Comparison of Estimated and Measured Long-Term ^{131}I Concentrations from a Continuous Elevated Source[1]

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The continuous release of very small, but detectable amounts of ^{131}I from the Brookhaven Graphite Research Reactor has provided an unusual opportunity for verification of dispersion estimates based on meteorological parameters. Analysis of an entire year's data shows the prediction system to be reliable within a factor of 2, and contains valuable information about such factors as effective stack height, the effectiveness of simplified dispersion models, and the variation of dispersion parameters with season and trajectory.

1. Introduction

One of the first joint undertakings of the Health Physics Division and the Meteorology Group at Brookhaven was a study of the possible environmental effects of the ^{41}Ar radioactivity in the cooling air plume from the Brookhaven Graphite Research Reactor (BGRR). A prediction system based on meteorological variables was developed to calculate the radiation at ground level (Lowry, 1950; Smith, 1951), and subsequent study of the monitoring data by Singer (1954) indicated that the system was substantially accurate although somewhat conservative in the sense that the dosages were overestimated.

In 1964, the Health Physics Division improved its technique for sampling and measuring ^{131}I sufficiently to permit study of the ground level concentrations of radioiodine in the reactor cooling air effluent, which is released from a stack 350 ft above the general level of the terrain. Following this development, Health Physics and Meteorology again joined forces in a project that has revealed important aspects of the actual behavior of the cooling air plume.

2. Estimation of ground level concentrations

The prediction method itself is not unique to Brookhaven, but includes the same series of steps that most such estimates entail.

The first element of this system requires good estimates of the dispersion parameters associated with the problem. Singer and Smith (1966) have shown that studies of oil fog test plumes and other tracers at Brookhaven have defined these conditions well, and the instruments on the 420-ft tower erected for these early experimental studies continue to supply the meteorological data needed for current estimates.

The second element is the estimation of the effective stack height of the plume. This quantity is the sum of the actual height of the release point and any additional rise associated with buoyancy or momentum effects of the plume itself. It is dependent upon wind and stability of the atmosphere, as well as the character of the cooling air release. The effective stack height of the BGRR plume has never been accurately determined, since the ^{41}Ar is as unsuitable for this purpose as it is for the determination of other details of the plume geometry. The plume, however, undoubtedly rises well above the stack top, since approximately 115 m^3 sec^{-1} of cooling air is released at 50C above ambient temperature and a vertical speed of 6 m sec^{-1}. Estimation of the plume rise is still recognized as an uncertain portion of air pollution evaluations, since few investigators have studied field data with sufficient precision to determine exactly what does happen. This study fortunately sheds some light on the problem, at least for emissions of this order.

Based on the effective stack height and the existing dispersion conditions, a Gaussian plume model of the following form is used as a representation of the dispersion process:

$$\chi(x,y,0)=\frac{Q}{\pi\sigma_y\sigma_z\bar{u}}\exp-\{(h^2/2\sigma_z^2)+(y^2/2\sigma_y^2)\},\quad (1)$$

where

χ = concentration (units m^{-3}) or dosage (units sec m^{-3}),
Q = release rate (units sec^{-1}) or a total release (units),
σ_y, σ_z = crosswind and vertical plume standard deviations (m),
$\bar{u}$ = mean wind speed (m sec^{-1}),
h = initial source height (m), and
x, y, z = downwind, crosswind, and vertical distances (m).

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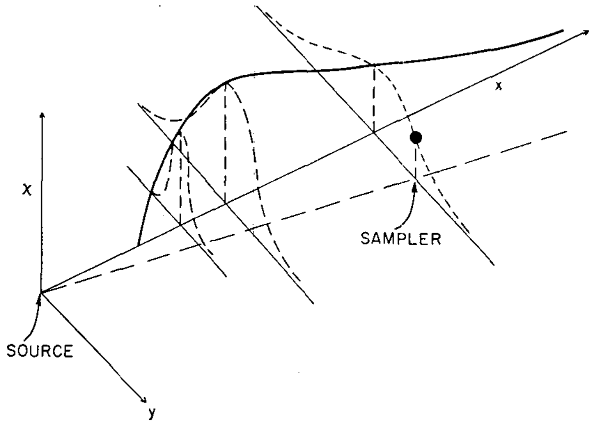

Fig. 1. Precise estimate of receptor concentration. The ground-level concentration pattern from an elevated source under typical daytime conditions is shown. The sampler concentration is estimated according to its exact position in reference to the pattern.

For relatively short periods (1–24 hr), one establishes for each hour the mean wind direction and values of $\bar{u}$, σ_y, σ_z and h, from which a ground-level concentration pattern is established as shown in Fig. 1. A given sampling point will occupy an x, y position (or series of positions) in relation to such patterns. This is the most accurate estimate possible with present techniques.

For longer periods such as a week or a month, it is more convenient to apply a simplified form of the dispersion equation, although it is by no means impossible to evaluate every hour of the period by the precise method and to obtain a sum of hourly contributions. The simplified method involves determining a concentration pattern that would be present if the wind direction were distributed uniformly throughout a given angular section of width θ, and a single meteorological dispersion condition persisted unchanged for the entire period. Mathematically this is achieved by integrating Eq. (1) in the y-direction and dividing it by $2\pi x\theta/360$, a quantity which closely approximates the sector width as long as the plume is relatively narrow compared to the arc segment $x\theta$, or the sector width itself is small compared to x. In most circumstances the error introduced by the arbitrary mixing of the coordinate systems is negligible. This procedure gives a new dispersion equation of the form

$$\chi(x,0)=\frac{360Q}{\theta\pi^{\frac{3}{2}}2^{\frac{1}{2}}\sigma_z\bar{u}x}\exp-(h^2/2\sigma_z^2). \tag{2}$$

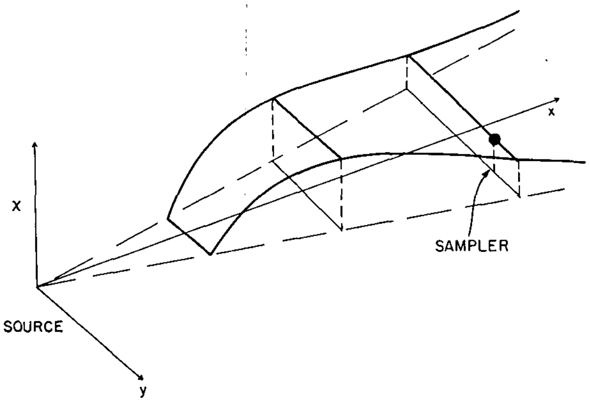

Fig. 2. Sector estimate of receptor concentration. The concentration pattern is similar to Fig. 1, but the assumption of uniform distribution in the y-direction makes the sampler concentration dependent only on x, h and the dispersion conditions.

A typical concentration distribution is shown in Fig. 2. The lateral position of the sampler within the sector is not important, and the concentration depends only on the dispersion condition, the height of release, and the distance from the source. It is assumed that whenever the wind direction is such as to include a sampler within the section width selected (30 deg in these studies), it will be affected by the concentration given by Eq. (2) computed for the appropriate distance and meteorological condition.

Several non-meteorological corrections and adjustments are required in this particular evaluation to account for variation in output rate Q, and sampler operation, as well as the radioactive decay of the isotope. All are included in the data presented.

In keeping with standard practice at Brookhaven, all hours are classified in terms of four dispersion conditions, determined from the wind direction trace at 355 ft on the meteorological tower. The dispersion parameters associated with these conditions are given in Table 1. In practice, all hours having D gustiness are discarded from the computations process, since extensive studies of tracer plumes in this area (Smith, 1956) have shown that there would be no ground-level concentrations whatever within the radius of the sampling stations. Transitory conditions, such as the alleged fumigation following an inversion break-up are also ignored, since there is no evidence that they occur at all with the BGRR plume, and their effects on long-term statistics would not be discernible even if they did.

Table 1. Dispersion parameters used in the study.

Brook-haven gustiness class*	General meteorological conditions	Plume standard deviations at a distance x downwind σ_y (m)	 σ_z (m)
B₂	Light winds, strong insolation	$0.40x^{0.91}$	$0.41x^{0.91}$
B₁	Moderate to strong winds, moderate insolation	$0.36x^{0.86}$	$0.33x^{0.86}$
C	Moderate to strong winds, overcast skies	$0.32x^{0.78}$	$0.22x^{0.78}$
D	Stable conditions	$0.31x^{0.71}$	$0.06x^{0.71}$

* The classification is based on the fluctuations of the 355-ft wind direction, as described by Singer and Smith (1966).

3. Comparison of predicted and observed samples

The first comparison of predicted and observed concentrations involved an unusual release of iodine (^{131}I and ^{133}I) which occurred during the first half of March 1965. The release rate began at 34 mCi hr^{-1} of ^{131}I and 365 mCi hr^{-1} of ^{133}I. The ^{133}I was apparent for only a few days since its half life is 21 hr, but the 8-day ^{131}I remained at above normal concentrations for two weeks.

Table 2 summarizes only ^{131}I data. The samples, which are the measured amounts at the end of the collection period, were obtained over periods ranging from 1 hr to 10 days depending on the release rate and meteorological conditions. Although there were variations, most of the dispersion conditions bringing material to the samplers were of the B_1 (typical daytime) type. Eqs. (1) and (2) were both used in the analysis, depending upon the length of the sampling period.

The first and most obvious feature of the table is the tendency for the predicted/observed (P/O) ratios to be large close to the source decreasing to values near the ideal of 1.0 between 2 and 3 km. This tendency must almost certainly be associated with an error in the estimation of the effective stack height h, since this seems to be the only way to obtain a grossly erroneous prediction close to the source and reasonable values at greater distances. Fig. 3 shows how an improper estimation of h can produce this result. The discrepancy between the concentration curves derived from the two stack heights is great in the region from 0.5 to 1.0 km, but decreases to small values at larger distances, where the effect of the initial source height becomes small.

The predicted sample amounts in Table 2 were based on the Bosanquet *et al.* (1950) equation for the plume effective stack height. This was originally chosen because it had given reasonable values for industrial stack emissions of somewhat larger magnitude than that of the reactor. Clearly, however, the estimate of the BGRR plume behavior under these typical meteorological conditions must be erroneous in view of the observed ratios shown in the table.

TABLE 2. Comparison of observed and predicted samples for March 1965 series using Bosanquet Δh equation.

Station	Distance (m)	^{131}I (Ci$\times 10^{-12}$) Obs.	Pred.	P/O
E-11	700	35	250	7.1
		43	260	6.0
		35	65	1.8
		13	22	2.8
		14	46	3.3
Igloo	1750	100	300	3.0
E-4	2200	6	6	1.0
E-7	2550	36	22	0.6
		75	140	1.8
		47	54	1.1
		20	32	1.6
E-9	2750	10	5	0.5
		12	5	0.4

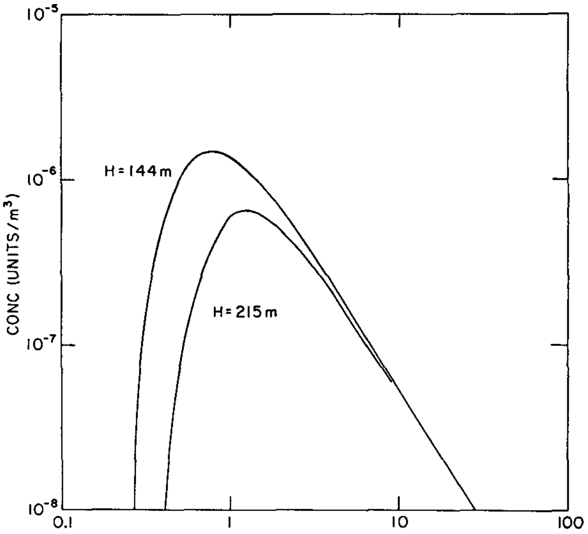

FIG. 3. Centerline concentrations with different effective stack heights. The importance of proper selection of the effective stack height to correct estimation of concentrations close to the source is shown. Between 0.3 and 1.0 km, the predicted values differ markedly, but beyond 3 km, they become almost identical.

Recent study of hot puff behavior at Brookhaven has suggested that the buoyancy effect of the warm gas emission might be more important than suggested by the Bosanquet equation, and the entire series of data were recomputed on the basis of the Central Electric equation (Lucas *et al.*, 1963). Comparison of the predicted effective stack heights for this and the Bosanquet equation are shown in Fig. 4. The Central Electric com-

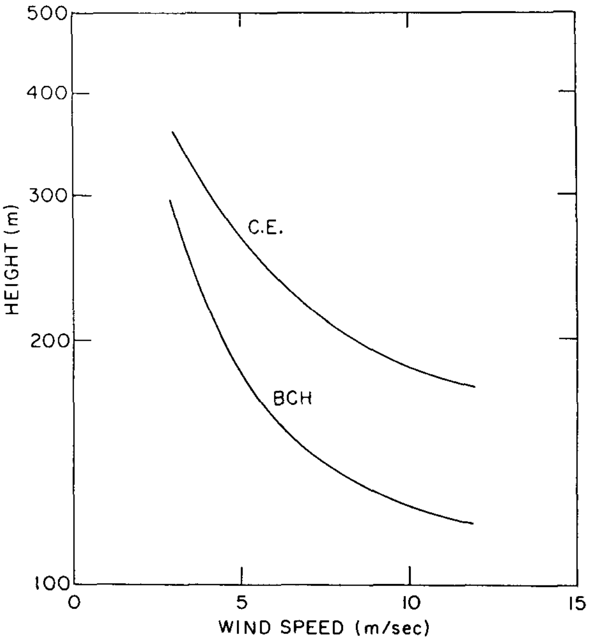

FIG. 4. Comparison of effective stack heights predicted by the Central Electric and Bosanquet equations for BGRR.

putation gives a more optimistic set of estimates of effective stack heights, exceeding those of the Bosanquet equation by more than 50 m at typical wind speeds.

The left hand portion (^{131}I) of Table 3 is identical to Table 2, except that the estimates are based on the Central Electric equation for effective stack height, and it is clear that an important readjustment has been made. There may be a very slight tendency for P/O ratios to be higher close to the source, but the overprediction has been greatly reduced without seriously affecting estimates at 3.0 km and beyond.

Note also that the comparison between the ^{131}I and ^{133}I data support the reliability of the sample measurements themselves. Comparison between the predicted/observed ratios for the two isotopes leaves little doubt that the measurements are consistent.

The data shown in Table 3 were sufficiently encouraging to warrant study of the entire set of routine ^{131}I samples taken at approximately 2-week intervals during 1965. Several sampling periods had to be eliminated owing to interference from Chinese bomb tests and other uncertainties, but acceptable data were available for approximately 75 per cent of the year.

A series of fixed and temporary monitoring stations (Fig. 5) were used to provide general coverage of the entire site, as well as detailed data along the lines of the prevailing wind direction during the warm and cold halves of the year (SW and NW, respectively).

The prediction system differs slightly from that used in the intensive evaluation of the March data, primarily to keep the technique simple. The same sets of dispersion parameters as shown in Table 1 were used, but in this instance it was assumed that the overall mean wind speed associated with each dispersion condition applied whenever it was present. Adjustments for half life were made in the usual fashion, but it was assumed that a constant emission rate applied throughout each sampling period unless marked changes were known to have occurred. In all, some 17 sampling periods were investigated, and in the following summaries these data have been combined with the March series to obtain the largest number of samples possible for study.

TABLE 3. Comparison of observed and predicted samples for March 1965 series using Central Electric Δh equation.

Station	Distance (m)	^{131}I (Ci×10^{-12}) Obs.	Pred.	P/O	^{133}I (Ci×10^{-12}) Obs.	Pred.	P/O
E-11	700	35	50	1.4	380	520	1.4
		43	52	1.2	380	520	1.4
		35	17	0.5	150	115	0.8
		12	9	0.7	50	32	0.6
		14	8	0.6	—	—	—
Igloo	1750	100	220	2.2	700	2440	3.5
E-4	2200	6	3	0.5	—	—	—
E-7	2550	36	19	0.5	380	198	0.5
		75	115	1.5	740	1150	1.5
		47	45	1.0	48	78	1.6
		20	26	1.3	—	—	—
E-9	2750	10	4	0.4	20	14	0.7
		12	4	0.3	—	—	—

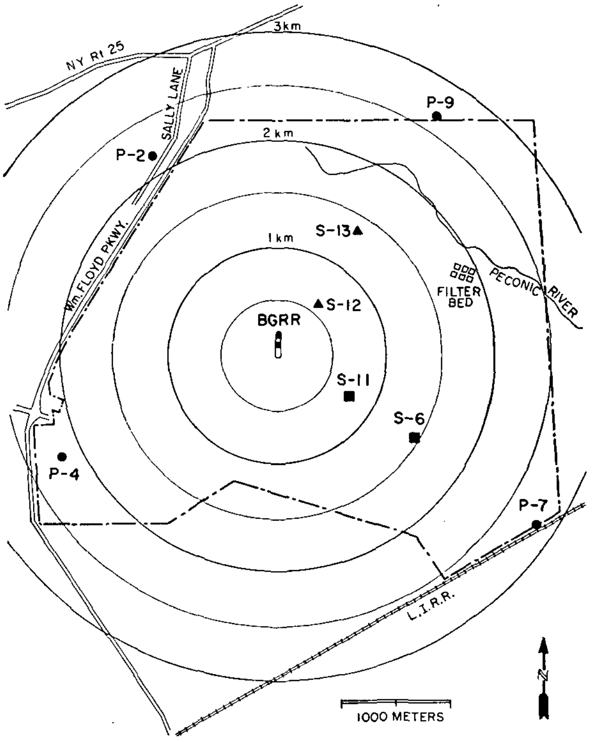

FIG. 5. Map of monitoring stations showing the locations of samplers used in this study.

There are many samples containing so little ^{131}I that they strain the resolution of the detection system, and it is equally true that if a sampling station is affected for no more than an hour or two in a 2-week period, these particular hours may not satisfy the assumptions of the simplified prediction model too well. The first processing of the data, therefore, was an attempt to isolate the reliable, meaningful cases. In Table 4 it is quite apparent that when the observed sample was assayed at less than 10^{-11} Ci, the scatter of the P/O ratios was enormous. Conversely, those samples containing more than 10^{-11} Ci fell into a much narrower and apparently a more orderly range.

Based on this separation and knowledge of the variability of the meteorological prediction scheme, it was decided to confine the analyses to samples having $\geq 10^{-11}$ Ci and estimated exposures of at least 8 hr. The latter restriction was of course not applied to a few of the samples obtained early in March where the precise plume model was used.

The readjustment of the estimates of effective stack height required to obtain reasonable P/O ratios in the early March series suggested study of the variation of the ratios with distance as a first step, and Table 5 shows such a breakdown. There are differences in the mean values of the P/O ratios and considerable scatter among individual cases, but it is extremely difficult to ascribe any features of the table to improper estimation of effective stack height. The two groups containing large numbers of samples (the closest and most distant stations) are not significantly different from each other and those at the intermediate distances have too few samples to show anything particularly startling. The obvious tendency for the P/O ratios to be smaller than 1.0 at all distances cannot be produced by a seriously incorrect estimate of source height, since the ratios do not change with distance. One may therefore conclude that the Central Electric equation on which these estimates were based accurately predicts the plume rise.

It is curious, however, to find that the geometric mean of all P/O ratios is 0.61, indicating a persistent tendency for the prediction system to underestimate the observed values. It is impossible with the available data to ascertain the reason for the underestimation, but there are at least two reasonable possibilities.

The wind speed parameter $\bar{u}$, appearing in the denominator of both Eqs. (1) and (2), applies strictly to a medium having a uniform flow throughout, and the atmosphere almost always shows a marked increase with height. Selection of a value of $\bar{u}$ is, therefore, an arbitrary process which cannot be guided by sound theoretical procedure since the mathematical model does not fit the natural atmosphere perfectly. The wind speeds used in all computations have been those at stack height, and this selection has been justified on the basis of its having appeared reliable for the oil fog tracer tests. It can be argued that a mean wind speed in the layer between the source and the ground is necessarily lower than that at stack height, and it is true that the use of such a reference wind would adjust the data in the proper direction. Following this line of reasoning, one would have to use the wind at approximately 50 ft to achieve a mean P/O ratio of 1.00, since typical daytime conditions (B_1 gustiness) prevail in these test cases and the change of wind with height is described by a $\frac{1}{4}$ power law. If improper estimation of $\bar{u}$ is wholly or partly to blame for the discrepancy, it is difficult to see why a fairly constant underestimation is not present in all portions of the data, but it appears in subsequent analysis that this is not the case.

Another possibility is that there may be a systematic overestimate of Q, the output rate. The only significant difference in any of the release values was that the March series came essentially from a single source whereas the iodine during the remainder of the year came from the reactor core generally. Comparison of these two groups does show that the 13 samples of early March had a mean P/O ratio of 0.80 while the 41 cases obtained during the rest of the year have a mean of only 0.56, but the authors cannot visualize any mechanism that could account for a discrepancy in the Q estimates.

Table 4. Distribution of samples by P/O ratios and amount collected.

P/O ratio	No. of samples with 10^{-13}–10^{-11} Ci	No. of samples with $>10^{-11}$ Ci
0.0–0.2	7	23
0.3–0.5	9	21
0.6–1.0	5	16
1.1–2.0	7	3
2.0–5.0	3	1
5.0–10.0	2	
>10	3	

Table 5. Predicted/observed ratios as a function of distance.

	Distance from source 500–999 m	1000–1499 m	1500–2200 m	2500–3000 m
	0.4	0.6	0.1	0.2
	0.6	0.4	0.6	0.4
	1.4	1.0	0.5	0.4
	1.2	0.8	0.9	0.3
	0.5	0.7	0.4	0.7
	0.7		5.5	0.9
	0.6		7.1	0.2
	0.5			0.3
	5.2			0.4
	0.3			0.5
	0.2			5.7
	0.1			0.4
	0.2			0.2
	0.4			0.8
	0.3			0.5
	0.2			1.5
	0.4			1.0
	0.5			1.3
	1.4			1.8
	0.7			1.2
				0.5
Mean	0.49	0.66	0.90	0.60
No. ⩾1.0	4	1	2	6
No. <1.0	16	4	5	15

All Data	
Mean	0.61
No. ⩾1.0	13
No. <1.0	40

Questions have frequently been raised concerning the difference in turbulence and dispersion in air coming from overwater and overland trajectories. It has not been possible to discover strong evidence of such differences in the Brookhaven wind studies, but this is not surprising, since air must travel over at least 8 miles of land before reaching the Laboratory, and some water surface would be involved with either the typical winter NW or the summer SW winds.

Table 6 shows the breakdown of P/O by wind direction, however, and one does find a very obvious difference between the SW and NW groups, the latter having a mean P/O ratio almost twice that of the SW set. Although this difference may be associated with the water vs. land trajectories, it is perfectly possible that it could be caused primarily by some seasonal difference in turbulence, appearing to be associated with wind direction only because the wind directions themselves are seasonably distributed. The NW set of data cannot be tested in this manner because there are too few

TABLE 6. Predicted/observed ratios as a function of wind direction.

	Wind direction (deg)			
	063	140	217	300–321
	0.5	0.1	0.2	0.4
	0.9	0.6	0.4	0.6
	7.1	0.4	0.4	1.4
		5.5	0.3	1.2
			0.7	0.5
			0.9	0.7
			0.2	0.6
			0.3	0.5
			0.4	5.2
			0.5	0.3
			5.7	0.2
			0.4	0.4
			0.6	0.5
			0.4	1.4
			1.0	0.7
			0.8	0.2
			0.7	0.8
			0.2	0.5
			0.1	1.5
			0.2	1.0
			0.4	1.3
			0.3	1.8
				1.2
				0.5
Mean	1.50	0.60	0.44	0.72
No. ⩾1.0	1	1	2	9
No. <1.0	2	3	20	15

TABLE 7. Seasonal distribution of predicted/observed ratios of samples obtained with SW winds.

	Oct.–March	April–Sept.
	0.4	0.2
	0.3	0.1
	0.7	0.2
	0.8	0.6
	0.2	0.4
	0.4	1.0
	0.4	0.9
	0.3	0.2
	5.7	0.3
	0.4	0.4
Mean	0.54	0.35

summer cases, but it is possible to compare the summer and winter seasons for the SW group (Table 7). Here also a noticeable difference appears, with the P/O ratios being obviously lower in summer than in winter.

Thus, from Tables 6 and 7, one would conclude that there are noticeable variations in long-term dispersion associated with both the trajectory of the air flow and with the season, but it is difficult to make a more detailed statement with the limited data available.

The discussion so far has been directed toward obvious and suspected flaws in both the prediction and evaluation systems, and the errors have been emphasized. It should also be clear that the data have shown the prediction system to be an excellent means of estimating long term dispersions of the reactor plume, probably more accurate than engineering estimates (as opposed to measurements) of pollutant emission rates, and therefore certainly adequate for general use.

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