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**COMPARISON OF AIRDOS-EPA PREDICTIONS
OF GROUND-LEVEL AIRBORNE RADIONUCLIDE
CONCENTRATIONS TO MEASURED VALUES**

FINAL REPORT

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CHAPTER 1:

INTRODUCTION

The AIRDOS-EPA computer code (reference 1) was developed at the Oak Ridge National Laboratory for use by the Environmental Protection Agency (EPA). The code is used to estimate environmental concentrations of radionuclides emitted to the atmosphere, and is commonly used by the EPA for analysis of the dose-reduction benefits resulting from regulations which govern such emissions.

AIRDOS-EPA uses annual average meteorological data coupled with a Gaussian plume model to estimate annual average ground-level concentrations at user-specified distances and directions from a continuous source. The plume is dispersed horizontally and vertically as it is blown downwind. The code can account for depletion of pollutants (radionuclides) in the plume by radioactive decay, gravitational fall, and scavenging and dry deposition processes.

The meteorological data consist principally of the annual frequency of wind direction, the annual frequency of each Pasquill stability category, and the annual average wind speed for each direction and stability category. The code allows for buoyant or momentum plume rise, but not both simultaneously. However, the simultaneous existence of both buoyant and momentum driven plume rise can be approximated by the user. The air concentrations are average values in the cross-wind direction over each of sixteen 22.5 degree sectors.

This study was a continuation of the evaluation of the AIRDOS-EPA code reported in "Comparison of AIRDOS-EPA Predictions of Ground-Level Radionuclide Concentrations to Measured Values at Five Facilities" (reference 2). In the original analysis, the results from five sites indicated that the code's predictions range from a factor of five on the low side to a factor of two on the high side at the 50 percent confidence level. Due to the small number of sites evaluated, the 95 percent confidence interval is significantly larger, ranging from a factor of 15 on the low side to a factor of six on the high side. The purpose of this study was to obtain results for additional sites in order to reduce these uncertainties.

As in the initial study, the accuracy and precision of the code was analyzed by comparing predicted ground-level concentrations to actual environmental measurements at five sites. The criteria for site selection were the same as used in the previous study. However, as we examined the candidate sites, it became clear that not only were there no "ideal sites" (see reference 2), but that the number of good "real world" sites was also very limited. While the sites selected for this comparison were those that most closely satisfied the selection criteria, some of them have characteristics which push AIRDOS-EPA to or even beyond its limits. Thus, while the initial study was a comparison at the "best" real world sites, this study may be thought of as comparison at "good" real world sites. It should be noted that although this comparison includes sites with less than ideal characteristics, our studies still do not provide a complete measure of the code's ability to estimate radionuclide concentrations at "typical" real world sites. To make such an estimate, we would have chosen the sites at random and not on the basis of their being the easiest to model.

For this study, the comparisons were made using data from two Department of Energy (DOE) facilities, two nuclear fuel-cycle facilities licensed by the Nuclear Regulatory Commission (NRC), and a fossil-fuel electric generating station. The DOE facilities, which release tritium, are the Hanford Reservation in Washington and the Brookhaven National Laboratory on Long Island, New York. In addition, krypton-85 is released at the Hanford Reservation. For both DOE sites, on-site meteorological data were used in the assessments. The NRC-licensed sites are Allied Chemical Company's uranium hexafluoride plant in Illinois and Nuclear Fuel Services' fuel fabrication plant in Tennessee. The Allied plant releases natural uranium and the Nuclear Fuel Services plant releases uranium of various enrichments. Neither of these facilities measures meteorological parameters on-site. Both present difficulties in modeling due to the number of release points at the Allied plant and the complex terrain in the vicinity of the Nuclear Fuel Services plant. The fossil-fuel electric generating station is located in the eastern United States. The facility emits sulphur dioxide. Although it lacks on-site meteorological measurements, meteorological data are available from a nearby airport. Each of the sites is described in more detail below.

Despite our efforts to select the best sites for this comparison, useful data were obtained from only two of the five sites: Allied Chemical Company's uranium hexafluoride plant and the Hanford Reservation. At the other sites, deficiencies in the environmental monitoring program and/or releases too small to be distinguished from

background by environmental measurement techniques rendered the comparisons of predicted versus measured concentrations meaningless. As a result, the evaluation of the precision and accuracy of the code was assessed using four of the data sets obtained from the first comparison study and the two meaningful data sets obtained in this comparison.

CHAPTER 2:

METHODOLOGY

Our procedures for obtaining measured and predicted concentrations are given below, together with a description of the AIRDOS-EPA input parameters and their uncertainties.

PREPAR (reference 3) is a complementary code to AIRDOS-EPA. Its purpose is to generate much of the input used by AIRDOS-EPA. We used PREPAR for this purpose, and the input parameters obtained from PREPAR are identified for each site along with those obtained from other sources.

ALLIED CHEMICAL COMPANY UF_6 PLANT

The Allied Chemical Company's uranium hexafluoride (UF_6) plant is located in Metropolis, Illinois on the north bank of the Ohio River, across from Paducah, Kentucky. The plant is located in a rural area where the terrain consists of gently rolling hills. The Allied plant was one of the five sites analyzed in the initial study. Running a second evaluation of the site using data from a different year serves to confirm or deny the validity of the initial results. For this study, we used data from the year 1980.

During the production of UF_6 , small amounts of natural uranium escape from the plant, and concentrations of uranium in the air are routinely measured as a part of the environmental monitoring program. Reference 4 contains data on both the release rates and the measured atmospheric concentrations for years 1979 through 1981. The measured concentrations of uranium for 1980 at the seven measurement locations are compared to our AIRDOS-EPA predictions in Table 1. To facilitate comparison, we have reproduced the results for 1981 obtained in the initial study.

There are two principal uncertainties in the predicted values. The releases of uranium from the Allied plant are from multiple stacks and vents, most of which are located on the feed material building. As the stacks that are not located on the feed materials

TABLE 1:

COMPARISON OF MEASURED AND PREDICTED AIRBORNE URANIUM*
CONCENTRATIONS AROUND THE ALLIED CHEMICAL COMPANY'S UF₆ PLANT

— 1980 —

Station	Direction	Distance, km	Concentration, pCi/m ³		Ratio Meas/Pred
			Measured	Predicted	
9	NNW	0.24	1.5E-2	1.7E-2	0.9
10	SW	0.22	4.0E-2	1.8E-2	2
12	SSE	0.18	3.7E-2	3.4E-2	1
13	NE	0.23	4.4E-2	2.2E-2	2
6	NNE	1.62	3.5E-3	3.4E-3	1
8	NE	0.32	2.1E-2	1.8E-2	1
11	N	0.23	2.9E-2	3.9E-2	0.7

— 1981** —

Station	Direction	Distance, km	Concentration, pCi/m ³		Ratio Meas/Pred
			Measured	Predicted	
9	NNW	0.24	1.0E-2	2.9E-2	0.3
10	SW	0.22	2.5E-2	2.3E-2	1
12	SSE	0.18	2.6E-2	4.1E-2	0.6
13	NE	0.23	3.4E-2	3.2E-2	1
6	NNE	1.62	2.5E-3	4.0E-3	0.6
8	NE	0.32	1.4E-2	2.5E-2	0.6
11	N	0.23	1.6E-2	5.0E-2	0.3

* natural uranium

** 1981 data reproduced from reference 2

building emit a trivial fraction of the total release, they were co-located with those on the feed materials building in the assessment. Most of the uncertainty lies in the treatment of the release point elevations, which vary from 3.6 to 47 meters. Further, about 60 percent of the stacks discharge horizontally and 40 percent discharge vertically.

AIRDOS-EPA allows a maximum of six stacks. As was done in the initial study, we grouped the stacks into six intervals and computed a release height for each interval based on the magnitude of the release from each stack in the interval. The assigned release heights are given in Table 2. Because many of the stacks and vents discharge horizontally and because of the possible downdraft effects from the building (which is 90 feet high, 60 feet wide, and 100 feet long), we neglected any plume rise from momentum or temperature effects.

The meteorology represents another source of uncertainty. There have been no on-site or close-by meteorological data available since 1964 when the weather station at Paducah, Kentucky, discontinued its measurements. Therefore, we used meteorological data from Evansville, Illinois, about 70 miles northeast of the site. The Evansville data were obtained from the National Climatic Data Center. Our comparison of the meteorological data from Evansville with that obtained from Paducah (see reference 2) indicates that the Evansville data are appropriate for the Allied plant.

The emissions data are based on stack samples which were obtained using an isokinetic probe and 0.6 to 0.8 micrometer membrane filters. Where moisture or chemical attack preclude the use of membrane filters, combination scrubber-mist impingers were used. Stack samples were collected once or twice a day. The samples were dissolved in nitric acid and analyzed fluorometrically for uranium. The releases are reported in terms of kg/yr of natural uranium. We used a specific activity of $6.77E-7$ Ci/gm for natural uranium to convert the release rates from mass to activity.

Environmental concentrations were measured by means of 2 cfm sampling devices which use a cascade impactor and a series of filters. The filters, which are changed weekly, are dissolved in nitric acid and analyzed flurometrically for uranium. At the minimum concentrations reported, on the order of 0.2 mg of uranium is collected per sampler per week. For the 25 percent aliquot used, this concentration is well above the expected nanogram sensitivity for fluorometry. The reported minimum detectable level

TABLE 2:

AIRDOS-EPA INPUT FOR ALLIED CHEMICAL COMPANY
 ATMOSPHERIC DISPERSION CALCULATIONS

1. Stack Parameters:

	<u>Stack 1</u>	<u>Stack 2</u>	<u>Stack 3</u>	<u>Stack 4</u>	<u>Stack 5</u>	<u>Stack 6</u>
Height, m	4.0	6.2	13.1	18.0	23.0	29.8
Diameter, m	NOT USED FOR ANY STACK					
Velocity, m/sec	NOT USED FOR ANY STACK					
Heat Release Rate, gm-cal/sec	NOT USED FOR ANY STACK					
Natural Uranium Release Rate, Ci/year	4.1E-3	1.1E-2	5.2E-2	2.4E-2	3.4E-2	1.5E-1
2. Height of Lid, m	1000					
3. Rainfall Rate, cm/yr	115					
4. Average Air Temperature, °K	NOT USED					
5. Surface Roughness Height, m	0.01					
6. Height of Wind Measurements, m	10					
7. Scavenging Coefficient	3.8E-6 sec ⁻¹					
8. Deposition Velocity, m/sec	4.5E-4					
9. Settling Velocity, m/sec	0*					

* We estimate the value to be about 0.001 m/sec; however, AIRDOS-EPA sets the settling velocity equal to zero if it is less than 0.01 m/sec.

is $1\text{E-}16$ microcuries/ml. The environmental concentrations are reported in terms of microcuries/cubic centimeter, based on a specific activity of natural uranium of $6.77\text{E-}7$ Ci/gm.

No hard estimates of the sampling errors are available. The quality assurance and control program includes running daily and weekly uranium standards and a monthly calibration. In addition, stack samples are regularly split and run by both Allied and the Nuclear Regulatory Commission's reference laboratory in Idaho. Agreement on these split samples is regularly obtained. Based on the split sample comparisons and discussions with cognizant personnel at the plant, we estimate that the emissions estimates and the environmental measurements have an uncertainty of ± 15 percent.

Table 2 gives the input parameters used in making the comparison between the measured and predicted environmental concentrations. The rainfall rate is from reference 4 and the height of the lid is the average of the winter and summer values from Figure 2.33 in reference 5. The remaining values are default values from reference 3.

BROOKHAVEN NATIONAL LABORATORY

The Brookhaven National Laboratory (BNL) is located on Long Island, about 97 km east of New York City. The site consists of about 2130 hectares, most of which is wooded, except for a developed area of about 655 hectares. The terrain in the vicinity of the plant is gently rolling, and comprises forests, agricultural land, and suburban developments.

BNL is a major Department of Energy research facility, and tritium is among the radionuclides released to the atmosphere from several facilities at the site. The facilities which emit tritium include: the Medical Research Center, the Chemistry Building, the High Flux Beam Reactor, the Van de Graaf Accelerator, and the Linac Isotope Facility. The environmental monitoring program at the site includes sampling of tritium in the releases from these facilities and measurements of the concentrations of tritiated water vapor in air at 18 environmental monitoring locations. The measured concentrations at these locations for 1985 are compared to our AIRDOS-EPA predictions in Table 3.

TABLE 3:

COMPARISON OF MEASURED AND PREDICTED AIRBORNE TRITIUM
CONCENTRATIONS AROUND THE BROOKHAVEN NATIONAL LABORATORY

Station	Direction *	Distance, km *	Concentration, pCi/m ³		Ratio Meas/Pred
			Measured	Predicted	
1	N	2.20	6.0E+0	2.1E-1	30
3	NE	2.60	9.9E+0	4.8E-1	20
4	ENE	2.50	1.0E+1	4.1E-1	20
5	E	2.60	5.8E+0	3.0E-1	20
6(P-7A)	ESE	2.90	5.9E+0	3.4E-1	20
6(P-7B)	ESE	2.90	5.9E+0	3.4E-1	20
7	SE	1.90	7.2E+0	5.1E-1	10
8	SSE	1.30	2.1E+1	9.3E-1	20
9	S	2.20	2.3E+0	5.2E-1	4
10	SSW	1.90	7.9E+0	9.3E-1	9
11(P-4)	SW	2.10	9.1E+0	7.7E-1	10
12	WSW	1.70	4.1E+0	8.5E-1	5
13	W	1.50	6.6E+0	7.3E-1	9
14	WNW	1.40	4.3E+0	4.4E-1	10
15	NW	1.60	1.9E+1	4.7E-1	40
16(P-2)	NNW	2.30	2.1E+0	3.1E-1	7
Control #1	NNW	> 16.00	3.7E+0	3.5E-2	100
Control #2	SW	> 20.00	5.3E+0	4.0E-2	100

* The direction and distances given are from Stack #1; appropriate corrections were made to these values in making the individual assessments.

In making the AIRDOS-EPA predictions, we used meteorological data from BNL's on-site meteorological station. While AIRDOS-EPA allows for as many as six stacks, it requires the stacks to be co-located at a single point. As the distances between the facilities which released tritium at BNL are on the order of 100 to 200 meters, modeling the site with co-located release points could have introduced significant errors. Therefore, we made separate assessments for each of the stacks listed in Table 4, and summed the results of the individual assessments to obtain the predicted values given in Exhibit 2.3. As the release from the Linac Isotope Facility (0.3 Ci/yr) is trivial compared to the total site release, we combined its release (Stack 4) with that of the Van de Graaf Accelerator (Stack 3). This assumption should have a negligible effect on the predictions.

There is one major uncertainty in the predicted values. In 1985, the total release of tritium at the site was 338 Ci, of which 200 Ci were released as elemental tritium from the Van de Graaf stack and 138 Ci as tritiated water vapor distributed between the stacks, as shown in Table 4. Since the environmental measurements are only of tritiated water vapor in air, we neglected the 200 Ci/yr of elemental tritium and used 138 Ci/yr as the source term for the site. The conversion rate of elemental tritium to tritiated water vapor in air is very uncertain. At the distances of the perimeter monitoring stations (approximately one to three km), the estimates of the percentage of elemental tritium converted to tritiated water vapor range from 0.7 to 50 percent (reference 6). In order to quantify the possible error introduced by the omission of the elemental tritium from the source term, we re-evaluated the AIRDOS-EPA predictions assuming a conversion rate of 50 percent for the elemental tritium released from the Van de Graaf stack. The results indicate that the assumption of no conversion could introduce an error on the order of 300 percent.

The emissions data for BNL are based on sampling devices in the stacks of each of the facilities. These sampling devices trap tritium vapor on a desiccant. The Van de Graaf stack also has an on-line Kanne chamber for monitoring total tritium. No estimates are available of the measurement errors associated with these sampling and monitoring devices.

TABLE 4:

AIRDOS-EPA INPUT FOR
 BROOKHAVEN NATIONAL LABORATORY
 ATMOSPHERIC DISPERSION CALCULATIONS

1. Stack Parameters:

	Stack 1	Stack 2	Stack 3	Stack 4
Height, m	100	9.5	21	16
Diameter, m	5.97	0.17	0.62	0.28
Velocity, m/sec	0.54	7.0	5.5	13.0
Heat Release Rate, gm-cal/sec			NOT USED	
H-3 Release Rate, Ci/y	9.3E+1	4.2E+1	2.4E+0	2.6E-1
2. Height of Lid, m	600			
3. Rainfall Rate, cm/yr	144			
4. Average Air Temperature, °K	NOT USED			
5. Surface Roughness Height, m	0.01			
6. Height of Wind Measurements, m	88			
7. Scavenging Coefficient	0.482E-05 sec ⁻¹			
8. Deposition Velocity, m/sec	0			
9. Settling Velocity, m/sec	0			

There are large uncertainties in the environmental measurements of tritium at BNL. The environmental measurements are based on liquid scintillation counting of the tritiated water vapor collected on silica gel at the sampling stations. The samplers draw a stream of air at approximately 200 cc/min across the silica gel, and the silica gel is changed weekly. The amount of water collected per sample is about 20 ml, or, given the sampling flow of 200 cc/min, about 10 ml/m^3 . About 8 ml of the collected water vapor are used in the liquid scintillation medium; thus the tritium activity in the aliquot corresponds to the tritium activity in about 0.8 m^3 of air. The minimum detection limit of the liquid scintillation analysis is approximately 300 pCi/l, or about 2.4 pCi/8 ml. Many of the sampling results are on the order of 5 pCi/m^3 , corresponding to 4 pCi in the aliquot, which is quite close to the minimum detectable level.

There are two further uncertainties in the environmental measurements. The first is introduced by the reporting and averaging results less than the minimum detectable level as zero. This practice, which was discontinued in January of 1986, biases the reported environmental concentrations for 1985 on the low side. The second uncertainty involves the background/blank value for the samples. The tritium concentration is 55 pCi/m^3 in the analytical laboratory where the silica gel for the samplers was dried (reference 7). We were unable to obtain results from the analysis of blanks which are run with the samples. Therefore, it is impossible to determine what fraction, if any, of the tritium in the environmental samples represents contamination of the silica gel during preparation.

Due to the large uncertainty in the conversion rate of elemental tritium to tritiated water vapor in air, the bias introduced by reporting and averaging less than minimum detectable values as zero, and the unknown contribution of background/blank tritium values to the measurements, it is impossible to determine what fraction of the discrepancy between the measured and predicted values is due to the lack of precision and accuracy in the AIRDOS-EPA model. It should be noted that the measured concentrations at the control stations (located at distances in excess of sixteen and twenty kilometers) are of the same order as the measured concentrations at the monitoring stations located at distances of 1 to 3 kilometers. Given this fact, and the fact that the reported environmental measurements are far greater than one would expect given the amount of tritium released and any reasonable X/Q , we conclude that the reported environmental concentrations do not represent actual tritium concentrations in the atmosphere. Because of these uncertainties in the measured data, the

comparisons between measured and predicted concentrations of tritium at the Brookhaven site are not useful in evaluating the accuracy of precision of the AIRDOS-EPA code. For further discussion see Chapter 3.

HANFORD RESERVATION

The Hanford Reservation is located in a rural region of southeastern Washington. The site, which occupies an area of 1500 square kilometers, is located about 320 km east of Portland, Oregon and about 200 km southwest of Spokane, Washington. The Hanford Reservation is a major Department of Energy research and production center. This study provides a comparison between measured and predicted environmental concentrations for two of the radionuclides released at Hanford during fuel reprocessing campaigns: tritium and krypton-85. The terrain in the vicinity of the 200 Area where these radionuclides are released is relatively flat. In making our comparisons, we used the on-site meteorological data from the Hanford Meteorological Station for 1984, the year in which the releases occurred.

The predicted results for tritium and krypton-85 are presented in Tables 5 and 6, respectively. In making both comparisons, we used an approximation to simulate the effects of both momentum and bouyant plume rise, and ignored the 1.4 Ci/yr of tritium emitted from the 100 Area and the 1100 Ci/yr of krypton-85 emitted from the 400 Area. Ignoring the tritium and krypton releases from these other areas of the site should have a negligible impact on the results, given the magnitude of the tritium and krypton-85 releases from the main stack of the PUREX Plant in the 200 Area. Input parameters used in the calculations are given in Table 7. The stack parameter data were provided by reference 8. The release rate data and the annual rainfall were obtained from reference 9. The height of the lid is the average of the summer and winter values estimated from Figure 2.33 in reference 5. The default values from reference 3 were used for all other parameters.

As all the tritium released from the Hanford Site is in the form of tritiated water vapor, the uncertainty encountered in the BNL comparison concerning the conversion rate of elemental tritium to tritiated water vapor does not apply at Hanford. The major uncertainty in the predicted tritium and krypton-85 concentrations results from the modeling of the plume rise from the effluent stack. The air in the 61 meter stack is about 90° F and has a velocity of about 15 m/sec. Both the temperature and velocity of

TABLE 5:

COMPARISON OF MEASURED AND PREDICTED AIRBORNE TRITIUM
CONCENTRATIONS AROUND THE HANFORD RESERVATION

Station	Direction	Distance, km	Concentration, pCi/m ³		Meas/Pred
			Measured	Predicted	
K Area	NNW	11.9	2.3E+0	5.7E-2	40
N Area	NNW	14.1	1.6E+0	4.8E-2	30
D Area	N	15.2	1.3E+0	4.6E-2	30
Fire Station	NNW	10.1	1.7E+0	6.7E-2	30
S of 200 E	S	3.10	1.7E+0	1.8E-1	9
E of 200 E	E	1.40	4.1E+0	3.3E-1	10
200 ESE	ESE	2.40	3.4E+0	5.2E-1	7
SW BC Cribs	SSW	4.40	1.9E+0	1.2E-1	20
GTE Building	W	3.60	2.2E+0	1.7E-1	10
400 E	SE	18.2	2.4E+0	7.5E-2	30
Hanford	ENE	10.7	2.3E+0	6.9E-2	30
Wye Barricade	SE	12.7	1.7E+0	1.1E-1	20
Rt. 11A, Mile 9	NW	5.70	2.1E+0	1.4E-1	10
N of 200 E	N	2.40	2.4E+0	1.4E-1	20
Benton City	S	28.7	1.6E+0	3.0E-2	50
Richland	SSE	36.3	1.4E+0	2.2E-2	60

TABLE 6:

COMPARISON OF MEASURED AND PREDICTED AIRBORNE KRYPTON-85
CONCENTRATIONS AROUND THE HANFORD RESERVATION

Station	Direction	Distance, km	Concentration, pCi/m ³		Ratio Meas/Pred
			Measured	Predicted	
E of 200 E	E	1.40	3.0E+2	6.6E+2	0.5
200 ESE	ESE	2.40	1.5E+3	1.0E+3	1
S of 200 E	S	3.10	2.7E+2	3.6E+2	0.8
300 Pond	SE	27.7	1.1E+2	9.5E+1	1

* Measured values have been corrected for background, see text.

TABLE 7:

AIRDOS-EPA INPUT FOR
 THE HANFORD RESERVATION
 ATMOSPHERIC DISPERSION CALCULATIONS

1. Stack Parameters:	
Height, m	61
Diameter, m	2.1
Velocity, m/sec	15.0
Heat Release Rate, gm/cal/sec	3.4E+5
H-3 Release Rate, Ci.y	2.0E+2
Kr-85 Release Rate, Ci/y	4.0E+5
2. Height of Lid, m	1265
3. Rainfall Rate, cm/yr	16
4. Average Air Temperature, °K	282
5. Surface Roughness Height, m	0.01
6. Height of Wind Measurements, m	61
7. Scavenging Coefficient	5.35E-7 sec ⁻¹
8. Deposition Velocity, m/sec	0
9. Settling Velocity, m/sec	0

the effluent are great enough for both momentum and bouyant plume rise to be significant. Although AIRDOS-EPA does not allow for simultaneous momentum and bouyant driven plume rise, the joint effect can be approximated by the user. We chose to model the stack by setting the stack height equal to the sum of the computed momentum rise and the physical height of the stack, and then selecting the bouyant plume rise option in the code. This results in an effective stack height of 68 meters. We calculated the momentum rise according to equation 2 in reference 1.

The environmental monitoring program at Hanford includes monitoring of tritium at the release point and measurement of tritiated water vapor in air at 16 sampling locations. The environmental sampling devices pass a stream of air at about $0.01 \text{ m}^3/\text{hr}$ through cartridges containing silica gel. The cartridges, which trap tritiated water vapor, are changed every two weeks. The samples, which represent about 3.4 m^3 of air, are analyzed monthly by liquid scintillation counting. The minimum detection level is about 300 pCi/l, or about 1.5 pCi for the 5 ml aliquot used in the analysis. The quality control and assurance program at Hanford includes routine instrument calibrations, daily source and background counts, replicate analyses to check precision, and participation in inter-laboratory comparisons with both the EPA and DOE. Results for tritium from these inter-laboratory comparisons show good agreement. However, since these samples are usually about 3000 pCi/l, the aliquot is about 24 pCi, about an order of magnitude greater than the environmental samples. Thus, while these quality control samples check procedural errors, they are not a very good check on the environmental samples as they do not check background/blank errors.

No hard estimates are available for the errors in the emissions or environmental measurements for tritium. However, all of the reported environmental concentrations for tritium are very close to the minimum level of detection. Moreover, examination of the reported environmental sampling data shows that the concentrations are virtually the same at all stations, even though the stations are located from 1.4 to more than 36 km from the source. Given these facts, and the fact that the reported environmental measurements are far greater than one would expect given the amount of tritium released and any reasonable λ/Q , we conclude that the reported environmental concentrations do not represent actual tritium concentrations in the atmosphere. Because of these uncertainties in the measured data, the comparisons between measured and predicted concentrations of tritium at the Hanford Reservation are not

useful in evaluating the accuracy or precision of the AIRDOS-EPA code. See Chapter 3 for further discussion.

The measured environmental concentrations of krypton-85 are based on "grab samples" of about 0.3 m^3 collected at low flow rates. The krypton in the sample is collected and purified using a cryogenic chromatography instrument, then mixed with a scintillation fluid, and counted on a low temperature liquid scintillation counter. The minimum detectable level for krypton-85 is reported (see reference 9) as 2 pCi/m^3 , well below the reported environmental concentrations. As the reported concentrations of krypton-85 include background, the measured results presented in Table 6 have been adjusted for background by subtracting 27 pCi/m^3 from the reported values. The value of 27 pCi/m^3 is the annual average atmospheric concentration of krypton-85 during 1984 at a network of 16 environmental sampling stations located in the southwest United States (reference 10).

NUCLEAR FUEL SERVICES, ERWIN

The NRC-licensed Nuclear Fuel Services facility in Erwin, Tennessee, fabricates both low-enriched and high-enriched uranium fuel. The plant occupies a 58-acre tract approximately 0.8 km southwest of the city limit of Erwin in northeast Tennessee. The site is located in the narrow valley of the Nolichucky River. The terrain at the site is relatively flat, but rugged mountains to the north, east, and south rise to elevations of 1000 to 1500 meters within a few kilometers of the site.

Small amounts of low- and high-enriched uranium escape from the plant during the fuel fabrication processes, and concentrations of uranium in the air are routinely measured as a part of the environmental monitoring program. Reference 11 contains data on both the release rates and the measured atmospheric concentrations for 1979 through 1984. The measured concentrations of uranium for 1984 at nine off-site measurement locations are compared to our AIRDOS-EPA predictions in Table 8.

There are two principal uncertainties in the predicted values. The releases of high- and low-enriched uranium from the Nuclear Fuel Services plant during 1984 were from 11 stacks and vents. However, since the Main Process Ventilation stack accounts for more than 90 percent of the total site release, we modelled the site using only a single stack. This assumption should not introduce a significant error to the predictions. Of greater

TABLE 8:

COMPARISON OF MEASURED AND PREDICTED AIRBORNE URANIUM *
 CONCENTRATIONS AROUND THE NUCLEAR FUEL SERVICES PLANT

Station	Direction	Distance, km	Concentration, pCi/m ³		Meas/Pred
			Measured	Predicted	
Little Mtn.	NE	0.80	3.1E-3	1.2E-4	30
Caroline Ave.	ESE	0.30	2.8E-3	1.4E-4	20
Emergency Hse.	S	0.28	3.5E-3	8.6E-5	40
Caroline Ave./ Stalling	SE	0.22	3.3E-3	7.4E-5	40
Stalling Lane	SE	0.32	3.4E-3	8.6E-5	40
Highland/ 1st Street	S	0.41	3.4E-3	9.4E-5	40
Meadowbrook Ln.	ENE	0.54	1.6E-3	2.1E-4	7
Security Fence	SSW	0.21	2.5E-3	4.2E-5	60

* Natural uranium

concern are the meteorological data which were used. Meteorological measurements are taken on-site; however, they are not in a form which is suitable for input to AIRDOS-EPA. Therefore, we had to use meteorological data obtained from the National Climatic Data Center from the airport near Bristol, Tennessee, approximately 50 kilometers north of the site. The validity of these data for a site located in a narrow and steep mountain valley could not be determined.

The AIRDOS-EPA code requires use of a specific radionuclide in the source term. The emissions data refer to high- and low-enriched uranium, expressed as microcuries of gross alpha. We used uranium-235 as the radionuclide for the source term, since no isotopic breakdown of the released uranium is available. This use does not affect the results since all isotopes of uranium have identical environmental transport parameters. Furthermore, as the environmental results are also reported as gross alpha in pCi/M^3 , the predicted and measured results are directly comparable.

The additional parameters used as input in making the comparison are presented in Table 9. The stack parameters, the emission rates, and annual rainfall rate are from reference 11. The height of the lid is the average of the summer and winter values obtained from Figure 2.23 in reference 5. All other values are default values from reference 3.

No estimates are available for the errors in the emissions or environmental measurements for uranium (gross alpha) at the Nuclear Fuel Services plant. We were unable to obtain documentation on the quality control and assurance of the analytical measurements. Plant personnel (reference 12) expressed concern over the validity of the measurements. Examination of the reported environmental sampling data shows that the concentrations are virtually the same at all stations. Furthermore, as the concentrations at these stations (located between 0.2 and 0.8 km from the stack) are all of the same order as the $2.6 \times 10^{-3} \text{ pCi}/\text{m}^3$ reported for a control station about 8 km from the source, it is unlikely that the reported environmental concentrations represent actual uranium emissions from the plant. Because of these uncertainties, we conclude that the comparison between measured and predicted concentrations of uranium (gross alpha) at the Nuclear Fuel Services plant are not useful in evaluating the accuracy or precision of the AIRDOS-EPA code. See Chapter 3 for further discussion.

TABLE 9:

AIRDOS-EPA INPUT FOR
 NUCLEAR FUEL SERVICES, ERWIN
 ATMOSPHERIC DISPERSION CALCULATIONS

1. Stack Parameters:	
Height, m	35
Diameter, m	1.5
Velocity, m/sec	11.0
Heat Release Rate, gm-cal/sec	NOT USED
H. E. Uranium Release Rate, Ci/y	5.00E-3
L. E. Uranium Release Rate,	5.97E-4
2. Height of Lid, m	1035
3. Rainfall Rate, cm/yr	137
4. Average Air Temperature, °K	NA
5. Surface Roughness Height, m	0.01
6. Height of Wind Measurements, m	10
7. Scavenging Coefficient	4.584E-6 sec ⁻¹
8. Deposition Velocity, m/sec	3.551E-5
9. Settling Velocity, m/sec	0*

* We estimated the settling velocity to be on the order of 0.001 m/sec; however, AIRDOS-EPA sets the settling velocity to zero if it is less than 0.01 m/sec.

FOSSIL-FUEL ELECTRIC GENERATING STATION

The fossil-fuel electric generating station is rurally located in the eastern United States. The site is adjacent to a major river. The terrain in the vicinity of the plant consists predominately of gently rolling agricultural land. During 1982, the year for which the comparison was made, the station burned both coal and #6 oil and emitted more than 100 thousand tons of sulphur dioxide (SO_2) to the atmosphere from two identical, adjacent stacks.

The measured environmental concentrations of SO_2 in the vicinity of the station and our AIRDOS-EPA predictions are presented in Table 10. In making the comparison, we approximated the effect of both momentum and bouyant driven plume rise by setting the stack height equal to the physical stack height plus the computed height of the momentum rise. The inputs used in making the predictions are presented in Table 11. The stack parameters and emissions data were obtained from the station. The rainfall rate and the average air temperature were obtained from reference 13. All other values are the default values from reference 3.

There are three principal uncertainties in the predicted results. The first uncertainty involves the meteorological data used in the comparison. The station does not have on-site meteorological data in a format suitable for input to AIRDOS-EPA. Therefore, we used data obtained from the National Climatic Data Center for the nearest weather station, which is located approximately 60 kilometers north of the station. While the terrain is flat between the site and the weather station, the suitability of these data cannot be verified. The second uncertainty concerns the modeling of the plume rise from the stacks. The air flow and the temperature of the effluent from these stacks make both momentum and bouyant driven plume rise potentially significant. As AIRDOS-EPA cannot directly account for both bouyant and momentum driven rise, we approximated their combined effect, using the method described in the Hanford comparison. The third uncertainty involves some of the input parameters for SO_2 , which is a non-radioactive pollutant. SO_2 emissions cannot be directly modeled by AIRDOS-EPA, as the code requires at least one radioactive pollutant be input as the source term. We used uranium-238 as the surrogate for SO_2 , thus assuring that there would be no radioactive decay during plume transport. However, using U-238 as the surrogate involved accepting the default values from reference 3 for the scavenging coefficient, deposition velocity, and settling velocity of U-238, rather than that of SO_2 .

TABLE 10:

COMPARISON OF MEASURED AND PREDICTED AIRBORNE SULPHUR DIOXIDE
CONCENTRATIONS AROUND THE FOSSIL FUEL GENERATING STATION

Station	Direction	Distance, km	Concentration, pCi/m ³		Ratio Meas/Pred
			Measured	Predicted	
M-1	N	5.90	2.0E+7	3.7E+6	5
M-2A	NNW	4.00	1.7E+7	1.6E+6	10
M-3	ENE	4.40	2.0E+7	2.8E+5	70
M-4	ESE	3.20	1.8E+7	2.2E+5	80

TABLE 11:

AIRDOS-EPA INPUT FOR
 THE FOSSIL FUEL GENERATING STATION
 ATMOSPHERIC DISPERSION CALCULATIONS

	Stack 1	Stack 2
1. Stack Parameters:		
Height, m	213	213
Diameter, m	5.9	5.9
Velocity, m/sec	22.7	22.7
Heat Release Rate, gm-cal/sec	1.61E+7	1.61E+7
SO ₂ Release Rate, g/y	4.71E+10	4.71E+10
2. Height of Lid, m	790	
3. Rainfall Rate, cm/yr	99	
4. Average Air Temperature, °K	287	
5. Surface Roughness Height, m	0.01	
6. Height of Wind Measurements, m	10	
7. Scavenging Coefficient	3.31E-6 sec ⁻¹	
8. Deposition Velocity, m/sec	4.5E-4	
9. Settling Velocity, m/sec	0	

Discussions with experts in the modelling of SO₂ transport (reference 14) indicated that the scavenging coefficient and deposition velocity would not be major factors in the SO₂ concentrations at the distances of the environmental measurements. Therefore, we conclude that using the default values for U-238 introduces a negligible error in the predictions.

The release rates for SO₂ from the station are estimated based on the quantity of sulphur in the coal and #6 oil fuels. The station assigns an uncertainty of ± 20 percent to the emission estimate. Similarly, the station estimates an analytical error of ± 10 percent to the environmental measurements. However, the environmental measurements include background SO₂, which introduces a major uncertainty to the results. Data on background concentrations in the vicinity of the station for 1982, the year under examination, were not available. However, annual average concentration data were obtained for 1984. These data, which represent the annual average concentrations of SO₂ at nine air quality monitoring stations in the vicinity of the facility, indicate the ambient (background) concentrations of SO₂ in the region ranged from 24 to 39 micrograms/m³ (reference 15). Although there has been a steady decline in ambient SO₂ concentrations over time, the rate of decline in the 1980's is fairly small (reference 16). The measured data reported in Table 10 range from 17 to 20 micrograms/m³. These values are even smaller than the expected range of background. Therefore, we conclude that the contribution of SO₂ from the station cannot be distinguished from background SO₂ in the environmental measurements, and that the comparison between measured and predicted concentrations are not useful in evaluating the accuracy or precision of the AIRDOS-EPA code. For further discussion see Chapter 3.

CHAPTER 3:

ANALYSIS OF PREDICTIVE PERFORMANCE

PRELIMINARY DISCUSSION ON DATA QUALITY

In this check of the AIRDOS-EPA air concentration predictions with experimentally measured field concentrations, useful data were obtained in only two situations: natural uranium at Allied Chemical Company's UF_6 Plant, and krypton-85 at the Hanford Reservation. These were the only two situations where environmental concentrations were well above minimum detectable levels. Over the past few years, radioactive emissions to the atmosphere have been substantially reduced, largely as a result of regulatory constraints by the EPA and the NRC. The DOE also has mirrored these constraints.

The validity of a comparison of measured concentrations of a material in the environment with those predicted by the AIRDOS-EPA model is limited in two ways. First, the measurement capability must be adequate for measuring the material under investigation at the lowest concentrations of interest. Second, the model does not really predict field concentrations; it only predicts additions to the environmental concentrations from plant effluents. The actual environmental concentrations are the sum of any non-plant (background) concentrations and the model-predicted concentrations. The measurement capability must therefore be adequate for subtracting the background concentration from the observed environmental concentration to obtain the net plant contribution.

At a number of sites investigated for this report, the model-predicted environmental concentrations were lower than the stated minimum detectable levels, and so could not be expected to be detected. Further, the measured environmental concentrations did not vary with distance from the plant, and were not much above the stated minimum detectable levels. These observations suggest that (1) the environmental concentrations were too small to be measured with the precision required for a check on AIRDOS-EPA, or (2) that there is a general background level which overshadows the plant-related and model-predicted concentrations.

A common major problem with measurement capability is an over-optimistic estimate of the minimum detectable level. The minimum detectable levels given are usually calculated from counting statistics: the random Poisson variability in the measurements of sample and of background. The assumption that there is no variability other than the random Poisson variability is generally incorrect. Other sources of variability affect the precision of any measurement, and therefore affect the actual minimum detectable levels, which in such cases are higher than the stated minimum detectable levels (reference 17). As an example, consider the lower limit of detection (LLD), which is often used to describe the minimum detectable level. Lower limits of detection are calculated on the following assumptions (reference 18):

- (1) The number of counts in both sample measurement and background measurement is large enough that the Poisson distribution function can be approximated by the normal distribution;
- (2) The standard deviation of the background is equal to the square root of the number of counts: i.e., there is no variability other than the inherent Poisson variability;
- (3) The standard deviation of the sample-plus-background count is the same as the standard deviation of the background count; i.e., there is no variability other than the inherent Poisson variability, and the count due to the background is negligible in comparison to the count due to the sample.

The tritium measurements at Hanford are probably an example of an uncertainty beyond the Poisson counting statistics uncertainty. The levels are only slightly above the quoted minimum detectable level, and so may very well be below the actual minimum detectable level. Although there is an extensive quality control program at Hanford, the capability to measure at very low levels (blanks) is not documented. The ability to analyze the EPA intercomparison samples does not demonstrate a capability at very low levels.

The tritium measurements at BNL are a more clear example of uncertainty beyond Poisson statistics, with a consequent inability to measure at very low levels. The presence of 55 pCi/m^3 in the laboratory where silica gel was dried introduces a large amount of unknown excess variability.

The gross alpha measurements at Nuclear Fuel Services are also close to the stated minimum detectable levels. The ability to measure adequately at low levels is not demonstrated. In fact, at this site there is a dearth of quality control data.

An example of the effect of a background concentration is seen in the krypton-85 data at Hanford, where an adjustment had to be made by subtracting 27 pCi/m^3 from the reported values. The value of 27 pCi/m^3 is the annual average atmospheric concentration of krypton-85 at a network of 16 environmental sampling locations in the southwest United States during 1984 (reference 10), and is also approximately the value found at distant locations around the site. In this case, because there is a well-measured background level and because the measured environmental levels are high compared to the minimum detectable levels, the corrected environmental monitoring data do give an adequate basis for comparison with the AIRDOS-EPA predictions.

The fossil fuel electric generating station is another example of a situation in which the background level swamps the immediate plant emissions. A general background level of 24 to 39 micrograms per cubic meter is typical for the region (reference 15), far above the model-predicted levels of 0.2 to 3.7 micrograms per cubic meter.

In the remainder of this chapter, an analysis of the predictive performance of the AIRDOS-EPA model is presented for five sites analyzed in this and the previous study. Included are four sites for which useful environmental concentration measurements were obtained in the previous study — Allied Chemical (1981), FMPC, Mound, and Savannah River. Of the five sites analyzed in the current study useful environmental measurements were obtained for one new site — the Hanford Reservation — and a second year (1980) of data was obtained for the Allied Chemical facility, yielding a total of six data sets at five sites useful for comparing AIRDOS-EPA predictions to measured values. The accuracy of the model at the five individual sites is discussed first, followed by a discussion of the precision of the overall predictive performance. Three methods are used for comparing the measured and predicted concentration values — graphical display of the data, correlation analysis, and regression analysis.

GRAPHICAL DISPLAY

The scatterplots in Figure 1 demonstrate the effect of applying a site-specific multiplicative adjustment factor to the predicted concentrations at each site, the

FIGURE 1A:
LOGARITHMIC SCATTERPLOTS OF (A) ORIGINAL AND (B) ADJUSTED
PREDICTED VALUES VERSUS MEASURED VALUES AT ALL SITES

A. PREDICTED VS. MEASURED VALUES

REFERENCE LINE: INTERCEPT = 0 AND SLOPE = 1

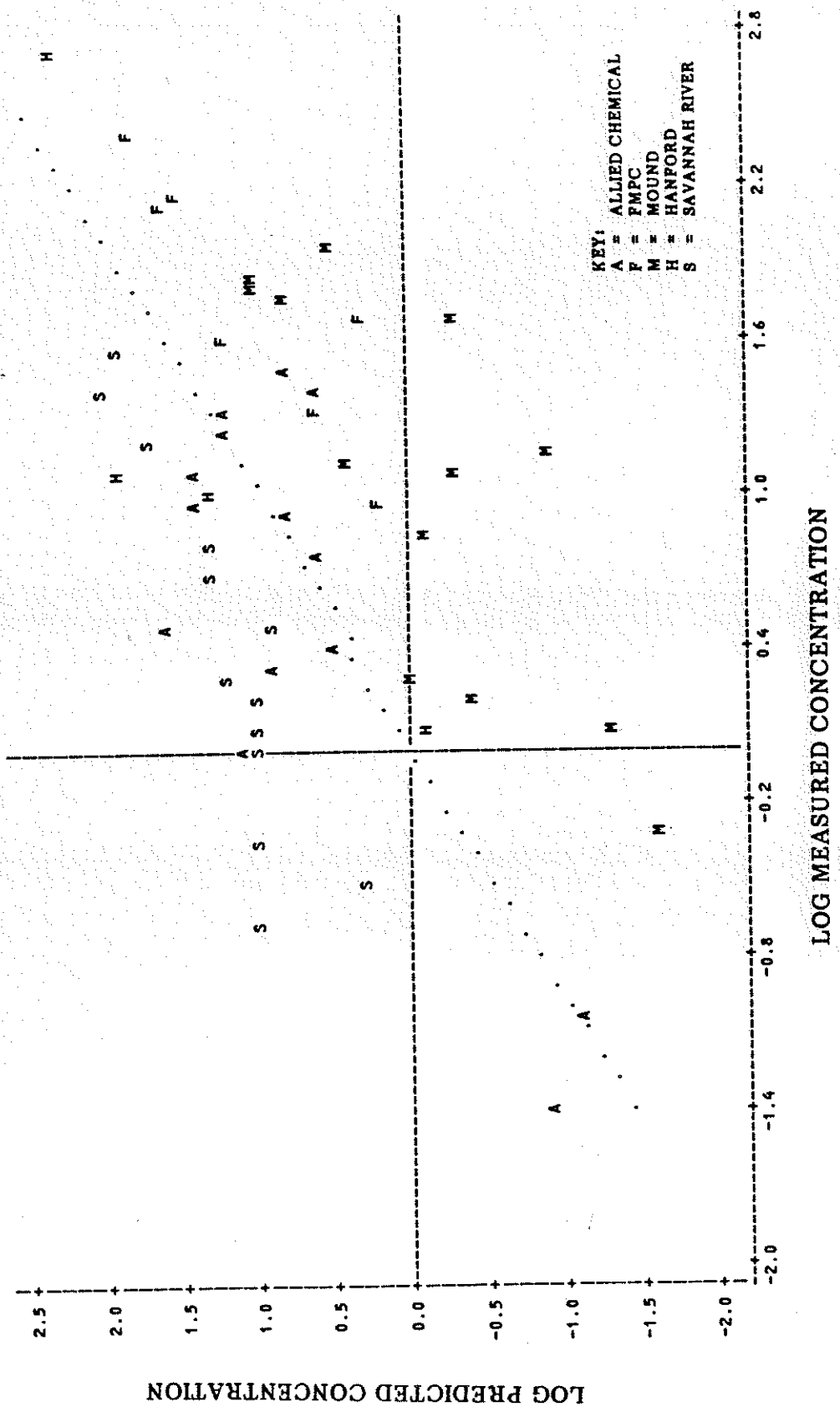
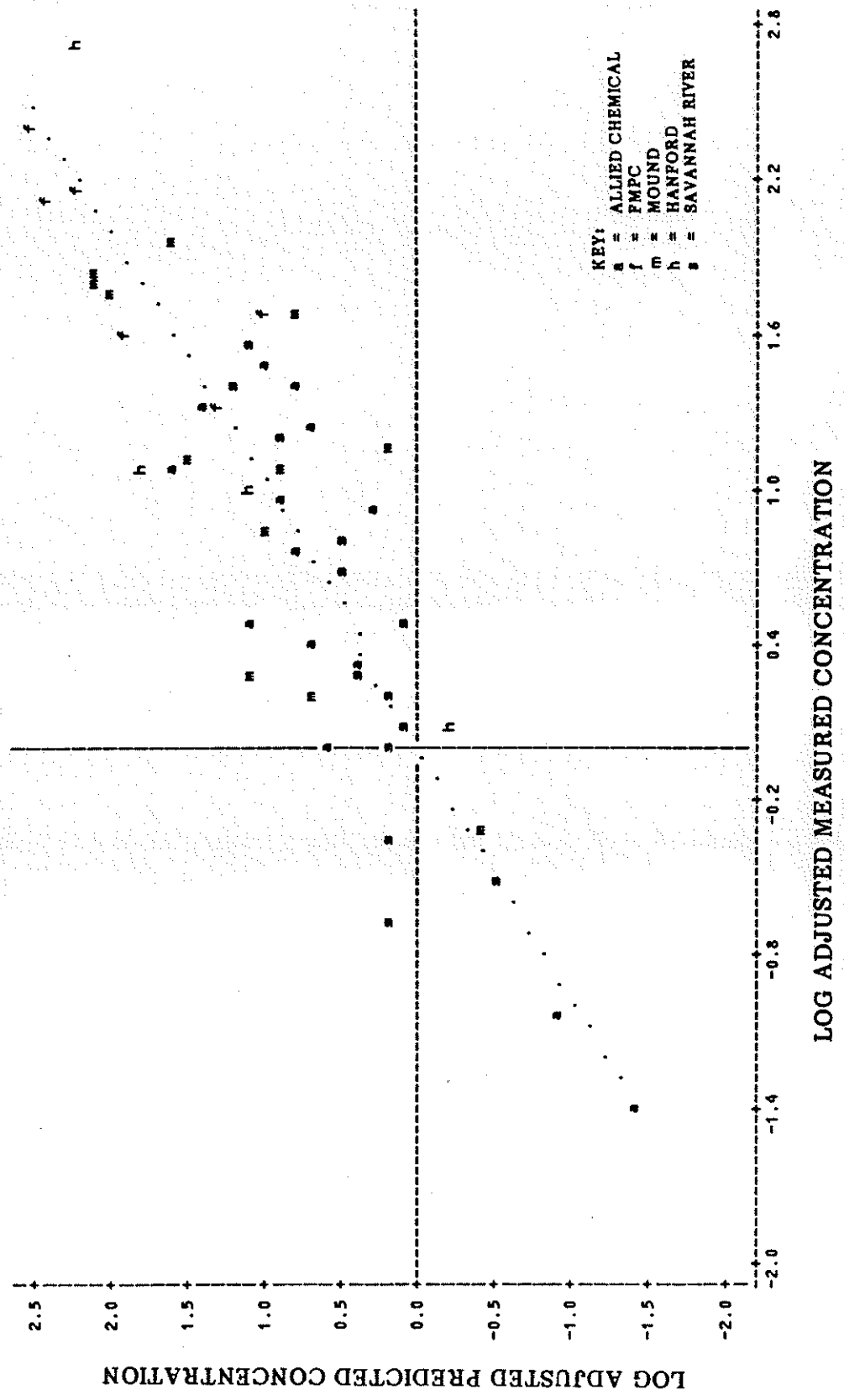


FIGURE 1B:
LOGARITHMIC SCATTERPLOTS OF (A) ORIGINAL AND (B) ADJUSTED
PREDICTED VALUES VERSUS MEASURED VALUES AT ALL SITES

B. ADJUSTED PREDICTED VS. MEASURED VALUES

REFERENCE LINE: INTERCEPT = 0 AND SLOPE = 1



KEY:
 a = ALLIED CHEMICAL
 f = FMPC
 m = MOUND
 h = HANFORD
 s = SAVANNAH RIVER

adjustment factor being equal to the ratio of the geometric means of the predicted and measured values. The scatterplots graph the natural logarithms of the predicted (A) and adjusted predicted (B) concentrations versus the logarithm of the measured concentrations. Examination shows that the unadjusted predictions for the Savannah River and Allied Chemical sites lie entirely above the $P=M$ reference line, which has slope 1 and intercept 0. The unadjusted predicted values lie entirely below the reference line for the remaining three sites. After scaling the predictions by the site-specific adjustment factors, the scatterplots are much nearer to the desired reference line. Using visual inspection, we are unable to reject the hypothesis that the combined data after adjustment has a regression line with intercept 0 and slope 1. This implies that the predictions are statistically equal to the measured concentrations after applying the adjustment factors.

The derivation of the adjustment factors is summarized in Table 12. The exhibit presents the geometric means of the measured and predicted concentrations at each site, and derives an adjustment factor equal to the ratio of the geometric means. Due to the spatial consistency of the relationship between predicted and measured concentrations, the adjustment factors have a special significance. If the predicted concentrations for all stations at a site are simply scaled by this factor, the resulting adjusted predictions are very similar to the measured values at almost all stations, regardless of direction or distance from the source.

This fundamental observation indicates that a straight-forward relationship exists between the measured and predicted concentrations. The predictive concentrations generated by the AIRDOS-EPA code may be higher or lower than measured concentrations by a factor of 3, but have a spacial pattern similar to the measured concentrations. The scalar difference between the measured and predicted values may be due to a variety of factors which include improper specification of the source characteristics for each site when generating predicted concentrations, or a systematic bias in the procedures used to obtain the measured concentration values at a given site.

CORRELATION ANALYSIS

A second measure of the predictive performance of the AIRDOS model may be obtained through correlation analysis. The (Pearson) correlation between P and M is given by

TABLE 12:
COMPARISON OF GEOMETRIC MEANS^a OF
PREDICTED AND MEASURED CONCENTRATIONS

Site (radionuclide)	Unit (pCi/m ³)	Number of Stations (N)	Geometric Means		Adjustment Factor for Predictions (\bar{M}/\bar{P})
			\bar{M}	\bar{P}	
Allied Chemical (1981) (uranium-238 and 234)	10 ⁻²	7	1.43	2.38	.601
* Allied Chemical (1980) (uranium-238 and 234)	10 ⁻²	7	2.14	1.77	1.21
* Hanford Reservation (Krypton-85)	10 ²	4	3.40	3.88	.877
FMPC ^(b) (uranium-238 and 234)	10 ⁻³	7	5.69	2.77	2.06
Mound (tritium)	10 ¹	13 ^(c)	2.81	.918	3.06
SRL (krypton-85)	10 ¹	13	1.48	3.31	.446
* All	—	51	2.34	2.07	—

Notes: (*) New data from current analysis.

(a) The geometric mean is defined as $\bar{X} = \left(\prod_{i=1}^N X_i \right)^{1/N}$.

(b) Based on data in Table 1 for 1981 only.

(c) Omits anomalous data for station 115.

$$r = \frac{\sum_{i=1}^N (M_i - \bar{M}) (P_i - \bar{P})}{\left(\sum_{i=1}^N (M_i - \bar{M})^2 \sum_{i=1}^N (P_i - \bar{P})^2 \right)^{1/2}}, \quad (1)$$

where \bar{P} and \bar{M} denote the arithmetic mean predicted and measured values. The correlation measure may be calculated either for the raw data or for the logarithms of the data. If a scatterplot of the M_i and P_i values lies entirely on an upward sloping straight line, then the correlation is $r = +1$. Values of r less between zero and one indicate scatter around this straight-line relationship.

Correlations were calculated for each site using both the raw data and the logarithms of the data. The results are shown in Table 13. The table also shows the correlations for all 51 stations for all sites, after adjusting the predictions to account for the site-by-site adjustment factors derived in Table 12. With the exception of the correlations based on the 1980 and 1981 raw data for the Allied Chemical site, all correlations exceed 0.75, indicating a relatively high level of correlation between measured and predicted values.

When correlations are calculated on a site-by-site basis, the adjustment factor does not affect the correlation, since the correlation measure is invariant with respect to scaling either the predicted or measured values. The overall correlation between the predicted and measured values for all sites is affected by the adjustments, however, since each site has a unique adjustment factor.

REGRESSION ANALYSIS

The almost linear relationship that exists between the logarithms of the predictions and measurements suggests that regression analysis may be applied to evaluate the slope and intercept of the linear relationship. The method of regression analysis determines the relationship between predicted and measured values, either at a site or across all sites, based on the following linear model:

$$\ln P_i = a + b \ln M_i + \ln e_i \quad (2)$$

Here P_i (M_i) represents the predicted (measured) concentration at station i , and the coefficients, a and b , are the regression parameters to be estimated via least-squares

TABLE 13:
ANALYSIS OF CORRELATION OF PREDICTIONS WITH MEASUREMENTS

Site (radionuclide)	Unit ³ (pCi/m ³)	Adjustment Factor for Predictions (M/P)	N	P vs. M log P vs. log M	Correlation	Summary Evaluation
Allied Chemical (1981) (uranium-238 and 234)	10 ⁻²	.601	7	.49	.84	Good correlation for log data. Predictions generally high (by factor of about 2).
* Allied Chemical (1980) (uranium-238 and 234)	10 ⁻²	1.21	7	.60	.89	Good correlation for log data. Predictions somewhat low (20%).
FMP(a) (uranium-238 and 234)	10 ⁻³	2.06	7	.93	.88	Highly correlated. Predictions low (by factor of 2).
* Hanford Reservation (krypton-85)	10 ²	.877	4	.87	.88	Only 4 stations, but highly correlated. Predictions 12% high.
Mound (tritium)	10 ¹	3.06	13 ^(b)	.80	.79	Good correlation. Predictions low (by factor of 3). One extreme outlier removed.
SRL (krypton-85)	10 ¹	.446	13	.95	.87	Highly correlated. Predictions high (by factor of 2).
* All	—	—	51	.86 ^(c)	.88 ^(c)	Overall correlation high (after adjustment). Predictions are 12 percent low overall (before adjustment); see previous table.

Notes: (*) New data from current analysis.

(a) Uses data for 1981 only.

(b) Omits anomalous data for station 115.

(c) Overall correlations shown for adjusted predicted values vs. measured values. Correlations before adjustment are .57 and .50 for the original and logged data, respectively.

techniques. The term e_i denote random predictive error terms which will be discussed below.

Equation 2 implies a linear relationship between the logarithms of the predicted and measured values. If the predicted values were exactly equal to the measured values at all stations, the least-squares estimates of the regression parameters would be $a=0$ and $b=1$, with a resulting R^2 of 1.0, indicating a perfect fit. The degree to which a , b , and R^2 differ from these desired values provides the third measure of the predictive performance of the AIRDOS model. The significance of observed deviations from the desired parameter values may be tested by making further assumptions concerning the predictive error terms in Equation 2. If the error terms are assumed to have a log-normal distribution, then the F-test for linear regression parameters may be applied to test the joint hypothesis that $a=0$ and $b=1$ in Equation 2. The hypothesis may be rejected when the F statistic is greater than the threshold value for the test, which varies with the desired level of confidence.

Equation 2 may also be written as

$$P_i = sM_i^b e_i \quad (3)$$

where the auxiliary parameter, s , denotes a common multiplicative scaling factor between predicted and measured values. This equation demonstrates that the predictive error term supplies a random multiplicative disturbance at each station. Taking the logarithm of Equation 3 yields Equation 2, with $a = \ln s$.

As indicated by Equation 3, a non-zero value of the parameter a indicates the existence of a multiplicative scaling factor in the predicted values. Values of b not equal to unity indicate that the predictive values are not proportional to M , but to a power of M . Due to the extremely high correlation of the levels of the measured value with distance from the source, testing for a power response of P to M is equivalent to testing for errors in the predictions that are related to distance. We shall find little evidence of such effects due to distance from the source.

We now address the assumptions made concerning the statistical properties of the predictive error terms in the equation. If it is assumed, in Equation 3, that the

predictive errors e_i have the lognormal distribution, then in Equation 2 the predictive error terms $\ln e_i$ have the normal distribution. The choice of the lognormal distribution may be considered as a convenient mathematical artifact, given that we believe the multiplicative disturbance model of Equation 3, yet desire to apply the F-test theory which is based on the normal distribution. Due to the small numbers of data points available at each site, it is difficult to select one distributional model for the predictive errors over other similar distributions.

The regression model of Equation 2 was estimated with the log adjusted predicted values as dependent variables and the log measured values as the independent variable using standard least-squares procedures. The regression results are summarized in Table 14, for each site and for all sites combined. The parameter estimates for the unconstrained model, which includes an intercept term, show that the estimate for the intercept is insignificantly different than zero for four of the five sites. The exception is the Savannah River facility, for which the estimate of a is slightly more than two standard errors greater than zero. For all sites combined, the estimate of a is less than two standard errors away from zero. These results indicate that a constrained model with no intercept term may provide better estimates of the slope parameter b . The constrained model results are discussed below.

The estimates of b in the unconstrained model are within one standard error of unity for four of the six cases — Allied Chemical in 1981, FMPC, Hanford and Mound. The first exception, Allied Chemical in 1980, has almost the same slope as for 1981, but the R^2 is higher and the corresponding standard error lower. The result is that the estimate for b is slightly more than one standard error from unity. The Savannah River site is also an exception, with the estimate of b being significantly less than unity. For all sites combined, the estimate of b is slightly more than two standard errors from unity.

The regression F-tests for the unconstrained models indicate that the estimated relationship between the adjusted predictions and the measurements is significant at the .05 level except at the Hanford site, which also has the fewest measurements and the largest standard error associated with the estimate.

The final column in the exhibit for the unconstrained model shows the result of the parameter F-test, which tests the joint hypothesis that $a=0$ and $b=1$. The degrees of freedom for the F statistic for this test depend on the number of parameters being tested ($m=2$) and the number of data points, $n=N-2$. Except for the Savannah River

facility, all F-statistics are sufficiently small that we cannot reject the hypothesis that $a=0$ and $b=1$ after adjustment.

The above result demonstrates the importance of the scaling adjustment factors derived in Table 12. After adjustment, the residual errors in the regression of the log predicted versus log measured values for all sites have a mean squared error of 0.161 and hence a standard error of 0.401. This standard error for the logarithmic relationship implies that the adjusted predictions are typically (50 percent confidence interval) in the range from 33 percent low to 49 percent high, when compared to the measured values. Although a detailed statistical analysis of the degree of measurement error is not possible due to the limited available data on measurement variance, the small residual error which remains in the predicted values after adjustment appears to be consistent with the degree of error in the measured values. In conclusion, after adjustment the predicted values are statistically equivalent to the measured values.

More precise estimates of the slope parameter b are shown in the constrained model results portion of Table 14. These estimates were derived by eliminating the intercept term of the model. Except for SRL, the estimates of b are very close to unity, indicating equality of the predicted and measured values after adjustment. The regression F-statistics show that the constrained models for all sites are significant at the .05 level. The R^2 values for the constrained model are only slightly smaller than those for the unconstrained model, indicating that the omitted intercept term has little explanatory power.

The F-test for the hypothesis that $b=1$ shows that we cannot reject the hypothesis, except at SRL. For this site, the 95 percent confidence interval for b extends from .50 to .88, falling slightly below the desired value of unity. Hence, the SRL predictions do not appear to be linearly related to the measurements, but rather are approximately fitted by the function $P=M^{0.7}$. This relationship indicates that after adjustment there is still a possibility of underprediction at high values of M , which occur close to the site, or overprediction at low values of M , which occur far from the site. Examination of the data for SRL in Figure 1 confirms the latter possibility — overprediction at large distances from the site. (There is also some slight evidence for the former possibility.) The SRL site is the only site for which measurements were made at very large distances from the site. Because krypton is an inert gas, the overprediction at large distances at SRL is not due to deposition or scavenging. Increased atmospheric

TABLE 14:
SUMMARY OF REGRESSION RESULTS FOR ADJUSTED
PREDICTED VALUES VS. MEASURED VALUES

Site (radionuclide)	Unconstrained Model (ln P = a + b ln M)					Constrained Model (ln P = b in M)				
	Parameter Estimates (standard error)		F for Regression (Prob > F)	R ² (m,n) for F test	F test for H ₀ : a = 0, b = 1.0 (Prob > F m,n)	b (standard error)	F for Regression (Prob > F)	R ² (m,n) for F test	F test for H ₀ : b = 1.0 (Prob > F m,n)	
	a	b								
Allied Chemical (1981) (uranium)	.072 (.203)	.801 (.229)	12.21 (.017)	71 (2,5)	0.38 (.424)	.833 (.194)	18.5 (.005)	70 (1,6)	0.74 (.422)	
Allied Chemical (1980) (uranium)	.152 (.208)	.800 (.185)	18.62 (.008)	79 (2,5)	0.58 (.592)	.893 (.130)	46.8 (.005)	77 (1,6)	0.68 (.442)	
FMPC (uranium)	-.283 (.501)	1.162 (.278)	17.49 (.009)	78 (2,5)	0.17 (.847)	1.011 (.069)	217.0 (.0001)	76 (1,6)	0.03 (.876)	
Hanford (krypton-85)	.208 (.492)	.830 (.319)	6.79 (.121)	77 (2,2)	0.14 (.648)	.937 (.166)	32.0 (.011)	75 (1,3)	0.15 (.729)	
Mound (tritium)	.153 (.250)	.852 (.197)	18.62 (.001)	63 (2,11)	0.28 (.760)	.950 (.112)	71.9 (.0001)	62 (1,12)	0.20 (.661)	
SRL (krypton-85)	.162 (.077)	.586 (.098)	35.83 (.0001)	77 (2,11)	8.92 (.005)	.688 (.096)	50.9 (.0001)	67 (1,12)	10.52 (.007)	
All	.128 (.079)	.850 (.065)	173.5 (.0001)	78 (2,49)	2.70 (.078)	.923 (.047)	387.7 (.0001)	77 (1,50)	2.67 (.109)	

mixing because of underestimation of the lid height may explain the discrepancy between measure and predicted values at very large distances, although, as noted earlier, doubling the lid height reduced the predicted concentration by less than 25 percent.

The constrained regression results for all sites combined yield a 95 percent confidence interval for b which barely includes the desired value of $b=1$. As a result, the F-statistic for testing the hypothesis $b=1$ indicates that we cannot reject the hypothesis at the 90 percent level. However, at any higher and less stringent confidence level, the hypothesis that $b=1$ may be rejected in favor of values less than unity. We must conclude, therefore, that, with the exception of a few sites at SRL located at large distances from the source, the predicted values are statistically equal to the measured values after applying the adjustment factors.

CHAPTER 4:

CONCLUSIONS

The accuracy of the AIRDOS-EPA code is determined largely by our ability to estimate the adjustment factor at each site. After the predictions are adjusted to account for the differences in the geometric means of the measured and predicted concentrations at each site, the adjusted predictions are equal to the measured values at almost all locations to within acceptable statistical error. Hence, the adjustment factors are a good measure of the accuracy of the model. The adjustment factors are .446, .601, .877, 1.21, 2.06, and 3.06 for the Savannah River, Allied Chemical (1981), Hanford, Allied Chemical (1980), FMPC, and Mound sites, respectively. (See Table 12.) Considering the six data sets at all five sites together, without adjustment, the predictions are 12 percent low overall when compared to environmental values.

Although we have only six data sets at five sites on which to base our opinions, we may formally derive a statistical confidence interval for the accuracy of the model based on the adjustment factors at each site. The 50 percent confidence interval for the accuracy of the model at a typical site ranges from a factor of 0.7 to a factor of 2.0, based on the log-normal distribution. As often as not, we may expect that AIRDOS-EPA predictions for ground-level concentrations will be within a factor of 2 of the measured values, when the model is applied at sites with well-known source characteristics and weather data. The 90 percent confidence interval for the accuracy ranges from a factor of 0.3 to a factor of 4.4. Due to the small number of sites, the 90 percent confidence interval for the adjustment factors based on all five sites is quite large.

The precision of the AIRDOS-EPA code in predicting ground-level radionuclide concentrations at varying distances from the source is high, after adjustment for incorrectly specified source characteristics or possible measurement bias at each site. The precision of the model is best measured by the constrained regression of adjusted predicted values versus measured values for all sites. See Table 14 (All Sites) and (Figure 1). The coefficient of determination (R^2) of this regression is 0.77 with a slope of 0.92, indicating that the predicted concentrations are statistically equivalent to the measured values after adjustment for site-specific bias. At the 50 percent confidence level, the predicted concentrations at all sites are within 50 percent of the measured values after adjustment.

REFERENCES

1. R.E. Moore, et al., "AIRDOSE-EPA: A Computerized Methodology for Estimating Environmental Concentrations and Dose to Man from Airborne Release of Radionuclides," U.S. Environmental Protection Agency, EPA 520/1-79-009, December 1979.
2. A.L. Sjoreen and C.W. Miller, "PREPAR — A User-Friendly Pre-processor to Create AIRDOS-EPA Input Data Sets," Oak Ridge National Laboratory, August 1984.
3. D.A. Fleming, M.W. Boback, and K.N. Ross. "Feed Materials Production Center Environmental Monitoring Annual Report for 1981," NLCO-1180, May 1, 1982.
4. D.A. Fleming and K.N. Ross. "Feed Materials Production Center Environmental Monitoring Annual Report for 1983," NLCO-2018, August 1984.
5. Letter from M.W. Boback (Director, Health and Safety Div., NLO, Inc.) to T.W. Oakes (ORNL Environmental Coordinator, Oak Ridge National Laboratory, Union Carbide Corp.), May 17, 1983.
6. D.H. Slade, ed., Meteorology and Atomic Energy 1968, U.S. Atomic Energy Commission, July 1968.
7. K. Telegadas, et al., "Measured Weekly and Twice-Daily Krypton-85 Surface Air Concentrations Within 150 km of the Savannah River Plant (March 1975 through September 1977) — Final Report," Air Resources Laboratories, Silver Spring, Maryland, NOAA Technical Memorandum ERL ARL-80, January 1980.
8. Proceedings of the First SRL Model Validation Workshop November 19-21, 1980, Hilton Head, South Carolina, E.I. du Pont de Nemours and Co. report DP-1597, October 1981.
9. "Iodine Species in Reactor Effluents and in the Environment", Electric Power Research Institute, Report NP-1269, December 1979.
10. "Application (to the Nuclear Regulatory Commission) for Renewal of Source Materials License," Sub 526, Docket 40-3392, Allied Chemical Corp., July 1982.
11. "Annual Environmental Monitoring Report: Calendar Year 1981, Mound Facility," Monsanto Research Corp., report MLM-2930, April 21, 1982.
12. "Background Information Document, Proposed Standard for Radionuclides," U.S. Environmental Protection Agency, EPA 520/1-83-001, March 1983.