Reports

Airborne Short-Lived Radionuclides of Unknown Origin in Sweden in 1976

Abstract. *On five occasions during the first half of 1976 traces of neptunium-239 and molybdenum-99 were detected in southern Sweden. These activities were accompanied by small amounts of fission products such as iodine-131 and barium-140. One of the events was coincident in time with large peaks in the atmospheric tritium gas concentration.*

In light of the current discussion of the possibility that the Soviet Union is developing a charged-particle beam weapon (*1*), some observations made in Sweden in the spring of 1976 should be of interest.

Since the middle 1950's the Swedish National Defense Research Institute has operated a network of nuclear fallout collection stations in Sweden. At these stations air is blown at a rate of 25,000 to 50,000 m^3/day through glass fiber filters, which are sent every second or third day to the main laboratories in Stockholm for analysis. The analysis is done mainly by γ-ray spectroscopy, utilizing large, high-resolution germanium detectors. The network consists of eight stations (Fig. 1), and the filters from each of them are checked regularly for fresh airborne radioactivity with a time resolution of 1 week. When needed, that resolution can be increased threefold. Quantitative determinations are made on a weekly basis for the Kiruna, Grindsjön, and Ljungbyhed stations and on a monthly basis for the other five. All but the Hagfors and Grindsjön ground-level air stations are combined with precipitation sampling units, and the surveillance system also includes equipment for sampling at high altitudes by use of jet planes from the Royal Swedish Air Force. The system is operated to give the necessary basic data for estimating the exposure of the Swedish population to fallout from nuclear tests, but also to detect violations of existing treaties concerning nuclear weapons tests. In a few instances, radioactive species originating in venting underground nuclear explosions have been detected in Sweden, and the 18 atmospheric nuclear explosion tests carried out by the People's Republic of China since 1964 have been studied. Radioactive species produced locally, mostly from hospitals, have sometimes been detected at single stations. However, on five occasions in 1976, in late February, March, April, May, and July, unusual mixtures of short-lived radionuclides were present at several stations in southern Sweden.

Most of the activity was due to the two nuclides ^{239}Np and ^{99}Mo (half-lives, 2.35 and 2.75 days, respectively), but some ^{131}I and ^{140}Ba (half-lives, 8.05 and 12.8 days, respectively) were also detected. China carried out an atmospheric nuclear test in the low-yield range on 23 January 1976 which resulted in small amounts of globally distributed debris. This explosion could easily account for the ^{131}I and ^{140}Ba activities in February and March, but could not explain the presence of the very short-lived ^{239}Np and ^{99}Mo activities from late February on or the ^{131}I and ^{140}Ba from April on.

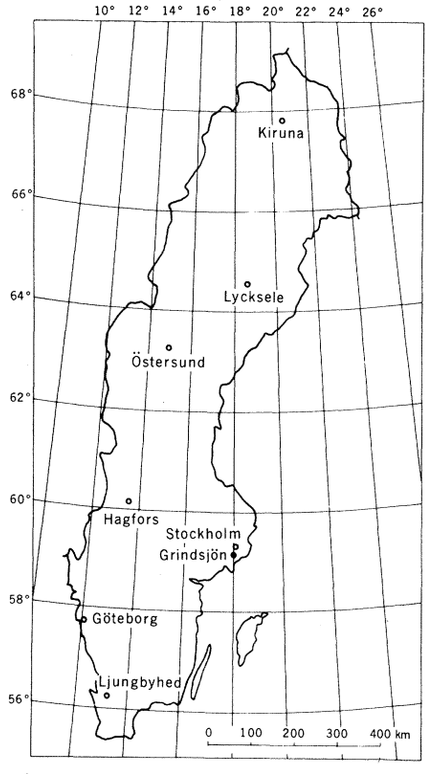

Fig. 1. Swedish stations for sampling radioactivity in ground-level air and precipitation, operated by the National Defense Research Institute.

The short-lived nuclides (half-life $\leq$ 12.8 days) that appeared in Sweden after the Chinese test and on the five subsequent occasions are listed in Table 1 for the sampling stations at Grindsjön and Ljungbyhed. The values given include the statistical, detector efficiency, and γ/β-branching errors. The limits of detection depend on the relative amount of background, the amount of air sampled, the time delay between sampling and counting, and the counting time. In the measurements accounted for here the detection limits vary, for ^{239}Np between 50 and 400, for ^{99}Mo between 5 and 40, for ^{131}I between 3 and 10, and for ^{140}Ba between 2 and 5 picocurie-seconds per kilogram of air.

The activities detected in February, March, and April were also present at the Stockholm, Hagfors, and Gothenburg (Göteborg) stations, while those in May and July were detected only at the Grindsjön station. No strange increases in activity were detected north of Hagfors, and the concentrations were larger at the two Stockholm stations and at Ljungbyhed than at Hagfors and Gothenburg. A study of regional wind patterns during the period of interest showed that all five events occurred during weeks when northeasterly or easterly winds prevailed for at least 2 days, indicating that the material arrived in Sweden by way of southern Finland or eastern Soviet Union and the Baltic Sea. This is supported by observations of ^{239}Np and ^{99}Mo in Finland on two of the occasions (*2*). Activities of about 5700 and 1300 pc-sec/kg, respectively, were detected at Helsinki during the week of 8 to 15 March (occasion 2), and activities a few times higher were detected from 6 to 19 April (occasion 3). At the time of the first event the sampling unit was out of operation. A more detailed study of the samples from the Stockholm station on the first two occasions revealed that the nuclides arrived between the afternoons of 20 and 23 February and of 12 and 15 March. During both periods the weather was characterized by easterly winds.

The Hagfors station is equipped with a unit for collecting gaseous tritium (HT + T_2). Twice a week samples are sent to Stockholm for processing and analysis. These measurements were started in November 1975 and have been carried out since then (*3*). In November and December 1975 a peak in the HT + T_2 concentration was detected that was interpreted as due to an underground nuclear explosion at Novaya Zemlya in October.

Table 1. Short-lived radionuclides at Grindsjön and Ljungbyhed during the first half of 1976 expressed in picocurie-seconds per kilogram of air. Dividing the numbers by 86.4 and the length of the sampling period in days yields the average concentration in femtocuries per kilogram of air.

Sampling period	Neptunium-239		Molybdenum-99		Iodine-131		Barium-140		Notes
	Grindsjön	Ljungbyhed	Grindsjön	Ljungbyhed	Grindsjön	Ljungbyhed	Grindsjön	Ljungbyhed	
2–9 Feb.	535 ± 170	730 ± 290	159 ± 40	80 ± 21	270 ± 19	350 ± 27	712 ± 51	1045 ± 80	Chinese debris
9–16 Feb.					177 ± 14	67 ± 9	691 ± 68	137 ± 16	
16–23 Feb.	490 ± 150	690 ± 240	134 ± 16	143 ± 20	38 ± 5	44 ± 5	80 ± 8	92 ± 9	Occasion 1; the I and Ba
23 Feb.–1 Mar.	320 ± 110		34 ± 9		20 ± 5	29 ± 8	61 ± 8	47 ± 8	mainly of Chinese origin
1–8 Mar.							20 ± 4		
8–15 Mar.	340 ± 100		88 ± 11		33 ± 4		39 ± 4		Occasion 2; the I and Ba
15–22 Mar.	260 ± 90	860 ± 340	82 ± 12	222 ± 26	41 ± 7	94 ± 11	48 ± 7	82 ± 7	mainly of Chinese origin
22–29 Mar.					12 ± 4				
29 Mar.–5 Apr.			25 ± 7						
5–12 Apr.									
12–20 Apr.	2030 ± 270		1053 ± 76	45 ± 13	173 ± 13		223 ± 18	21 ± 7	Occasion 3
20–26 Apr.			36 ± 7	43 ± 14	32 ± 4	78 ± 19	10 ± 3		
26 Apr.–3 May									
3–10 May									
10–17 May					68 ± 8				
17–24 May	320 ± 110		118 ± 15		17 ± 4		16 ± 3		Occasion 4
24–31 May			25 ± 9						
⋮									
12–19 July			14 ± 5				11 ± 4		Occasion 5

After that the tritium level was quite constant through 1976 at about 60 atoms per milligram of air, except for two large peaks in March, when about 200 atom/mg were detected between 12 and 15 March and a maximum of 580 atom/mg occurred between 19 and 23 March. These events coincide very well in time with the second occasion on which short-lived radionuclides were observed, even if they could not be correlated with very high levels of ^{239}Np or ^{99}Mo at Hagfors at the same time.

The ^{239}Np/^{99}Mo activity ratio in most measurements varies between 2 and 4, which is compatible with ratios in bomb-produced debris during the first few weeks (the ratio decays with a half-life of 16 days). Irradiation of natural uranium by thermal neutrons results in a ^{239}Np/^{99}Mo activity ratio that is larger than 10 at the time of irradiation and takes about 1 month to decrease to 3. If natural molybdenum is present in addition to the uranium the ratio can vary freely below 10, as ^{99}Mo is produced by neutron capture in the naturally abundant isotope ^{98}Mo. Also, if the uranium is enriched in ^{235}U lower ratios will result. Ratios of 2 to 4 are consistent with ^{235}U concentrations between 4.4 and 2.2 percent, which covers enrichments used in light-water reactor fuel.

If the ^{99}Mo found originated in a fission process, which is suggested by the presence of ^{131}I and ^{140}Ba on the third and fourth occasions, as much as 12 and 17 weeks after the weak Chinese explosion, it is strange that other short-lived fission products such as ^{132}Te (half-life, 3.25 days) were not detected along with the ^{239}Np and ^{99}Mo. Mechanisms of transport from the source to the atmosphere and to the samplers can alter nuclide relations considerably, but the measurements show that 99 percent or more of the ^{132}Te that would be expected from the ^{99}Mo activity was missing (*4*). A natural conclusion is that the ^{99}Mo was not directly produced in a fission process. It could, of course, have been produced in some laboratory work on fresh fission products, but it could also have been produced by neutron capture in natural molybdenum used as construction material in a nuclear fission or fusion device.

The only sources within Scandinavia that could account for the observed activities are the nuclear power plants at Oskarshamn, Barsebäck, and Ringhals in southern Sweden and the research reactors at Studsvik (about 70 km southwest of Stockholm), at Risö in Denmark, and at Helsinki in Finland. However, none of these reactors and research laboratories reported any airborne effluents during 1976 that could be correlated with the ^{239}Np and ^{99}Mo detected.

The highest concentrations measured at Grindsjön would be compatible with an annual discharge of the order of 10 curies from the closest nuclear power plant, the one at Oskarshamn, 200 km to the southwest. However, during 1976 less than 4 mc of airborne ^{239}Np was released from that station and no discharge of ^{99}Mo could be detected (< 0.1 mc/week). Airborne particulate effluents from nuclear power plants are usually dominated by ^{140}Ba, ^{89}Sr, ^{131}I, ^{58}Co, ^{60}Co, ^{134}Cs, and ^{137}Cs, of which all but ^{89}Sr are γ-ray emitters. As most of these were not detected, and no one was predominant, an ordinary nuclear power plant does not appear to be a very probable source of the activities observed.

No atmospheric nuclear explosions were reported during the period of interest, and the underground nuclear tests that were performed cannot be correlated with our findings. Furthermore, it would be very unlikely for ^{239}Np and ^{99}Mo to be the predominant nuclides released by such a test.

As the observed events cannot be readily accounted for in terms of any known source, the observations and speculations concerning charged-particle beam experiments at Semipalatinsk in the Soviet Union raise an interesting possibility. A recent article (*1*) discusses the possibility that a nuclear explosive generator is being developed as the power source to drive the accelerator producing the charged-particle beam. Steel segments used for the construction of two large spheres that could be part of such a generator have been observed at Semipalatinsk from reconnaissance satellites, along with large releases of hydrogen and explosive burn of the hydrogen at some altitude. Tests have been observed on seven occasions since November 1975 that may be related to the development of such a device at Semipalatinsk.

Meteorological trajectories at 850 mbar, corresponding to a height of about 1500 m, were estimated for the five occasions. According to these, nuclides from the Semipalatinsk area could have been transported to southern Sweden by easterly winds on the occasions in March, April, and May, but probably not in late February and July. Whether the short-lived radioactive species detected in

Sweden and Finland in 1976 emanated from experiments at Semipalatinsk is problematic. The large distance (about 4000 km) and the failure to find trajectories for two of the five occasions argue against such an explanation, but the lack of a known source and the concentration of the observations in the first half of 1976, when the Semipalatinsk experiments were reportedly started (*1*), suggest that it should be considered.

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References and Notes

1. C. A. Robinson, Jr., *Aviat. Week Space Technol.* **106**, 16 (2 May 1977).
2. O. Castrén, Finnish Institute of Radiation Protection, private communication.
3. B. Bernström, *Report C40062-T2* (National Defense Research Institute, Stockholm, Sweden, 1977).
4. This is based on the facts that a ^{99}Mo activity of 1053 ± 76 pc-sec/kg was detected at Grindsjön between 12 and 20 April, that the detection limit for ^{132}Te in the same measurement was 5 pc-sec/kg, and that the ^{132}Te/^{99}Mo activity ratio at formation is larger than 0.56 for all common types of fission of ^{235}U and ^{238}U (the ratio then grows with a doubling time of 17.9 days). Much of the work concerning measurements and analysis of the samples has been done by R. Arntsing and I. Vintersved at this institute.

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Primordial Noble Gases in Chondrites: The Abundance Pattern Was Established in the Solar Nebula

Abstract. *Ordinary chondrites, like carbonaceous chondrites, contain primordial noble gases mainly in a minor phase comprising ≤0.05 percent of the meteorite, probably an iron-chromium sulfide. The neon-20/argon-36 ratios decrease with increasing argon-36 concentration, as expected if the gas pattern was established by condensation from the solar nebula, and was negligibly altered by metamorphism in the meteorite parent bodies. Meteoritic and planetary matter apparently condensed over a substantial range of temperatures.*

Chondrites (*1, 2*), Earth (*1*), and Mars (*3*) contain primordial noble gases in similar proportions, and hence presumably of similar origin. They seem to have been trapped in dust grains growing from the solar nebula (*1, 4*). Efforts to localize these gases in primitive, carbonaceous chondrites such as Allende (*5, 6*) have shown that they occur mainly in "phase Q," an ill-defined Fe,Cr-sulfide comprising 0.04 percent of the meteorite, which is insoluble in HCl-HF but soluble in HNO_3 (*5-8*). Smaller amounts of gas, of distinctive isotopic composition, are found in chromite and amorphous carbon, comprising some 0.4 percent of the meteorite (*5, 6*).

To test whether these results were applicable to less primitive chondrites, we have examined five LL chondrites by the same technique (*9*). They ranged from petrologic type LL3 to LL6, representing progressively greater metamorphism and equilibration (*10*) in the meteorite parent bodies, at temperatures ranging from ~400°C for LL3 (*11*) to 950° ± 100°C for LL6 (*12*). It has been known for some time that the content of Ar, Kr, and Xe correlates with metamorphism, decreasing by some two orders of magnitude from type 3 to type 6 (*2*). But there has been some controversy over the cause of this correlation. Dodd (*13*) and others (*11, 14*) argue that "all ordinary chondrites were derived from material similar to the type 3 chondrites" and lost progressively greater amounts of volatiles during "metamorphism in an open system" (*13*). Larimer and Anders (*4*) suggest instead that the differences are intrinsic and reflect cooling of the nebula: meteorites from the deepest, most metamorphosed parts of the parent body accreted first, when nebular temperatures were highest, and therefore trapped the smallest amounts of gas.

Our study has succeeded in shedding some light on this problem. All five meteorites examined contain most of their primordial gas in minor, gas-rich phases similar to those of carbonaceous chondrites. And the proportions of Ne and Ar strongly suggest that the noble gas pattern was established during condensation, not metamorphism.

Upon treatment with HCl-HF (*5*), all meteorites gave residues of 0.3 to 0.6 percent, consisting mainly of chromite (Table 1). The amount and Cr content fell gradually through the sequence. Krymka, the least equilibrated member of the group (*15*), had a markedly lower Cr content, and hence must have contained substantial amounts of other phases. In this respect it resembles Allende, where the extra phase has been identified as amorphous carbon (*5, 6*). Two etches with red fuming HNO_3 (overnight at 50° to 70°C) dissolved only a minor part of each sample, as shown by the weight losses and the Cr and Fe contents of the etch solutions. The Fe/Cr ratio of the dissolved fraction (1.5 to 13) is much higher than that of the chromite [~0.5 (*16*)], which suggests that some reactive, Fe-rich phase is being dissolved before the less reactive chromite. Chemically, this phase thus resembles phase Q of Allende in both HNO_3 solubility and high Fe/Cr ratio. But is it also the main host phase of the noble gases?

The answer is yes. Figure 1 shows that noble gases (*17*) are greatly enriched in the unetched residues (heavy solid lines) over the bulk meteorites (dashed lines), but are largely removed on etching with HNO_3 (open circles). Evidently the noble gases are located mainly in a minor phase that fits the operational definition of Q [insolubility in HCl-HF and solubility in HNO_3 (*5*)] and that also has other properties in common with Allende Q, namely Fe/Cr > 1 and total mass fraction ≤0.05 percent of the meteorite.

The enrichment factor, residue/bulk, shows some correlation with petrologic type. For the three heavy gases (Ar, Kr, and Xe), it rises from ~4 for St. Séverin to ~150 for Hamlet, before leveling off at ~120 for Parnallee and Krymka. The fraction of heavy gases removed by etching also rises, from ~60 percent for St. Séverin to >99 percent for Hamlet. Krymka again resembles Allende, by re-

Table 1. The HCl,HF-insoluble residues from LL-chondrites. The PMD is the percentage mean deviation of Fe content of olivine (*15*), a measure of disequilibrium. Chromite contents were calculated from the Cr analyses, using reported values for chromites from individual meteorites or group means (*16*). Samples were etched twice, to remove Q as completely as possible. The figures in the last three columns refer to weight losses in the first and second treatments; they have substantial errors for the smaller samples (Krymka and Olivenza). The Cr and Fe data in the last two columns (determined by instrumental neutron activation analysis) refer to first treatment only; the Fe values have errors of ± 20 to 50 percent.

Type	Name	PMD	Residue (%)	Cr (%)	Fe (%)	Chromite (%)	Loss on HNO_3 etching		
							Weight (%)	Cr (%)	Fe (%)
LL6	St. Séverin		0.63	36.8	22.1	98	7, 4	0.21	2.7
LL5	Olivenza		0.60	34.2	23.7	90	4, 10	0.54	1.3
LL4	Hamlet	3.5	0.54	30.9	17.6	76	11, 11	0.32	2.5
LL3	Parnallee	19	0.37	33.6	18.9	81	11, 22	2.00	3.0
LL3	Krymka	45	0.32	19.1	15.1	47	4, 12	1.22	5.2
C3V	Allende (*5*)	90	0.40	13	9	39	7	0.77	2.67