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EARLY RESULTS ON THE LONG-RANGE TRANSPORT OF CHERNOBYL RADIOACTIVITY

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• Environment Canada

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MANAGEMENT PERSPECTIVE/EXECUTIVE SUMMARY:

The measured levels of the Chernobyl radionuclides in Canada are compared with those reported for various locations in western Europe. The levels in Canada,about 10,000 times lower than the average levels in Europe,are unlikely to have reached Canadian populations deriving drinking water from municipal sources.Nor are the radionuclides being detected currently likely to persist in the Canadian aquatic environment. On the other hand,cesium-137 with a half-life of 30 years will persist in the local aquatic ecosystems in some European countries where it is likely to obliterate the 1963 weapons-testing fallout activity maximum. In such locations the Chernobyl cesium-137 will constitute a new marker for dating sediments.Possible use of the Chernobyl radionuclides for tracing tropospheric transport patterns is pointed out.The detection of some radionuclides arising from an additional source,perhaps the 10 April 1986 accident at the Nevada test site,is further ascertained. The fractionation of different radionuclides is reported.It is pointed out that in the absence of zirconium-95,the reference radionuclide in reporting fractionation factors in all previous studies of nuclear fallout transport,the fractionation factors are readily explained using cesium-137 as the reference radionuclide.

RESULTATS PRELIMINAIRES DE L'ETUDE SUR LE TRANSPORT A GRANDE DISTANCE DE LA RADIOACTIVITE DE TCHERNOBYL

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SOMMAIRE

Les teneurs en radionucléides provenant de Tchernobyl mesurées au Canada sont comparées à celles qui sont signalées à divers points de l'Europe de l'Ouest. Il est très peu vraisemblable que les teneurs canadiennes, d'environ 10 000 fois inférieures aux teneurs européennes, aient atteint les populations canadiennes qui tirent leur eau de boisson de source municipale. De plus, il est plus probable que les radionucléides présentement détectés persistent longtemps dans le milieu aquatique canadien. Par ailleurs, le césium-137, qui a une période de 30 ans, persistera dans les écosystèmes aquatiques locaux de certains pays européens, ce qui entraînera vraisemblablement un dépassement du maximum de 1963 dû aux retombées des essais nucléaires. Dans ces endroits, le césium-137 de Tchernobyl constituera un nouveau marqueur permettant de dater les sédiments. L'utilisation possible des radionucléides comme traceur pour mesurer les profils de transport dans la trophosphère a été soulignée. La détection de certains radionucléides provenant d'une source additionnelle, peut-être l'accident du 10 avril 1986 sur le site d'essai du Nevada, fait également l'objet d'études supplémentaires. On signale le fractionnement des différents radionucléides. On souligne qu'en l'absence de zirconium-95, qui est le radionucléide de référence servant à déterminer les facteurs de fractionnement dans les études antérieures de transports de retombées radioactives, les facteurs de fractionnement sont facilement expliqués si l'on utilise le césium-137 comme radionucléide de référence.

ABSTRACT

The levels of Chernobyl-derived radionuclides were measured in several rain,snow and water filtration plant floc samples collected from various locations in Canada.The data indicate that the Chernobyl release had negligible effect on the radiological quality of Lake Ontario waters.The detection of some activation products in pre-Chernobyl samples ,presumably arising from the accidental release of radioactivity during weapon testing at the Nevada site,is also reported.Some data from the ongoing measurements on rain samples are presented to discuss the possible use of Chernobyl radionuclides for delineating tropospheric transport processes.Fractionation factors for the major radionuclides are also reported.

REFERENCE

Les teneurs en radionucléides provenant de Tchernobyl ont été mesurées dans plusieurs échantillons d'eau de pluie, de neige et d'usines de filtration d'eau (floc), recueillis dans plusieurs endroits au Canada. Les données indiquent que la radioactivité de Tchernobyl a un effet négligeable sur la qualité radiologique des eaux du lac Ontario. La détection de certains produits radioactifs dans des échantillons d'avant Tchernobyl, provenant probablement de la libération accidentelle de radioactivité au cours d'un essai nucléaire au Nevada, est aussi signalée. Certaines données provenant des mesures actuelles d'échantillons de pluie sont présentées pour permettre un examen des utilisations possibles des radionucléides de Tchernobyl, qui pourraient servir à mesurer les processus de transport dans la troposphère. Les facteurs de fractionnement des principaux radionucléides sont également signalés.

RESULTATS PRELIMINARIES DE L'ETUDE DU TRANSPORT A GRANDE DISTANCE
DE LA RADIOACTIVITE DE TCHERNOBYL
S.R. Joshi

INTRODUCTION

In the aftermath of the accident at the Chernobyl nuclear power plant,the well-founded anxiety about immediate hazards to humans is slowly being replaced by concerns about the long-term consequences of the disastrous radioactive release.Concomitant with these concerns is a need for understanding the environmental processes responsible for the transport of Chernobyl radioactivity.This need is further highlighted by the realization that some of the major differences between the Chernobyl fallout and the nuclear weapons-testing fallout (Table 1) preclude direct application of the conclusions drawn from the weapons-testing fallout studies.In retrospect,therefore,the Chernobyl release paralleled a major emission of industrial pollutants to the atmosphere more closely than the release of radioactivity from an above-ground nuclear explosion.This apparent similarity with industrial stack emissions makes Chernobyl-delivered radionuclides very useful tracers for studying the long-range transport of pollutants released to the lower atmosphere.The present report gives early results of ongoing NWRI measurements designed to assess the impact of the Chernobyl release on the Canadian aquatic environment and to characterize the long-range transport of this radioactivity.The data collected thus far indicate that measureable amounts of the Chernobyl radioactivity were present in Canadian rains at least until mid-june, 1986.This observation may be compared to the general reckoning that the mean tropospheric aerosol residence time is about one month with the air in the lower 3000m being washed clean on the average about every three days (Libby,1959).

MATERIALS AND METHODS

Sample locations

Rain samples were collected from locations shown in Fig.1 using NWRI-designed automatic precipitation collectors.

Analytical Techniques

The samples were analyzed by high-resolution gamma-ray spectrometry using hyperpure germanium detectors in planar and coaxial configurations.The characteristics of the detectors and the low-background shielding used have been described earlier(Joshi,1985a). Frequent assays of reference materials procured from the International Atomic Energy Agency and the U.S.National Bureau of Standards,and the results from regular participation in intercomparison programmes organized by the U.S.Environmental Protection Agency and the IAEA form the backbone of our quality assurance programme with respect to identification and energy calibration.Each sample was counted for 2.5×10^5 seconds or more.Very low levels of some radionuclides or small sizes of the samples available resulted in relatively high counting errors in some instances;the presence of the radionuclide(s) concerned, however,was never in doubt.The radioactivity levels reported in the present study are corrected for decay to mid-point of the collection period.

RESULTS

The results of our measurements on the Burlington rain samples,collected only from May 6 to May 24,1986,are shown in Fig.2. In order to assess the impact of Chernobyl fallout on Lake Ontario waters,three floc samples from the Burlington water treatment plant were also assayed (Roy et al.,1979;Durham and Joshi,1981).The results of these measurements are given in Table 2.Following detection of ^{65}Zn and ^{110m}Ag in the May 6-7 Burlington rain sample,some rain samples and a snow sample from other locations were also analyzed. The collection dates for these samples correspond to actual or predicted pre-Chernobyl time periods.Radionuclides detected in these samples and their levels are given in Table 3.

DISCUSSION

The results given in Fig.1 show that the Chernobyl radioactive cloud reached Burlington on 6-7 May,in agreement with the air transport model-based prediction by Environment Canada's Atmospheric Environment Service.Two phases are indicated from the measurements. That the two phases are distinct is evident from their radioisotopic composition (Fig.2) and radionuclide activity ratios.While the first activity maximum (14-16 May) is characterized by $^{137}Cs/^{134}Cs$ and $^{137}Cs/^{103}Ru$ activity ratios of about 1.7 and 1.9,respectively,the corresponding ratios in the second activity maximum (20-24 May) are

2.1 and 0.9.These activity ratios are similar to those obtained for samples collected in Sweden (Devell et al.,1986),UK (Fry et al.,1986), West Germany (Hohenemser et al.,1986),and USA (Bondietti and Brantley, 1986).Two separate phases have also been observed in Sweden (Devell et al.,1986) and in the USA (Bondietti and Brantley,1986).The two separate maxima reported in the Swedish study were observed about 10 days apart,while the maxima reported in the present study are separated by about seven days.The detection of ^{237}Np,via the 312-keV gamma-emission of its daughter,^{233}Pa,in the 20-24 May rain sample suggests that the second phase was probably derived from the 'northern' cloud which was relatively richer in transuranics than the 'southern' cloud (Hohenemser et al.,1986).

The levels of the artificially-produced radionuclides ^{137}Cs and ^{125}Sb in Lake Ontario waters (Table 2) are very similar to those measured previously in the nearshore (Durham and Joshi,1981) and open (Durham and Joshi,1984) waters of the lake.Unlike ^{137}Cs and ^{125}Sb which are known to be persistent in the lake as a result of their relatively long half-lives and significant production in the nuclear weapons testing,the remaining three radioisotopes are unlikely to persist following introduction of weapons-testing fallout in the lake. We have previously detected both ^{103}Ru and ^{106}Ru in the Lake Ontario nearshore waters (Durham and Joshi,1981),but not ^{65}Zn and ^{131}I though the latter is extensively used in the nuclear medicine procedures in the area medical facilities.The levels of ^{103}Ru obtained in the present

investigation are somewhat higher than those measured earlier (Durham and Joshi,1981). Even if the levels of these particle-reactive radionuclides were significantly higher than those prevailing during the sampling period,most of the extraneous radioactivity would be removed from the raw lakewater during the treatment process since the floc is known to effectively scavenge a large variety of radionuclides including those detected in the present study (Roy et al., 1979,1981;Durham and Joshi,1981).Radioelements with conservative chemical behaviour,such as strontium,if present,would not be retained on the floc.The data available todate,however,suggests that very little strontium activity was released from Chernobyl.Our own measurements support these observations since other non-volatile are either not detected (^{95}Zr) or are present in extremely low amounts (^{141}Ce and ^{144}Ce). It is thus reasonable to infer that very little,if any, Chernobyl radioactivity reached populations deriving water from municipal water supplies located around large freshwater bodies in Canada.

A corollary to the results on Lake Ontario waters provides that the Chernobyl-delivered ^{137}Cs is unlikely to serve as a 'marker' in the Canadian aquatic environment.Similarly,by inference,the very low levels of this radionuclide that might reach the bottom sediments are not expected to alter the sedimentary ^{137}Cs profiles in the Canadian lakes (Joshi,1985b and references therein) by downward migration.In contrast,in several locations in Europe where localized Chernobyl ^{137}Cs fallout was several times higher than that from

extensive weapons testing in the late 1950's and the early 1960's, this ^{137}Cs should serve as a new,distinct horizon for dating sediments. Figure 3 depicts the reported (Devell et al.,1986;Fry et al.,1986; Hohenemser et al.,1986; FCRNS,1986;WHO,1986) levels of deposition of Chernobyl-derived ^{137}Cs in some West European countries.Depending upon the sedimentation rate and the extent of mixing of old and new sediments,these levels could either dwarf or completely obliterate the 1963 weapons-testing fallout ^{137}Cs activity maximum in the bottom sediments.

An interesting aspect of the measurements on the Burlington rain samples (Fig.2) involves the detection of activation products ^{65}Zn and ^{110m}Ag in the 6-7 May rain.While the Chernobyl ^{110m}Ag has been detected in the UK (Jones et al.,1986) and in Canada (J.-C.Roy, Laval University,personal communication,1986), the presence of ^{65}Zn in the Chernobyl fallout has not been reported.It is quite possible that the detected low levels of these radionuclides in our samples arise from a source other than the Chernobyl reactor.In order to check this possibility,one snow sample and four rainwater samples from different locations and corresponding to pre-Chernobyl time periods (predicted or actual) were also analyzed.The results,given in Table 3, show that several fission and activation products,including ^{65}Zn, are invariably detected in samples collected after April 8,1986.The occurrence of small peaks in the 696-keV region of the gamma-ray spectra of some of the analyzed samples,notably the 8-21 April rain,also suggests the presence of antimony isotope(s).Unfortunately,as yet,we have not been able to unambiguously identify the particular radioisotope(s) since the low levels of the activity and the small

sample sizes available to us make it exceedingly difficult to assign other characteristic gamma-emissions.These observations,nevertheless, indicate that some areas in Canada were probably impacted by the very low levels of radioactivity,presumably released during the April 10 accident at the Nevada test site(The Hamilton Spectator,3 July 1986). The levels of the Nevada radionuclides appear to be too low to cause any concern;nor are they expected to interfere in our measurements on the Chernobyl radioactivity in Canada as no ruthenium radioisotope was detected in the Nevada-released radioactivity .Also,the Nevada cesium activity levels at the currently-monitored Algoma site had considerably reduced before the arrival of the Chernobyl cloud.

The release characteristics of some commonly observed Chernobyl radionuclides are shown in Table 4.The emitted radionuclides, clearly belonging to three distinct classes,are expected to show both primary (i.e. at the time of release) and secondary(i.e. during subsequent atmospheric transport) fractionation.That this fractionation has indeed occurred is evident from Fig.4 where the activity ratios of radionuclides measured in the Burlington rain samples are plotted as a function of time.The dashed line in each case is derived from the radionuclide inventory of a light water reactor core running for two years(Lewis et al.,1975;Hohnemser et al.,1986) and indicates the activity ratio obtainable in absence of fractionation.The magnitude of fractionation is usually expressed in terms of the fractionation factor, f(A), for a given radionuclide,A.The standard definition (Edvarson et al., 1959) of f(A) utilizes ^{95}Zr as a reference radionuclide.Unlike the fractionation of nuclear weapons-testing fallout where ^{95}Zr was easily

detectable,the Chernobyl fallout is characterized by the general absence of this radionuclide.Therefore,for the Chernobyl fallout studies, fractionation factor may be defined as

$$f(A,^{137}Cs) = \left[\frac{N(A)}{N(^{137}Cs)}\right]_{meas} \cdot \left[\frac{N(^{137}Cs)}{N(A)}\right]_{reactor} ,$$

where N is the activity of the radionuclide in question.The first brackets contain the measured activity values and the second the activity values as obtained from the radionuclide inventory of a light water reactor core (Lewis et al.,1975).The latter values are corrected for the decay of the radionuclide A to the time period corresponding to the measured values;the decay of ^{137}Cs is negligible for the time period under consideration.Table 5 gives the values of $f(A,^{137}Cs)$ for various radionuclides detected in Canadian rain samples.The results are qualitatively in agreement with the expected release characteristics of these radionuclides.

Our current measurements are made on rain samples collected from the Algoma site,the lead-location for the NWRI research project on acid deposition.Figure 5 depicts the results obtained for samples collected during May/June,1986.It is possible that the 3-10 June peak denotes completion of one cycle around the earth by the Chernobyl cloud with reference to this location.It remains yet to be seen if the second cycle will be detectable.On the other hand,it may be suggested that the 3-10 June peak is continuation of the phase detected in the Burlington rain beginning with the 16-20 May rain sample,as has been suggested by Bondietti and Brantley (1986).Whether the observed peak represents global circulation of the radioactive cloud or belongs

to a different emission,it appears reasonable to infer that the residence time of the Chernobyl emission is at least about six weeks if one assumes that the reactor stopped spewing significant amounts of radioactivity within at most a week of the 26 April release.In comparison,the results from the previous studies,using both weapons-testing fallout (Thein and Kuroda,1967) and naturally-occurring (Moore et al.,1973) radionuclides,yielded tropospheric aerosol residence times ranging from a few days to several weeks.

The controversy over the accuracy and interpretation of atmospheric residence time data has often surfaced in the literature (see,for example,Martell and Drevinsky,1960;Nevissi et al.,1974),and indeed is expected considering the complex transport processes involved.In this regard,the information derived from the Chernobyl radioactivity should be more realistic for two major reasons.Firstly, the characteristic Chernobyl radionuclides do not suffer from the 'natural' interferences (such as inputs from uranium mining areas and forested regions) one may encounter in using uranium-series radionuclides for deriving residence times.Secondly,since,as far as we know,the Chernobyl release was essentially confined to the troposphere, the 'stratospheric reservoir' effect (Libby,1959) would not apply. Inputs from this 'stratospheric reservoir' of weapons-testing fallout radionuclides to each subsequent event are not readily distinguished or realistically estimated for the short-lived radionuclides previously used in estimating tropospheric residence times.In this context,it may also be noted that the characteristic cesium or ruthenium radioisotopic

ratios in the Chernobyl fallout may also be quite useful in furthering our understanding of the interhemispheric transfer/exchange processes. Further measurements should lead to a better understanding of some of the processes operative in the lower atmosphere

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TABLE 1

SOME OF THE MAJOR DIFFERENCES BETWEEN THE CHERNOBYL FALLOUT AND THE WEAPONS TESTING FALLOUT

	Chernobyl	Nuclear weapons testing
Release	- relatively intermittent - temperature 2000-2800 K	- spontaneous - temperature 10^8 K
Radioactivity	- full range of radionuclides, but preferential release of volatile fission products	- full range of radionuclides dispersed
Transport	- tropospheric - residence time (?) - rapid mixing expected	- mainly stratospheric - residence time 1-10 years (stratospheric); up to few weeks (tropospheric) - slow mixing
Deposition	- very heavy 'local' fallout - significant 'tropospheric' fallout	- significant 'stratospheric fallout (long-lived radionuclides) - less pronounced 'local' and 'tropospheric' fallouts
Levels	- 8-10 kBq m^{-2} (West Germany) - $\sim 10^{-3}$ kBq m^{-2} (Burlington, Canada)	- 1963 fallout maximum: 0.8 kBq m^{-2} (West Germany) - 1963 fallout maximum: 1.3 kBq m^{-2} (40-50° N)

TABLE 2
RESULTS OF MEASUREMENTS ON THE BURLINGTON WATER FILTRATION PLANT FLOC SAMPLES

Sampling date	Level (mBq/L)				
	^{137}Cs	^{131}I	^{125}Sb	^{103}Ru	^{65}Zn
12 May 1986	ND[a]	0.11±0.04	ND	ND	ND
20 May 1986	0.15±0.04	ND	0.11±0.07	0.08±0.04	0.08±0.04
29 May 1986	0.09±0.02	ND	ND	0.08±0.04	0.11±0.04

[a]ND, not detected

TABLE 3

LEVELS OF RADIONUCLIDES DETECTED IN PRE-CHERNOBYL RAIN AND SNOW SAMPLES

Sample location	Sample type	Collection date	Level (mBq/L)							
			^{144}Ce	^{141}Ce	^{137}Cs	^{134}Cs	^{125}Sb	^{110m}Ag	^{95}Zr	^{65}Zn
Algoma	Rain	1-8 April	ND[a]	ND	ND	ND	ND	ND	ND	ND
Algoma	Rain	8-21 April	ND	345±255	44±26	ND	ND	ND	ND	ND
Algoma	Rain	29 April - 6 May	16±11	ND	5±3	5±3	ND	ND	ND	ND
Longwoods	Rain	20 April - 3 May	ND	ND	224±131	ND	618±300	278±134	978±442	479±313
Slave River	Snow	2 May	ND	ND	4±2	ND	11±5	4±2	ND	ND

[a] ND, not detected

LE 4

RELEASE CHARACTERISTICS OF SOME COMMONLY OBSERVED CHERNOBYL RADIONUCLIDES

Radionuclide	Half-life	Approximate core inventory[a] (EBq)	Release characteristics[a]
Iodines			
^{131}I	8.0 days	3.145	- b.p. 184°C;does not bind effectively to the melt - largely associates with the aerosol formed by condensed metal oxides and with other vapours
Cesiums			
^{134}Cs	2.06 years	0.063	- b.p. 670°C - not retained in the melt
^{137}Cs	30.1 years	0.215	
Volatile Oxides			
^{103}Ru	39.35 days	3.700	- b.p. 3900°C - less stable oxides - unpredictable release patterns;simulated vary from < 1% to 100%
^{106}Ru	368.2 days	0.703	
Nonvolatile Oxides			
^{141}Ce	32.4 days	5.920	- b.p. 3426°C - react with H_2O and CO_2 to form stable oxides - oxides mix with UO_2 and released in proportion to vapourized UO_2
^{144}Ce	284.2 days	4.070	

[a]Lewis et al. (1975)

TABLE 5
RADIONUCLIDE FRACTIONATION FACTORS IN CANADIAN RAINS

Location	Sample date	f(A, ^{137}Cs)					
		^{144}Ce	^{141}Ce	^{140}Ba	^{131}I	^{106}Ru	^{103}Ru
Algoma	13-20 May	ND[a]	ND	ND	D[b]	ND	0.294
Burlington	14-16 May	ND	ND	ND	3.09	ND	4.21×10^{-2}
Burlington	17-20 May	ND	4.56×10^{-3}	2.68×10^{-2}	4.73	6.47×10^{-2}	8.22×10^{-2}
Burlington	20-24 May	2.06×10^{-3}	ND	ND	10.37	0.124	9.67×10^{-2}
Algoma	27 May-3 June	ND	1.34×10^{-3}	ND	D	0.264	0.224
Algoma	3-10 June	6.76×10^{-3}	5.28×10^{-3}	ND	D	0.377	0.236
Algoma	10-17 June	ND	ND	ND	D	1.04	0.218

[a] ND, not detected ; [b] D, decayed prior to analysis

FIGURE CAPTIONS

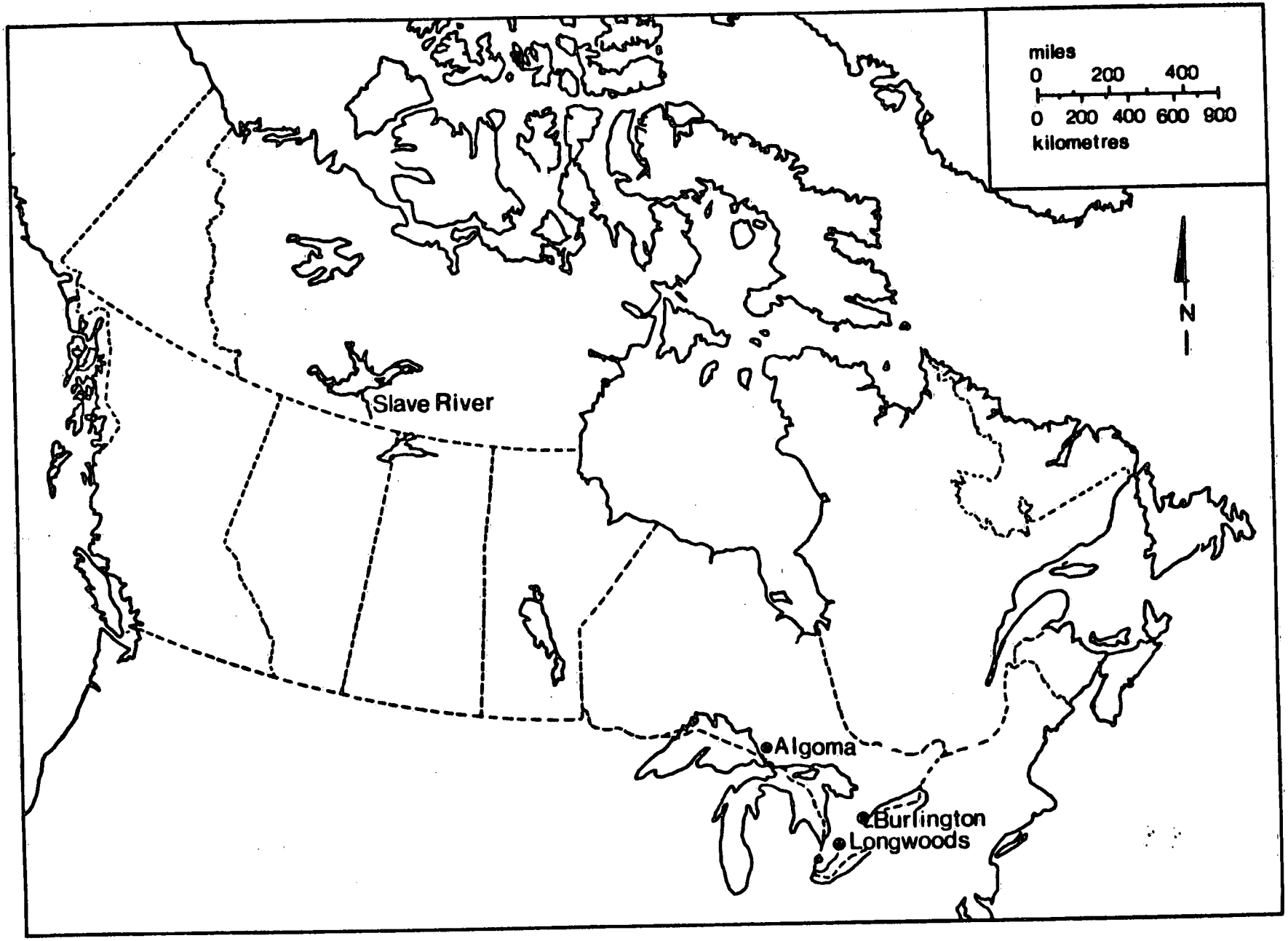
Fig.1 Locations for rain and snow samples.

Fig.2 Measured levels of radionuclides in the Burlington rains (May 6-24,1986).

Fig.3 Deposition of Chernobyl-delivered ^{137}Cs in some West European locations.

Fig.4 Activity ratio profiles of major radionuclides detected in the Burlington rains.

Fig.5 Variations in the concentrations of cesiums and rutheniums in the May/June 1986 Algoma rain samples.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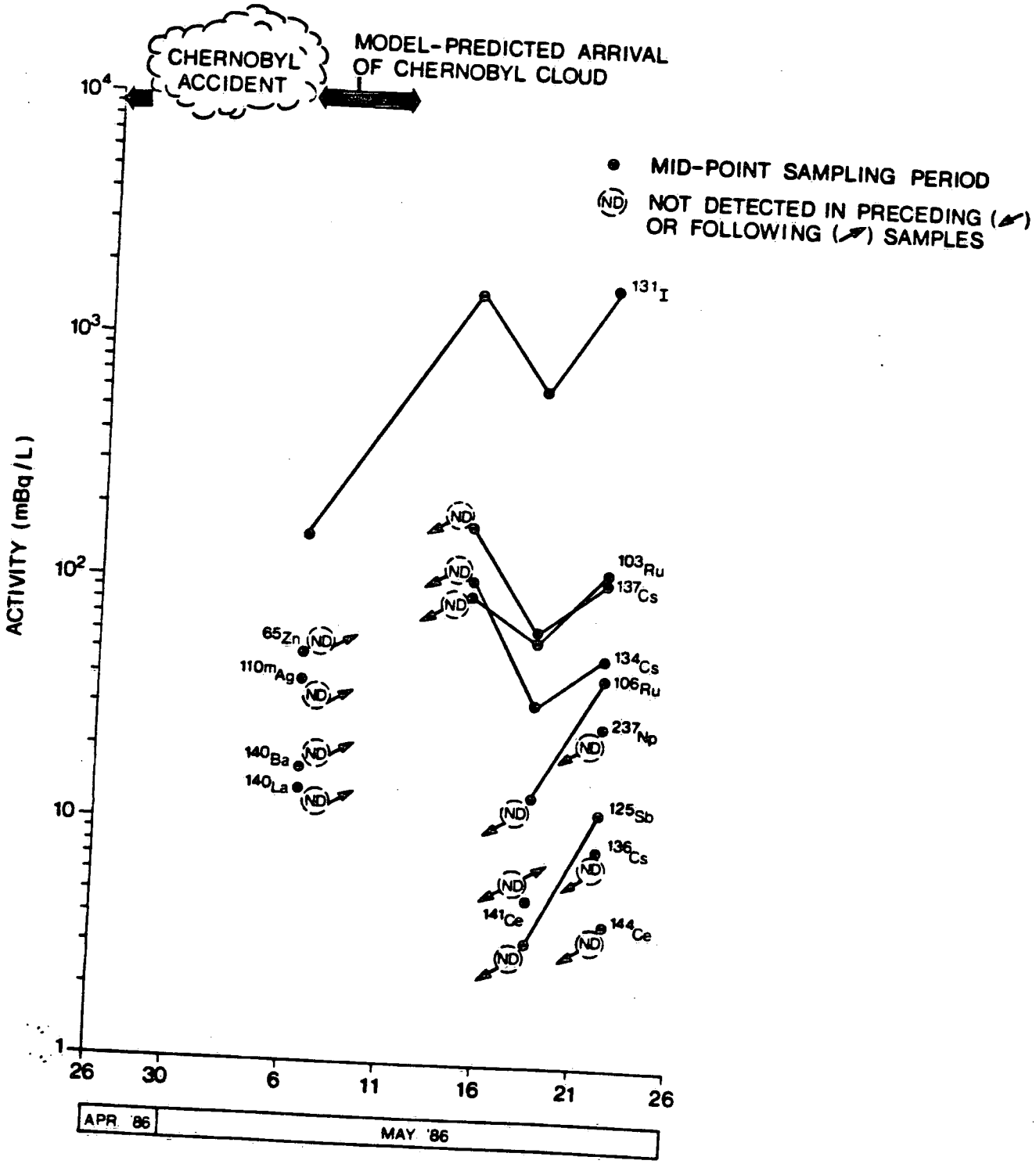
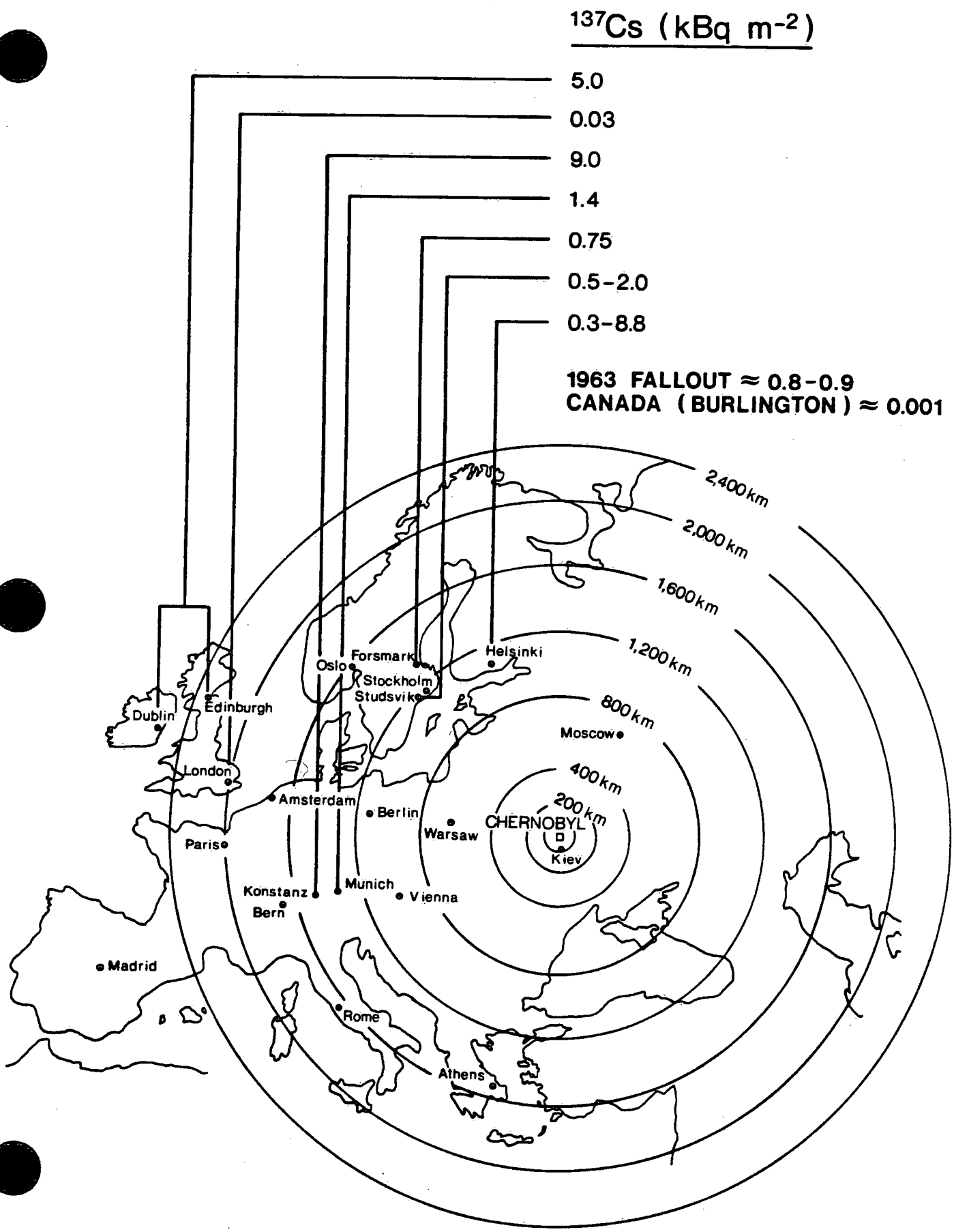
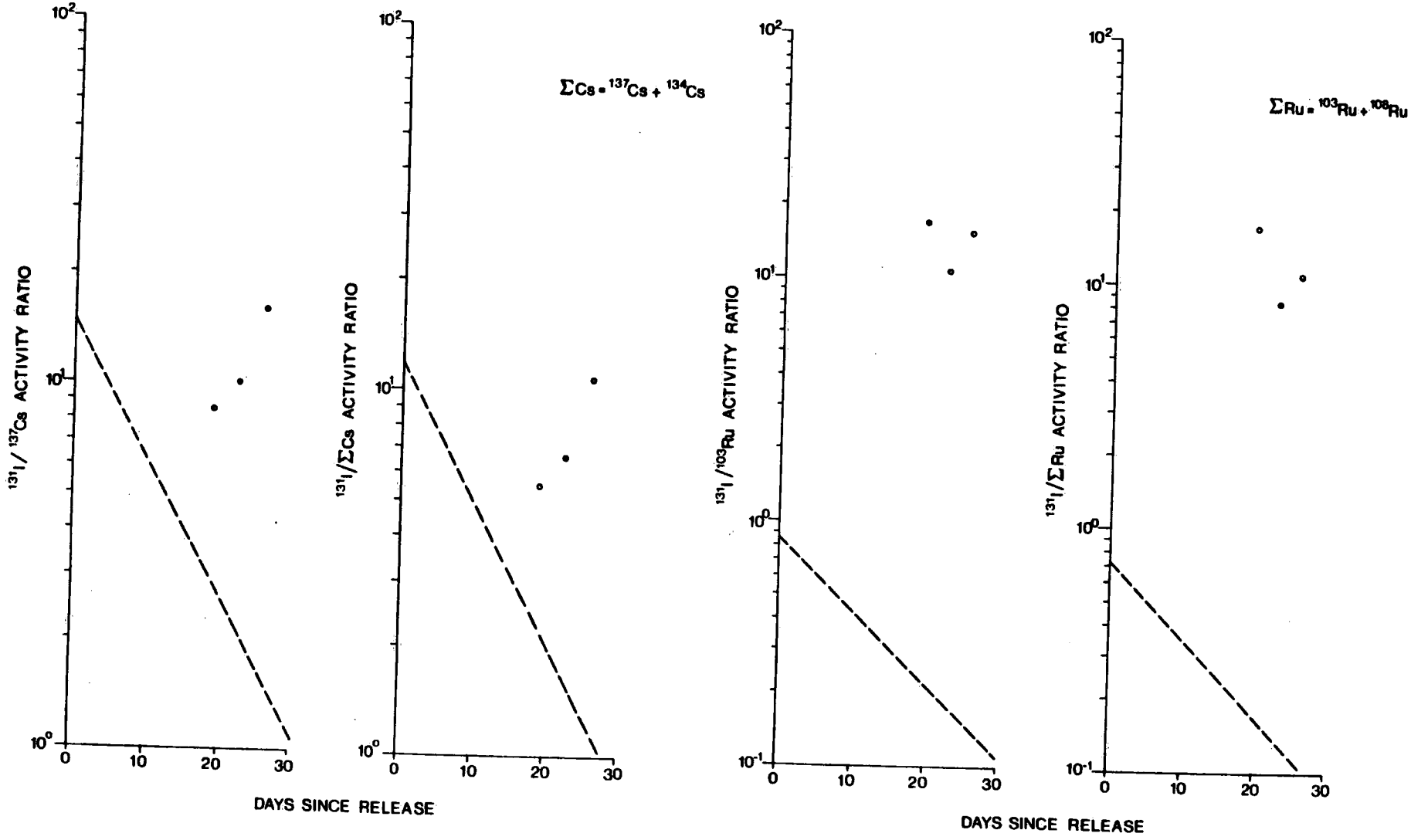
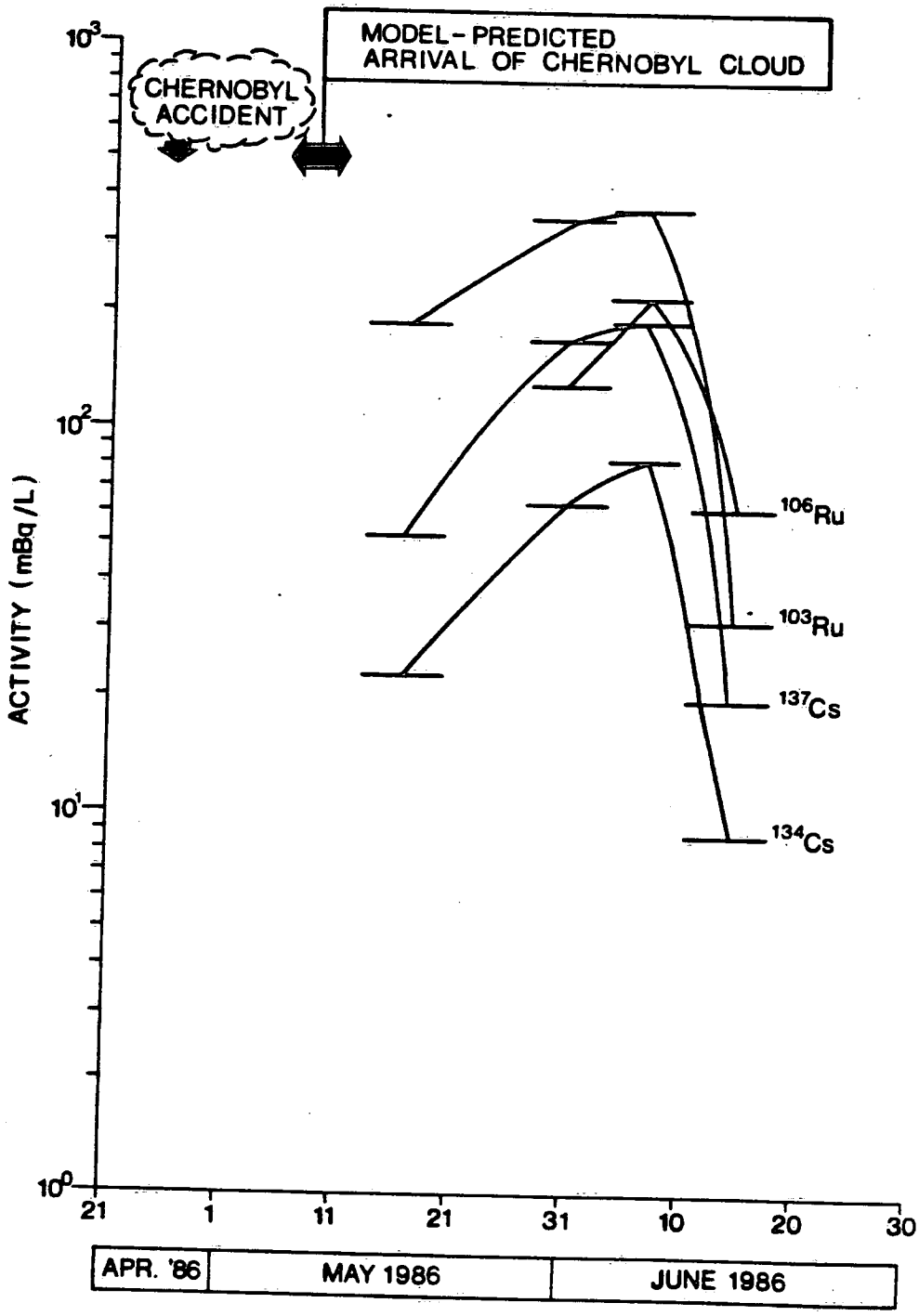
0 200 400
0 200 400 600 800
kilometres
N
Slave River
Algoma
Burlington
Longwoods


CHERNOBYL ACCIDENT
MODEL-PREDICTED ARRIVAL OF CHERNOBYL CLOUD
MID-POINT SAMPLING PERIOD
(ND) NOT DETECTED IN PRECEDING (↙) OR FOLLOWING (↗) SAMPLES
ACTIVITY (mBq/L)
10^4
10^3
10^2
10
1
^{131}I
^{103}Ru
^{137}Cs
^{134}Cs
^{106}Ru
^{237}Np
^{125}Sb
^{136}Cs
^{141}Ce
^{144}Ce
^{65}Zn
^{110m}Ag
^{140}Ba
^{140}La
(ND)
26
30
6
11
16
21
26
APR '86
MAY '86


^{137}Cs (kBq m^{-2})
5.0
0.03
9.0
1.4
0.75
0.5-2.0
0.3-8.8
1963 FALLOUT ≈ 0.8-0.9
CANADA (BURLINGTON) ≈ 0.001
2,400 km
2,000 km
1,600 km
1,200 km
800 km
400 km
200 km
Dublin
Edinburgh
London
Amsterdam
Paris
Oslo
Forsmark
Stockholm
Studsvik
Helsinki
Moscow
Berlin
Warsaw
CHERNOBYL
Kiev
Konstanz
Bern
Munich
Vienna
Madrid
Rome
Athens


$^{131}I/^{137}Cs$ ACTIVITY RATIO
$^{131}I/\Sigma Cs$ ACTIVITY RATIO
$\Sigma Cs = ^{137}Cs + ^{134}Cs$
$^{131}I/^{103}Ru$ ACTIVITY RATIO
$^{131}I/\Sigma Ru$ ACTIVITY RATIO
$\Sigma Ru = ^{103}Ru + ^{106}Ru$
DAYS SINCE RELEASE
DAYS SINCE RELEASE


MODEL-PREDICTED
ARRIVAL OF CHERNOBYL CLOUD
CHERNOBYL
ACCIDENT
ACTIVITY (mBq/L)
10^3
10^2
10^1
10^0
^{106}Ru
^{103}Ru
^{137}Cs
^{134}Cs
21
1
11
21
31
10
20
30
APR. '86
MAY 1986
JUNE 1986