Sandia National Laboratories

P.O. Box 5800
Albuquerque, New Mexico 87185

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May 7, 1994

Dr. R. Y. Lee
Accident Evaluation Branch, NLN344
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555-0001

Re: Comments on Doctoral Thesis entitled "The Chernobyl Accident Revisited: Source Term Analysis and Reconstruction of Events During the Active Phase"

Dear Dr. Lee:

In response to your request, I have examined the technical issues raised in the doctoral thesis submitted to the Massachusetts Institute of Technology by Alexander Sich and entitled "The Chernobyl Accident Revisited: Source Term Analysis and Reconstruction of Events During the Active Phase." My comments on this rather polemical thesis are enclosed. I have tried in these comments to address two technical issues:

- Was the release of radioactivity from the Chernobyl site substantially greater than estimated shortly after the accident?
- Is close examination of the Chernobyl accident likely to improve our understanding of hypothetical severe accidents in Western reactors?

I conclude in my comments, in contrast to Sich, that the answer to both of these questions is no. I still believe the accident holds little important insight to the progression of severe accidents in Western reactors. There may be some information of use from the Chernobyl accident concerning the behavior of radioactive materials in the environment. This, however, is a topic not addressed by Sich.

I would, of course, be happy to discuss my comments further with you and your associates.

Sincerely yours,

Dana A. Powers

Dana A. Powers

cc: T. Speis (USNRC)
N. Grossman (USNRC)
C. Alexander (BCL)
T. Kress (ORNL)
N. Ortiz 6400

Enclosure 3

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Comments by D. A. Powers on "The Chernobyl Accident Revisited: Source Term Analysis and Reconstruction of Events During The Active Phase" by A. R. Sich

The thesis "The Chernobyl Accident Revisited: Source Term Analysis and Reconstruction of Events During the Active Phase" is a welcome re-examination of the accident that improves upon the understanding of this serious nuclear reactor accident derived in the hectic months shortly after the accident [1]. Sich has done a service to the reactor safety community compiling additional descriptive information that has come available since 1986. Whether such a polemical if not political thesis is appropriate for fulfilling the requirements of a doctorate in nuclear engineering is outside the scope of this examination. The very limited quantitative analysis presented in the document makes it difficult to assess the plausibility arguments made throughout the thesis.

In the next two subsections of these comments, I examine two of the points Sich tries to make. In the first of these subsections, I examine Sich's contention that the radionuclide release during the Chernobyl accident was much greater than previously thought. In the second subsection, I examine his contention that the improved understanding of the accident shows it holds important insight into severe accidents at Western reactors. In the third subsection, I comment on other technical and not-so-technical points raised by Sich.

A. Magnitude of Radionuclide Release

Soviet scientists assembled a massive tome of information about the Chernobyl accident and shared it with the world reactor safety community just four months after the accident [1]. It is not surprising then that this information might not be as accurate and complete as information now available. Sich berates the Soviets for underestimating radionuclide releases during the accident. The Soviets, however, were very clear in their presentation of the information that the releases they reported were only for materials deposited in the USSR. They did not, at the time, have access to radionuclide data from other countries nor did they feel they had time to digest such data from other countries. They thought it would be presumptive to tell how much fallout had landed in other countries!

It was precisely because of the limitations of the data presented by the Soviet scientists that Warman [15] and others undertook an effort to define the total magnitude of radionuclide releases from the Chernobyl accident. All at the Vienna meeting understood that the releases cited by the Soviets were not complete especially for the very volatile radionuclides, cesium and iodine. Soviets undertook the borehole sampling of core debris in Chernobyl so that they could get better estimates of the total release.

Sich, however, claims that releases were even greater than later Western estimates. In large part, his estimates of larger releases come from assuming a monumental inventory for ^{136}Cs. I have been unable to discover the reason Sich would claim the ^{136}Cs inventory is so large as

169 x 10^6 curies.* This is a factor of 50 to ŀ0 larger than any of the previous estimates. To examine Sich's arguments I have:

- compared Sich's estimates of the inventories to those made with the CINDER code by Los Alamos,
- compared Sich's estimate of the ^{136}Cs inventory to those made by others with the ORIGEN code, and
- compared $^{136}Cs/^{137}Cs$ ratios predicted with inventories estimated by Sich to measurements made of fallout collected in several countries.

Details of these comparisons are described in the subsections below. The comparisons uniformly indicate that Sich's estimate of the ^{136}Cs inventory is grossly high.

There is abundant evidence in Sich's thesis that points to an error in the ^{136}Cs inventory. When he attempts to rationalize the release fractions of radionuclides using corrected measurements and his inventory, he concludes that ^{136}Cs is released to an extent 1 to 2 orders of magnitude less than ^{134}Cs and ^{137}Cs. We might well expect ^{136}Cs to be somewhat slower to release initially than ^{134}Cs and ^{137}Cs. ^{136}Cs is a short half-life radionuclide ($t_{1/2}$ ~13.19 days) so more of this isotope would be found in the grains of the fuel rather than at grain boundaries in comparison to longer half-life radionuclides such as ^{134}Cs ($t_{1/2}$ = 2.06 years) and ^{137}Cs ($t_{1/2}$ = 30 years). There had been more operating time for the long-life isotopes to migrate to grain boundaries where release from the fuel can more easily occur. But, in low burnup fuel such as that at Chernobyl, the differences in release rates of the isotopes of cesium ought to be less than a factor of 2 and certainly not orders of magnitude. It is far easier to explain the releases of ^{136}Cs reported by Sich in comparison to releases of ^{134}Cs and ^{137}Cs in terms of an incorrect inventory for ^{136}Cs than in terms of different release mechanisms.

1. COMPARISON OF ESTIMATED CESIUM ISOTOPE INVENTORIES

Cesium isotope inventories provide useful indicators of reactor behavior. The ratio of $^{134}Cs/^{137}Cs$ is usually considered a good indicator of fuel burnup since this ratio increases almost linearly with burnup. This is shown in Figure 1 for a calculation with the CINDER code for a 3200 MW reactor. The $^{136}Cs/^{137}Cs$ ratio decreases with initial burnup to a plateau that is indicative of the somewhat recent operating power (~1 month) of the reactor. Because ^{136}Cs is produced by the activation of a relative stable cesium isotope, ^{135}Cs, the plateau in power reactors is relatively insensitive to burnup.

Cesium isotope inventories of the Chernobyl reactor at the time of the accident (April 26, 1986) estimated by A. Sich [1] and others are shown in Table 1. Inventories estimated by most investigators assume a uniform fuel burnup varying from about 10500 to 12600 MWd/t. For this

* The most plausible explanation has been suggested by L. Soffer. He suggests that Sich has assumed equilibrium concentrations of ^{136}Cs so the fission yield (6.3%) matches the rate of decay ($t_{1/2}$ = 13.16 days).

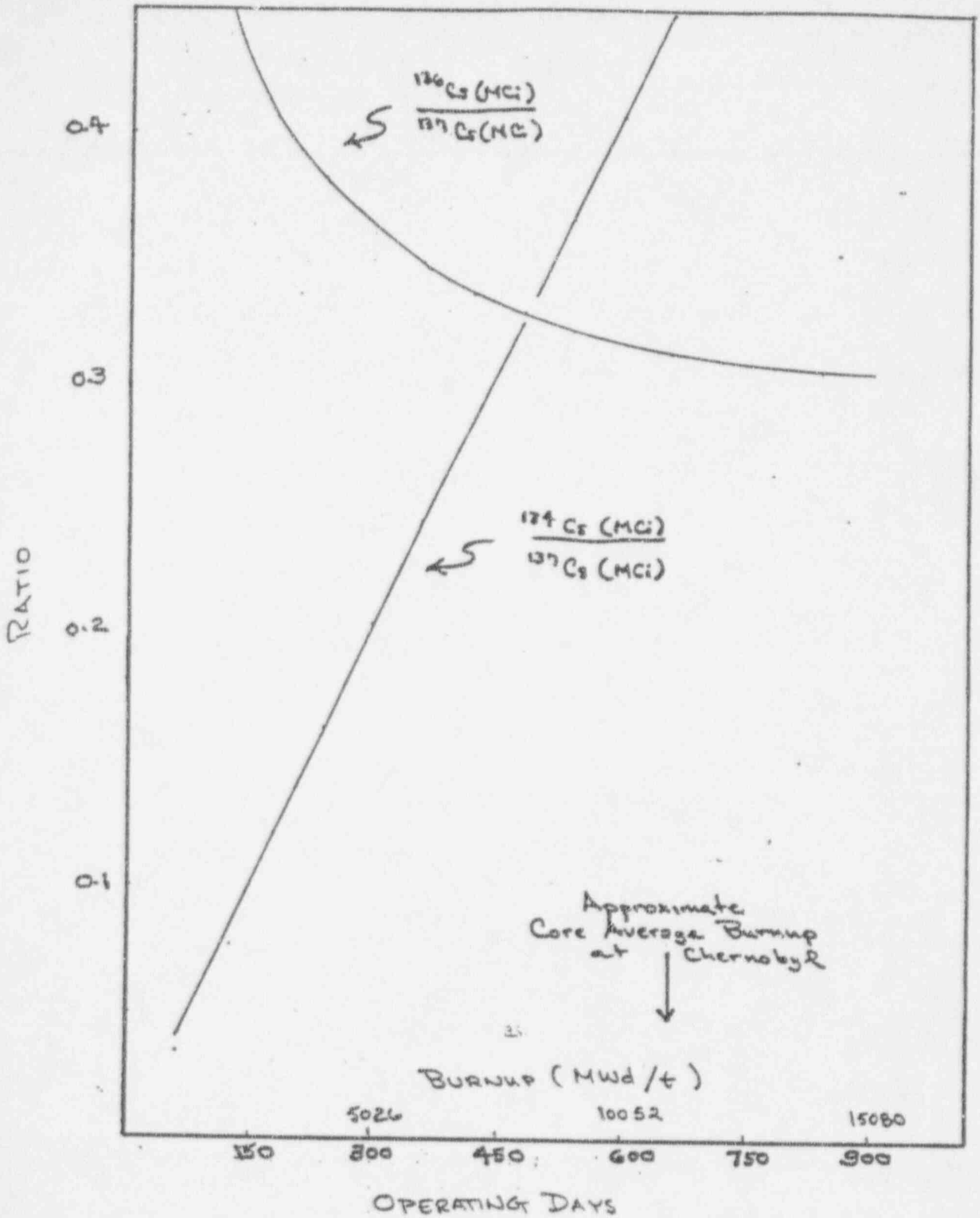

Figure 1. Behavior of $^{136}Cs/^{137}Cs$ and $^{134}Cs/^{137}Cs$ Ratios With Fuel Burnup

Table 1. Estimates of Cesium Isotope Inventories

Isotope	A. Sich [1]	G. Kirchner C. S. Noack [2]	Begicher et al. [3]	[4]	This work
^{134}Cs	4.6	4.3	5.0	5.6	3.3 ± 0.46
^{136}Cs	169	2.3	no estimate	1.7	2.06 ± 0.17
^{137}Cs	7	8.3	7.7	5.9	6.59 ± 0.55
$^{134}Cs/^{137}Cs$	0.66	0.52	0.65	0.95	0.501 ± 0.081
$^{136}Cs/^{137}Cs$	24	0.28	--	0.29	0.313 ± 0.036

work, the variability in the burnup of various fuel assemblies [3] was taken into account as shown in Table 2. Inventories as a function of burnup were taken from approximate descriptions of an RBMK-1000 reactor obtained with the CINDER code. ^{134}Cs isotope inventories are predicted to be somewhat lower than other estimates. Otherwise, the estimates of the inventories are all in acceptable agreement except the inventory of ^{136}Cs made by Sich. His estimate seems to be physically impossible for normal uranium oxide fuel. His estimate would be plausible only if the fuel had been deliberately doped with ^{135}Cs. There is no evidence that this was done.

Table 2. Estimation of Cesium Inventories Considering Varying Burnup of the Chernobyl Fuel

Number of Fuel Assemblies	Burnup Range (MWd/t)	Inventory of Whole Core With Indicated Burnup (MCi)		
		^{134}Cs	^{136}Cs	^{137}Cs
721	14766 - 13029	5.251 - 4.101	2.717 - 2.421	8.888 - 7.858
392	13029 - 11292	4.101 - 3.092	2.421 - 2.133	7.858 - 6.824
154	11292 - 9554	3.092 - 2.221	2.133 - 1.851	6.824 - 5.786
101	9554 - 7817	2.221 - 1.491	1.851 - 1.572	5.786 - 4.748
35	7817 - 6080	1.491 - 0.919	1.572 - 1.288	4.748 - 3.700
43	6080 - 4343	0.919 - 0.475	1.288 - 0.992	3.700 - 2.650
41	4343 - 2606	0.475 - 0.171	0.992 - 0.672	2.650 - 1.592
172	2606 - 0	0.171 - 0	0.672 - 0	1.592 - 0
Weighted Inventory		3.758 - 2.846	2.224 - 1.893	7.132 - 6.040
		(3.302 ± 0.456)	(2.059 ± 0.166)	(6.586 ± 0.546)

2. COMPARISON OF ISOTOPE RATIOS IN FALLOUT

The RBMK-1000 reactor at Chernobyl is a complicated machine. The precise loading, burnup, and power history of the reactor is still not known with precision. This makes calculation with any code questionable. Another key to the cesium isotope inventories is provided by fallout collected at many locations following the accident. If ^{136}Cs was as abundant as expected by Sich, unusual isotopic ratios would be found in the fallout since it is difficult to imagine how ^{136}Cs release could be dramatically different than ^{134}Cs release or ^{137}C release.

Because the isotopes have long half-lives, the $^{134}Cs/^{137}Cs$ ratio ought to be time invariant over the several days of the accident. The $^{136}Cs/^{137}Cs$ ratio ought to decrease with time:

Days After Accident	Date	Relative $^{136}Cs/^{137}Cs$ Ratio
0	25/APRIL	1.0
1	26	0.949
2	27	0.900
3	28	0.854
4	29	0.810
5	30	0.768
6	1/MAY	0.729
7	2	0.692
8	3	0.656
9	4	0.622
10	5	0.591
11	6	0.560
12	7	0.532
13	8	0.504
14	9	0.478
15	10	0.454

Some isotope ratios reported from various countries at various dates are shown in Table 3. Though there is significant scatter among the results, the results are, by and large, consistent with the estimates of inventories made by others and shown in Table 1. That is, at the time of the accident, the $^{134}Cs/^{137}Cs$ ratio was between 0.47 and 0.60. The $^{136}Cs/^{137}Cs$ ratio was between about 0.24 and 0.32. None of the evidence supports the very large value of the ^{136}Cs inventory published by Sich. Before much is made of this large value, Sich should show

- the code used for the prediction is valid, and
- the mechanism for inhibition of the release of ^{136}Cs.

I am not familiar with the WIMS and CACHE codes used by Sich to obtain his isotopic inventories and he provides no references for these codes. In view of the extensive validation of the ORIGEN

Table 3. Isotope Ratios in Fallout

Country	Date	^{134}Cs(Ci)/^{137}Cs(Ci)	^{136}Cs(Ci)/^{137}Cs(Ci) (value at 25/April)	Ref.
Finland				
air samples	29/APR	0.57	0.19 (0.23)	6
	30/APR	0.60	0.27 (0.35)	6
	28/APR	0.611	0.255 (0.30)	6
		0.586	0.228 (0.27)	6
		0.650	0.260 (0.30)	6
		0.562	0.219 (0.26)	6
	30/APR	0.517	0.183 (0.24)	6
	2/MAY	0.569	0.170 (0.25)	6
		0.558	0.163 (0.24)	6
		0.562	0.140 (0.20)	6
	3-5/MAY	0.577	0.185 (0.30)	6
	8/MAY	0.562	0.125 (0.25)	6
	15/MAY	0.517	0.083 (0.24)	6
	16/MAY	0.513	0.0795 (0.24)	6
Hungary	5/MAY	0.56	0.22 (0.37)	7
	6/MAY	0.56	0.2 (0.36)	7
	7/MAY	0.48	0.12 (0.23)	7
	8/MAY	0.48	0.13 (0.26)	7
	9/MAY	0.48	0.117 (0.24)	7
	30/APR	0.89	0.33 (0.43)	7
	1/MAY	0.63	0.22 (0.30)	7
	3/MAY	0.54	0.21 (0.32)	7
	5/MAY	0.51	0.15 (0.25)	7
	6/MAY	0.61	0.19 (0.34)	7
United Kingdom				
air sample	3/MAY	0.533	--	8
grass	6/MAY	0.541	--	8
Sweden				
ground contamination	30/APR	0.793	--	9
		0.727	--	9
		1.14	--	9
Netherlands	2-9/MAY	0.458	--	10
"Kiev Trouser"		0.42	--	11
"Minsk Shoe"		0.69	--	11
"NE Poland Trouser"		0.43	--	11

Table 3. Isotope Ratios in Fallout (Concluded)

Country	Date	$^{134}Cs(Ci)/^{137}Cs(Ci)$	$^{136}Cs(Ci)/^{137}Cs(Ci)$ (value at 25/April)	Ref.
Germany	3/MAY	0.556	0.166 (0.25)	2
		0.452	0.110 (0.17)	2
	6/MAY	0.589	0.166 (0.30)	2
		0.507	0.137 (0.24)	2
	14/MAY	0.462	0.151 (0.41)	2
		0.523	0.138 (0.38)	2
		0.512	0.088 (0.24)	2
		0.468	0.087 (0.24)	2
		0.537	0.085 (0.23)	2
		0.526	0.087 (0.24	2
		0.578	0.126 (0.34)	2
		0.562	0.087 (0.24)	2

and CINDER codes, it is difficult to ignore predictions of these codes. This is especially true when all other evidence suggests Sich has incorrectly stated the ^{136}Cs inventory.

B. Pertinence of Chernobyl to Hypothetical Severe Accidents in Western Reactors

Sich contends that information derived from the events of the Chernobyl accident are pertinent to the understanding of severe accidents in western boiling water reactors and pressurized water reactors. He seems to understand that western reactors are not susceptible to the exact type of accident experienced at Chernobyl. (Indeed, I suspect RBMK reactors are also not susceptible to a repetition of this exact accident!) His contention seems, instead, to be based on the dilution, spreading and freezing of core debris in the lower regions of the reactor. Sich contends that these observations suggest an excellent basis for design of a core retention system.

As noted by Sich, the idea of dilution of core debris as a core retention device has long been known. There have even been tests of the concept in the U.S. [12] and in Germany [13]. What Sich does not address is how to design such a device for high power density fuel that has not cooked for several days before contacting the sacrificial material. Nor does he address compatibility issues for core debris rich in metallic zirconium. Simple scoping calculations would show that rather thick sacrificial beds would be required. Retrofitting existing reactors with such beds would be nearly impossible. Furthermore, such beds would not substantially affect risk estimates for reactors [14].

Sich concludes (p. 410) that Chernobyl may place an upper bound on severe reactor accidents at Western reactors. I think this conclusion, which is not supported by anything in the main text, is not well conceived. Certainly, it ignores issues of:

- energetic plumes at Chernobyl,
- containment bypass accidents, and
- higher burnup fuels.

In any event, I have no idea how this bound might be employed even if it were recognized as a bound.

Sich points to a substantial retention of cesium in the Chernobyl core debris (p. 335). He does not attempt to explain this retention. But, fuel exposed to air at modest temperatures might well react to form cesium uranates that are of low volatility. This might have some pertinence to issues of air intrusion into the coolant system of Western reactors late in a severe accident.

C. Other Topics

Sich has found that most of the materials dropped into the Chernobyl reactor did not fall on the core debris. This is an interesting finding. His contention that this refutes claims made by the Soviets that the dropped material mitigated the accident is inaccurate. Soviets did not make such claims. They had release data showing it did not.

At the meeting in Vienna, the Soviets never claimed the nitrogen purge arrested the accident. They were themselves unsure about the relative timing of the start of the purge and the sudden drop in the rate of radionuclide release.

Sich has found that most of the graphite at Chernobyl burned. In August of 1986, there were two schools of thought among the Soviet scientists on graphite combustion. There was an official position that only 20 percent burned. There was a significant body of scientists that disagreed and felt most of the graphite burned. Sich's finding supports this dissenting group.

Sich contends that there is now proof carburization was not responsible for release of refractory radionuclides (p. 331). After all the graphite has burned away, any carbides formed would also have been oxidized. Sich does not provide an explanation of why both ruthenium and barium releases were so high. Typically, oxidizing conditions enhance ruthenium release and suppress barium release, but both were high at Chernobyl. Carburization does provide such an explanation. Sich's stages 1 and 2 of the accident provide the opportunity for carburization to occur. He presents no quantitative analysis to show it will not. All evidence of carburization would be destroyed by accident stages after the sixth day of the accident.

Sich has taken more recently available data bases on radionuclide releases and attempted more refined analyses of the release process. Unfortunately, he attempts nothing quantitative. Qualitative results he obtains are not greatly different than what was known in 1986. He concludes there was a prolonged period of isothermal release. But, since rates are so similar for elements of greatly different volatilities, it might be better to conclude there was a prolonged period in which release was limited by gas phase mass transport.

As an aside, Sich spends a lot of time wondering about the source of early (April 26) release data. He concludes it was not obtained from air sampling. Rather it was obtained from integrated ground deposition measurements. He is correct in this conclusion. This is exactly what the Soviets said when they presented the data in the meeting in Vienna.

There are a variety of troublesome features about Sich's thesis. Not the least of these is the points he makes about childhood thyroid cancers. Sich presents no data or analysis. His discussions of the point are by reference to mainly nonscientific sources. His treatment of the issue goes from speculation in the early chapters to a definite link between releases and cancer incidences in the conclusions. I can find nothing in the thesis that supports this evolution in the accident consequences.

REFERENCES

1. U.S.S.R State Committee on the Utilization of Atomic Energy, The Accident at the Chernobyl Nuclear Power Plant and Its Consequences, report presented at the International Atomic Energy Agency's Experts' Meeting, August 25-29, 1986, Vienna, Austria.

2. G. Kirchner and C. C. Noack, Nuclear Safety 29 (1988) 1.

3. S. N. Begicher, A. A. Borovoj, E. V. Burlakov, A. Ju. Gagarinsky, V. F. Demin, I. L. Khodakovsky, and A. A. Khrulev, "Radioactive Releases Due to the Chernobyl Accident," International Seminar Fission Product Transport Processes in Reactor Accidents, Dubrovnik, Yugoslavia, 1989.

4. Technical Research Centre of Finland, "First Rough Estimation (1.5.1986) of Source Strength at Chernobyl."

5. Not used.

6. Finnish Centre for Radiation and Nuclear Safety, Second Interim Report Radiation Situation in Finland From 5 to 16 May 1986, STUK-B-VALO 45, Government Printing Center, Helsinki, Finland, May 1986.

7. A. Andrasi et al., Monitoring the Radiation Consequences Due to the Disaster at the Chernobyl Nuclear Facility From April 28 to June 12, 1986.

8. S. Nair and P. J. Darley, A Preliminary Assessment of Individual Doses in the Environs of Berkely, Gloucestershire Following the Chernobyl Nuclear Reactor Accident, TPRD/B/0796/R86, Central Electricity Generating Board, Berkeley, Gloucestershire, UK, May 1986.

9. M. Jensen and J.-C. Lindhe, Activities of the Swedish Authorities Following the Fallout From the Soviet Chernobyl Reactor Accident, 2nd revised preliminary report, National Institute of Radiation Protection, Stockholm, Sweden, 12 May 1986.

10. "Summary of Radiological Measurements Related to the Chernobyl Accident Carried Out by the Netherlands Energy Research Foundation Petten," presentation to the NEA/CSNI Principle Working Group 4 Meeting, June 12, 1986, Paris, France.

11. J. van der Veen, A. van der Wijk, and W. G. Mook, Nature 323 (1986) 399.

12. D. A. Powers, "A Survey of Melt Interactions with Core Retention Materials," p. 379, Proceeding of the International Meeting Fast Reactor Safety Technology, Seattle, WA, August 19-23, 1979.

13. Experiments and analyses by Dalle-Donne at the Kernforschungszentrum, Karlsruhe, Germany, on borax core retention devices.

14. NUREG-1150.

15. E. A. Warman, Soviet and Far-Field Radiation Measurements and an Inferred Source Term From Chernobyl, TP 87-13, Stone and Webster Engineering Corp. Boston, MA, 1987.