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**EARLY OBSERVATIONS ON  
CHERNOBYL FALLOUT IN CANADA**

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### MANAGEMENT PERSPECTIVE

Following the Chernobyl nuclear reactor accident in April 1986, the National Water Research Institute initiated measurements on samples obtained as part of our Acid and Toxic Rain projects designed to study the impact of the accident on Canadian freshwater systems.

The results obtained so far show that the radionuclide deposition in Canada was several thousand times lower in Canada than in Western Europe. Specific measurements failed to detect any Chernobyl radionuclides in Lake Ontario, even when highly sensitive NWRI techniques were used. Presumably, the dilution effect pushed the levels below our detection capabilities. It is unlikely that any Chernobyl-derived radionuclides reached populations deriving drinking water from water filtration plants. Also, unlike in Europe, the Chernobyl radioactivity will not leave a "marker" in the Canadian aquatic environment.

During the course of the above investigations, we have fingerprinted the specific isotopes released and compared our data with available European measurements and estimated releases from a reactor core. The results indicate that Chernobyl radioactivity may be used to delineate lower atmospheric circulation patterns. Such measurements provide useful information with regard to the long-range transport of pollutants since the Chernobyl release closely parallels a large stack emission. The limited data collected so far also indicates the presence of some pre-Chernobyl radionuclides in very small amounts. It may be argued that these radionuclides were released on April 10 from the Nevada test site when an underground nuclear explosion resulted in accidental releases of radioactivity to the atmosphere.

## OBSERVATIONS PRÉLIMINAIRES SUR LES RETOMBÉES DE CHERNOBYL AU CANADA

### PERSPECTIVE-GESTION

Dans le but de comprendre les répercussions de l'accident survenu à la centrale nucléaire de Chernobyl en avril 1986 sur les systèmes d'eau douce au Canada, l'Institut national de recherche sur les eaux a entrepris d'analyser des échantillons obtenus dans le cadre du projet sur les pluies acides et les pluies toxiques.

Jusqu'à maintenant, les résultats obtenus démontrent que les dépôts de radioéléments sont plusieurs milliers de fois plus faibles au Canada qu'en Europe de l'Ouest. Des mesures spécifiques n'ont permis de détecter aucun des radioéléments provenant de Chernobyl dans le lac Ontario, même en appliquant les techniques hautement sensibles de l'INRE. Il est probable qu'à cause de la dilution, les niveaux de radioélément ont baissé en deça de notre capacité de détection. Les radioéléments de Chernobyl n'ont probablement pas contaminé l'eau potable de consommation provenant des usines de filtration d'eau. D'autre part, contrairement à ce qui s'est produit en Europe, la radioactivité de Chernobyl ne laissera pas de "marqueur" dans l'environnement aquatique au Canada.

Au cours des enquêtes mentionnées plus haut, nous avons identifié les isotopes spécifiques libérés et avons comparé nos données avec les mesures prises en Europe et les émissions provenant probablement de la partie centrale du réacteur. Les résultats indiquent qu'on peut utiliser les émissions de radioéléments de Chernobyl pour déterminer les tendances des courants aériens dans la basse atmosphère. Ces mesures sont utiles dans le cadre des études sur le transport à grande distance des polluants, étant donné que l'accident de Chernobyl ressemble d'assez près aux émissions des grosses cheminées d'usine. Les quelques données rassemblées jusqu'à maintenant révèlent également que certains radioéléments en très petites quantités étaient présents avant Chernobyl. On pourra supposer que ces radioéléments ont été libérés le 10 avril dernier du site expérimental au Nevada, lorsqu'une explosion nucléaire souterraine a provoqué la libération accidentelle de radioactivité dans l'atmosphère.

## Early Observations on Chernobyl Fallout in Canada

SIR- Fission products deriving from the Chernobyl reactor accident were first detected in a Burlington rain sample on 6-7 May, 1986, in agreement with the air transport model-based prediction by Environment Canada's Atmospheric Environment Service. Two distinct maxima were indicated during subsequent measurements. The first maximum (14-16 May) was characterized by  $^{137}\text{Cs}/^{134}\text{Cs}$  and  $^{137}\text{Cs}/^{103}\text{Ru}$  activity ratios of about 1.7 and 1.9, respectively;  $^{131}\text{I}$  was the only other radionuclide detected. The second maximum, discerned in the 20-24 May rain, was characterized by  $^{137}\text{Cs}/^{134}\text{Cs}$  and  $^{137}\text{Cs}/^{103}\text{Ru}$  activity ratios of about 2.1 and 0.9, respectively. These activity ratios are similar to those obtained in Sweden<sup>1</sup>, UK<sup>2</sup>, West Germany<sup>3</sup> and USA<sup>4</sup>. In addition,  $^{106}\text{Ru}$ ,  $^{125}\text{Sb}$ ,  $^{136}\text{Cs}$ ,  $^{144}\text{Ce}$  and  $^{237}\text{Np}$  were also unambiguously identified, in the 20-24 May rain, using low-background hyperpure germanium coaxial and planar detectors. The presence of  $^{237}\text{Np}$  suggests that this phase of airborne radioactivity was derived from the 'northern' cloud<sup>3</sup>.

An intriguing feature of the measurements, summarized in Table 1, was the easy detection of very small amounts of activation products  $^{65}\text{Zn}$  and  $^{110\text{m}}\text{Ag}$  in the 6-7 May rain. While Chernobyl-delivered  $^{110\text{m}}\text{Ag}$  has been detected in UK<sup>5</sup> and in Canada<sup>6</sup>, the presence of  $^{65}\text{Zn}$  in Chernobyl fallout has not been reported. In order to trace the source of this  $^{65}\text{Zn}$ , one snow sample and three rainwater samples from widely different locations and corresponding to pre-Chernobyl time periods (predicted or actual) were also assayed.

The results of these measurements, also given in Table 1, indicate the presence of several fission and activation products in these samples. That their origin is different from Chernobyl is suggested by the detection of  $^{95}\text{Zr}$ . Our continuing analyses have consistently shown the absence of  $^{95}\text{Zr}$  in post-Chernobyl samples. It may be argued that the pre-Chernobyl radionuclides in Canada are derived from the reported<sup>7</sup> accidental release of radioactivity during the April 10 underground testing of a nuclear weapon at the Nevada test site. Although our measurements seem to support this scenario, a definite conclusion must await completion of analyses on several other samples we have recently obtained.

Measurements on Lake Ontario water samples using the highly-sensitive floc technique<sup>8</sup> did not reveal above-background levels of any of the radionuclides of concern. It is thus reasonable to infer that negligible, if any, radioactivity reached populations deriving water from municipal water supplies located around large freshwater bodies in Canada. This also implies that the Chernobyl radioactivity is unlikely to leave any 'marker' in the Canadian aquatic environment. In contrast, in Europe, where localized<sup>1-3</sup> Chernobyl fallout was heavier than that from extensive weapons testing<sup>9</sup> in the late 1950's and early 1960's and over four orders of magnitude higher than that in Canada, a long-lived radioisotope such as  $^{137}\text{Cs}$  should serve as a distinct dating horizon for the next 150 years or more.

The Chernobyl fallout differs from the weapons-testing fallout in that it involved preferential release of volatile elements

and oxides. Furthermore, the bulk of weapons-testing debris was injected into the stratosphere from where it came down with the residence time of at least one year. The Chernobyl fallout, on the other hand, was essentially confined to the troposphere and would probably come down in relatively short period of time. In this respect, the Chernobyl cloud may be likened to the plume of pollutants transmitted by a given industry. Unlike the releases of industrial pollutants to the atmosphere, the Chernobyl emission is characterized by the presence of unique radionuclides which may be used to study the tropospheric transport patterns. It appears that ruthenium and/or cesium radioisotopes would best serve this purpose. The results of our measurements on  $^{103}\text{Ru}$ ,  $^{106}\text{Ru}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in Burlington and Algoma rains are plotted as a function of time in Fig. 1. The dashed line in each case is derived from the radionuclide inventory of a light water reactor core running for two years<sup>3,10</sup>. The activity ratio  $^{103}\text{Ru}/^{106}\text{Ru}$  is similar to that observed in Sweden in pure ruthenium particles<sup>1</sup>. The activity ratios  $^{103}\text{Ru}/^{137}\text{Cs}$  and  $\Sigma\text{Ru}/\Sigma\text{Cs}$  are clearly influenced by the complex fractionation and atmospheric mixing patterns. The initial departure from the predicted  $\Sigma\text{Ru}/\Sigma\text{Cs}$  activity ratio seems to wane with the passage of time. This probably reflects removal of larger hot particles by dry or wet deposition. The ensuing preponderance of smaller particles not only improves atmospheric mixing patterns but also results in ground measurements which are more representative of sample population.

The ease with which rutheniums are detectable is significant in two respects. Firstly, very little is known about the behaviour of ruthenium-bearing particles in the environment. Secondly,

the laboratory experiments designed to study the release of ruthenium from reactor fuels have failed to adequately explain the observed patterns<sup>10</sup>. In this context, the Chernobyl-delivered ruthenium provides a unique opportunity to delineate its release and atmospheric dispersion characteristics.

Our current measurements are made on rain samples collected from the Algoma site, the prime monitoring location for NWRI project on acid deposition. The results for samples collected from 13 May to 17 June are shown in Fig. 2. As yet we do not have enough data to ascertain whether the 3-10 June peak is part of the second maximum noted earlier or denotes completion of one cycle around the earth<sup>11</sup> with reference to this location. Further measurements should provide a clearer picture of the circulation pattern and residence time of the Chernobyl cloud.

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Table 1 Levels (mBq/L) of radionuclides in precipitation

Location	Sample	Date	131I	137Cs	134Cs	136Cs	Radionuclide 103Ru 106Ru	141Ce	144Ce	140Ba	125Sb	95Zr	65Zn	110mAg	237Np
Burlington, Ont.	Rain	6-7 May	158±118	ND	ND	ND	ND	ND	ND	ND	ND	ND	50±38	40±20	ND
Burlington, Ont.	Rain	14-16 May	1583±92	181±8	109±7	ND	94±8	ND	ND	ND	ND	ND	ND	ND	ND
Burlington, Ont.	Rain	17-20 May	631±63	64±17	32±1	ND	61±2	5±3	ND	14±6	ND	ND	ND	ND	ND
Burlington, Ont.	Rain	20-24 May	1743±123	109±4	51±3	9±4	115±3	42±20	4±2	ND	11±6	ND	ND	ND	27±11
Algoma, Ont.	Rain	1-8 April	NM	ND	ND	NM	ND	ND	ND	ND	ND	ND	ND	ND	ND
Algoma, Ont.	Rain	29 April- 6 May	NM	5±3	5±3	ND	ND	ND	16±11	ND	ND	ND	ND	ND	ND
Longwoods, Ont.	Rain	20 April- 3 May	NM	224±131	ND	ND	ND	ND	ND	ND	618±300	978±442	479±313	278±134	ND
Slave River, NWT	Snow	2 May	NM	4±2	ND	ND	ND	ND	ND	ND	11±5	ND	ND	4±2	ND

Count time, 2.5-3.5x10<sup>5</sup> s; Levels decay-corrected to mid-point sampling period; ND, not detected; NM, not measured

### FIGURE CAPTIONS

**Fig.1** Activity ratio profiles of cesiums and rutheniums in Burlington and Algoma rains.

**Fig.2** Variations in the concentrations of cesiums and rutheniums in Algoma rain samples collected following the Chernobyl accident.



