitted to ANDROS [11] for subsequent dose and health risk calculations from the overhead cloud.

The output of ANEMOS is presented for 16 sectors of a circular grid with nuclide-specific air concentrations and ground deposition rates. ANEMOS can calculate both the sector-average concentrations and deposition rates at a given set of downwind distances in each sector, and the average of these quantities over an area within each sector bounded by two successive downwind distances.

3. THE SRP DATA BASE

The SRP is a major production facility of the U.S. Department of Energy (DOE). The SRP includes a nuclear fuel manufacturing facility, three production reactors, two chemical separation plants, a heavy water production plant, and various waste management activities. These facilities are located on a 770 km^2 site south of Aiken, South Carolina. The terrain within 150 km of the SRP is gently rolling hills ranging in elevations from 150 m above sea level to the northwest to about 25 m toward the southeast. The SRP is covered with mixed hardwood and pine forests; the surrounding area consists of mixed forests and clear land [3].

Fission product Kr-85 is released as a nonbuoyant plume at a height of 62 m during dissolution of irradiated fuel. Kr-85 is an inert gas with a (10.76 years) radioactive half-life. Therefore, it can be used as a tracer of atmospheric dispersion processes without the complicating effects of wet deposition, dry deposition, and chemical transformation.

Kr-85 air concentration measurements began in March 1975 and continued through September 1977 at 13 stations surrounding SRP. These stations are shown in Fig. 2 [4]. Cryogenic air samplers were used to collect the Kr-85 for laboratory processing and counting. The sampling stations ranged in distances from 28 to 144 km from the release point.

Meteorological data for the years of the Kr-85 Savannah River experiments are available. There are separate meteorological data bases for hourly surface weather observations, twice daily rawinsonde observations, and hourly-average meteorological tower observation. In addition to these meteorological data, the on-site 62 m meteorological tower data and an acoustic sounder located at the SRP have been used to compile wind rose statistics representative of the source area and vertical mixing characteristics of the lower atmosphere, respectively.

4. METHODOLOGY

Separate ANEMOS simulations of Kr-85 transport were performed for each study period from September 1975 through August 1976 from the SRP data base.

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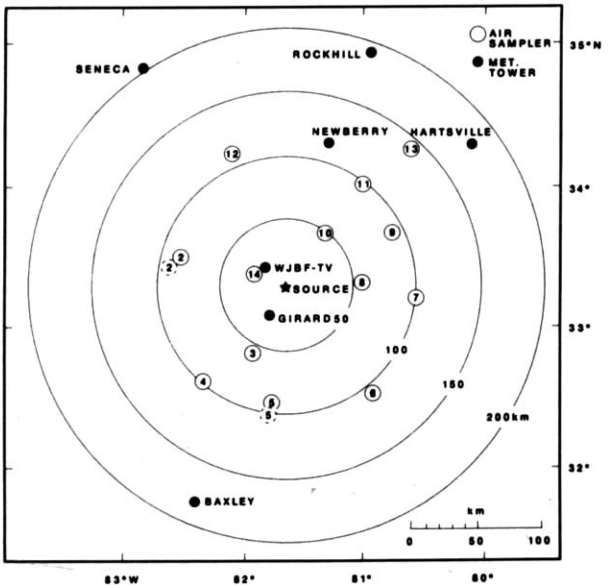

Fig. 2. Kr-85 cryogenic air sampling stations, meteorological towers and surface weather stations within 200 km of the SRP Source [4].

Study periods of interest were twelve months, four seasons, and one annual value. Model parameters were chosen appropriate to release conditions at the SRP. The source term was assumed to be a steady atmospheric release from a single point source (stack). Values of simulated ground-level Kr-85 air concentrations were computed for each of the 13 locations corresponding to the monitoring stations specified in the SRP data base.

Comparisons were made between measured values of Kr-85 air concentrations and values of Kr-85 air concentrations predicted for each of the stations and time periods considered in this study using the statistical analysis package SAS 79 [13]. A background concentration of 14 pCi/m^3 was subtracted from the measured concentrations at each station to derive the "observed" values used in these comparisons. The tendency of ANEMOS either to overpredict or underpredict was evaluated by examining the values of the ratio of predicted to observed air concentrations. The frequency distribution, median and geometric standard deviation of predicted-to-observed (P/O) ratios, and the slope, intercept, and Pearson's correlation for the regression of the log of the predicted air concentrations versus the log of the observed air concentrations have been computed for the annual, seasonal and monthly time periods. Also, Test of Significance [14] and the Reliability Index [15] have been applied to our results.

5. RESULTS

Table 1 shows a station-by-station comparison of the observed and predicted annual average Kr-85 air concentrations. Table 2 shows the frequency distribution of the comparison of predicted to observed ratios for the annual, seasonal and monthly air concentrations of Kr-85. A plot of log-predicted versus log-observed ground-level concentrations is given in Fig. 3. Given in Table 3 for each time period considered are the median and geometric standard deviation of the P/O values and the slope, intercept and correlation coefficient for the regression of the log of the observed air concentrations versus the log of the observed air concentrations. The results of the Test of Significance are also given in Table 3. The results show that the annual predicted ground-level Kr-85 concentrations exceed the observed values for 7 of the 13 stations. The predicted-to-observed concentration ratio was less than 1 in 6 of the 13 cases. Also this ratio was less than 0.5 for 1 of the 13 stations.

It is assumed that the observed air concentrations at a given location are lognormally distributed. The Reliability Index is given in Ref. [15].

One may use either of the following formulas:

$$k_g = \frac{1+\sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{x_i-y_i}{x_i+y_i}\right)^2}}{1-\sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{x_i-y_i}{x_i+y_i}\right)^2}} \qquad (6)$$

or,

Table 1. Comparisons of observed and predicted annual average air concentrations of Kr-85 at the SRP, September 1975 through August 1976.

Monitoring station	Distance from source (km)	Kr-85 Concentrations (p/Ci/m^3) Observed	Predicted	Predicted / Observed
2	94	8.6	10.0	1.16
3	60	12.0	21.1	1.76
4	99	15.0	16.1	1.07
5	98	5.4	7.7	1.43
6	109	10.0	11.2	1.12
7	100	21.0	19.4	0.92
8	57	51.0	38.2	0.75
9	93	32.0	17.5	0.55
10	50	73.0	35.9	0.49
11	98	25.0	15.3	0.61
12	112	10.0	16.9	1.69
13	144	14.0	9.9	0.71
14	28	38.0	47.1	1.24

$$ks = \exp\sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\ln\frac{y_i}{x_i}\right)^2} \qquad (7)$$

where:

kg = the geometrically based index

ks = the statistical reliability index

x_i = ith model prediction

y_i = observations corresponding to x_i

n = number of pairs (x_i, y_i)

From Eq. [6] and Eq. [7] the values kg = 1.473 and ks = 1.497 are found. These results indicate that the model is accurate within a factor of about 1.5, in the sense described in Ref. [15]. Since the samples are from lognormal distributions in Ref. [15], this means that, within one standard deviation (that is about 68% of the time) the prediction will be within a factor of 1.5 of the observation ($\frac{x_i}{y_i} < 1.5$ and $\frac{y_i}{x_i} < 1.5$, 68% of the time).

The frequency distribution of the comparison of predicted and observed annual, quarterly and monthly concentrations of Kr-85 is

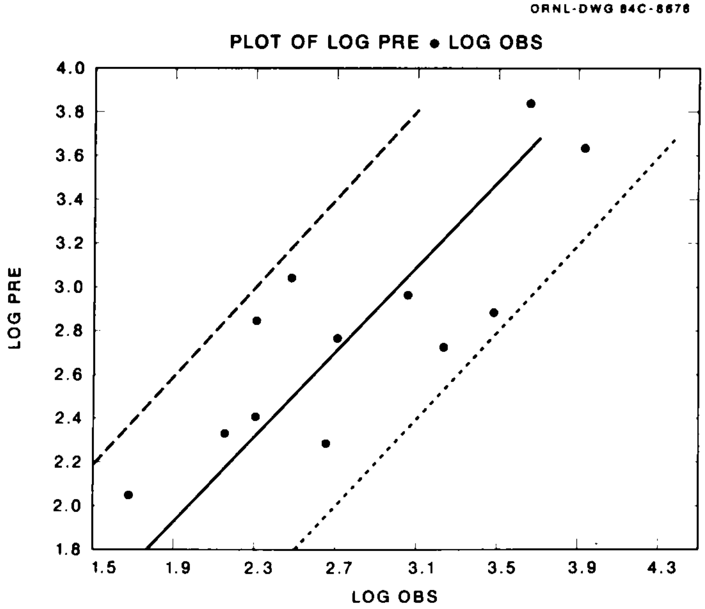

Fig. 3. Comparison log-predicted and log-observed annual Kr-85 ground-level concentrations at SRP monitoring stations. Values are plotted in units of pCi/m^3. The solid line implies prediction = observation; dashed line, prediction = 0.1 x observation; and broken line, prediction = 2 x observation.

given in Table 2. The monthly results tend to show more scatter than do either the seasonal or the annual comparisons. In other words, the results tend to be more scattered as the averaging time being considered decreases.

The statistical analysis of comparisons between predicted and observed annual, quarterly, and monthly air concentrations of Kr-85 are given in Table 3. From the Test of Significance it is found that the correlation coefficient for the long term periods considered in this study all significant at $p < 0.01$ level. The correlation coefficients tend to be more insignificant as the averaging time being considered decreases. Annual and quarterly values give very high correlation coefficients. For example, annual correlation coefficient is 0.84 and summer correlation coefficient is 0.95. If we look at the monthly results, three months (September, November and December) are not significant at $p < 0.01$ and give very low correlation coefficients.

The median ratio of predicted-to-observed values indicates a slight tendency towards underpredictions. For example, the underprediction of annual values is less than one order of magnitude (Median = 0.96). There are some overpredictions in our analysis; however, they were always less than one order of magnitude. For example, the median ratios of September 1975 and October 1976 are 4.63 and 6.49, respectively. The geometric standard deviation of

Table 2. The frequency distribution of the comparison of predicted and observed annual quarterly and monthly air concentrations of Kr-85 at the SRP, September 1975 through August 1976.

	Frequency of ratio (P/O)[a]					
	≤0.1	0.1-0.5	0.5-1	1-2	2-10	≤10
ANNUAL						
Sep. 1975-Aug. 1976	0	1	5	7	0	0
QUARTERLY						
Sep.-Nov. 1975	0	0	0	5	7	0
Dec. 1975-Feb. 1976	0	4	5	2	0	0
Mar.-May 1976	0	2	4	5	0	0
June-Aug. 1976	0	2	5	3	2	0
MONTHLY						
Sep. 1975	0	0	0	5	3	5
Oct. 1975	0	0	0	2	8	3
Nov. 1975	0	1	7	1	1	1
Dec. 1975	0	6	4	2	1	0
Jan. 1976	0	3	7	1	2	0
Feb. 1976	0	0	5	4	3	1
Mar. 1976	0	9	1	1	1	0
Apr. 1976	0	0	7	4	1	0
May 1976	0	0	3	4	6	0
June 1976	0	3	4	4	1	0
July 1976	0	1	4	3	0	1
Aug. 1976	0	1	2	3	6	1

[a] A value of 1 signifies a perfect prediction.

Table 3. The statistical analysis of comparisons between predicted and observed annual, quarterly and monthly air concentrations of Kr-85 at the SRP, September 1975 through August 1976.

	Median	Geometric Standard Deviation	Intercept[b]	Slope[b]	Correlation Coefficient[b]
ANNUAL					
Sept.1975-Aug. 1976	0.96	1.52	-0.43	1.16	0.84*
QUARTELY					
Sep.-Nov. 1975	2.26	1.59	-0.81	1.00	0.80*
Dec. 1975-Feb. 1976	0.64	1.52	-0.98	1.44	0.90*
Mar.-May 1976	0.85	1.57	-0.45	1.22	0.88*
June-Aug. 1976	1.04	1.80	-2.00	1.69	0.95*
MONTHLY					
Sep. 1975	4.63	3.39	1.49	-0.07	0.07
Oct. 1975	6.49	2.61	-2.29	1.13	0.65*
Nov. 1975	0.94	2.92	1.00	0.63	0.49
Dec. 1975	0.61	2.36	2.61	0.32	0.35
Jan. 1976	0.72	2.01	0.74	0.84	0.64*
Feb. 1976	0.86	2.40	-0.49	1.24	0.80*
Mar. 1976	0.44	2.12	0.76	1.02	0.76*
Apr. 1976	0.96	1.61	-0.51	1.20	0.85*
May 1976	1.86	2.12	-0.85	1.08	0.81*
June 1976	0.82	1.87	-0.98	1.44	0.90*
July 1976	1.08	2.60	-0.63	1.18	0.82*
Aug. 1976	2.03	2.77	-1.83	1.40	0.69*

[b] For a perfect fit, slope=1, intercept=0, and correlation coefficient=1.

*Signigicant at $p < 0.01$.

predicted-to-observed ratios are lower for long time periods, such as months, considered in this study. Also, the regression of the log of prediction versus log of observation is closer to a perfect fit for the long time periods than any of the other short time periods considered in this study.

6. DISCUSSION

The comparisons presented here assume that Kr-85 is emitted by SRP in a continuous manner when, in fact, it is emitted intermittently. Because of the relatively long averaging times considered in this study, and since there were no long shutdown periods suggested in the monthly emission data, this assumption should not be critical to the conclusions of the study. These results also assume no significant problems with the cryogenic Kr-85 sampling system or with the meteorological data acquisition system.

A more critical problem is the selection of a value for the limit to vertical mixing, or lid height. Simulated ground-level air-concentration values at mesoscale distances are quite dependent on this parameter. For distances at which Eq. (3) is used, computed concentration is an inverse function of lid height. Furthermore, the effective limit to vertical mixing may be much higher than the classical lid height. For example, convective activity may serve to remove material from the lower layers of the troposphere [16].

These results show the importance of considering averaging times when discussing the accuracy of Gaussian plume model air concentration predictions. The monthly comparisons were generally less accurate than the quarterly comparisons which were, in turn, generally less accurate than the annual average comparison. This decrease in accuracy with decrease in averaging time has been demonstrated previously for the Gaussian model [17,18]. This trend has been attributed to the less uniform distribution of wind direction within a sector for the shorter averaging times [17].

No attempt has been to judge the "acceptability" of the accuracy of results presented here. For example, are the annual average predictions too conservative, not conservative enough, or acceptable as presents? Only the user of these predictions should make this judgment.

7. CONCLUSIONS

Comparisons between predicted and observed ground-level Kr-85 air concentrations have been made by using the ANEMOS computer code. Comparisons were made for annual, seasonal, and monthly time periods.

There was a general tendency for the model to underpredict slightly the observed air concentrations for the time periods considered. The results for long term periods were more accurate than the results for short term periods, such as months. In other words, the general accuracy of the results tended to decrease as the averaging time being considered decreased.

The results of model validation studies such as this one should be considered whenever ANEMOS or similar computer codes are used to determine compliance with EPA radionuclide emission standards or applied to other radiological assessment problems. The acceptability of the model accuracy indicated by such studies must be determined by the model user on the basis of the use to which the model is being applied.

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