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Searching for the “smoking gun” of the miscarried 2019 Nenoksa nuclear cruise missile test: a null result

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Dedicated to Prof. Dr. Thomas M. Klapötke on the Occasion of his 60th Birthday

On August 8, 2019, an explosion of a military missile occurred at the Nenoksa (also transcribed as Nyonoksa) Missile Test Center (Russian Federation). Russian authorities confirmed a release of radioactive material in the course of this incident, which fueled rumors that it could have involved a nuclear-propelled missile of the Burevestnik/Skyfall type. In this study, our radioanalytical efforts are summarized searching for the “smoking gun” of the incident. These included the gamma-measurements of air filters from two vessels that were in some proximity to the event as well as one Greek high-volume air filter. In addition, we tested the hypothesis that radioactive ⁴²Ar

may have been used to operate a radiothermal generator. If the incident had released ⁴²Ar, it may have become detectable by measuring characteristic gamma radiation emitted from a tank containing liquefied atmospheric argon. No traces whatsoever were found that could provide clues about the release. It is possible that the presumably small amounts of radionuclides released from either a small nuclear reactor or a powerful radionuclide source dispersed quickly over Russian territory to non-detectable levels before reaching any of our assayed samples.

Introduction

Following the end of atmospheric nuclear testing and especially after the 1986 Chernobyl nuclear accident, a powerful monitoring network for anthropogenic radionuclides was established in Europe (and worldwide). Many European monitoring stations are connected through a network called “Ring of Five” (Ro5) that allows rapid exchange of data and discussions. The Ro5 is an informal network gathering institutes on a laboratory level and scientists involved in measurements of ultra-low airborne concentrations of unusual radionuclides (or more common radionuclides in unexpected concentrations). Most Ro5 information comes from routine monitoring programs in Europe, which

usually refer to a weekly sampling frequency. The Ro5 is characterized by a rapid data exchange.

Most of the Ro5’s monitoring stations employ high-volume samplers and low-level radiation detectors that allow detecting trace or ultra-trace levels of radionuclides. Today, the Ro5 network encompasses 178 radionuclide monitoring stations in 29 (mainly European) countries. The Ro5 has successfully detected large,^[1] medium^[2] and small^[3] releases of anthropogenic radionuclides, including ¹⁰⁶Ru, ¹³¹I, ¹³⁷Cs, and other fission/activation products.

Since 1996, the Comprehensive Nuclear-Test-Ban Treaty (CTBT) has been open for signature and an International Monitoring System (IMS), consisting of seismic, hydroacoustic,

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infrasound and radionuclide stations has been established by the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) for its verification.^[4] Some Ro5 members are also affiliated to national CTBTO-contact institutes and vice versa. This close relationship makes a reciprocal and rapid data exchange possible. Once detections are observed in the IMS network or communicated by national institutes via the Ro5 more quantitative information on released amounts, simulated air-concentrations and additional analyses can further be investigated. Radionuclide stations within the IMS (80 world-wide) in support to the CTBTO are providing information on a daily basis thus with a higher time resolution but with about a 5–10 fold higher detection limit, as regards to gamma spectrometry results.

On Thursday, August 8, 2019, around 06:00 UTC, an explosion occurred at the Nenoksa (russ. Ненокса) Missile Test Center (64.6479 N, 69.2237 E) near Severodvinsk, Arkhangelsk Oblast, Russian Federation. According to the IMS-analysis conducted by the CTBTO International Data Centre, four geophysical detectors (3 seismic, 1 infrasound) recorded the detonation that occurred at an offshore platform in the White Sea.^[4] This incident caused eight injuries of technical and scientific staff, seven of which were reported fatal.^{[5][6]}

Immediately after the accident, the Ministry of Defence of the Russian Federation released a statement confirming an explosion during a test of a liquid-propellant rocket engine, without specifying the exact location. Although little information is available on the nature of the test at the Russian naval test range near Nenoksa, the Rosatom State Nuclear Energy Corporation said that the test involved a nuclear-powered engine, allegedly a cruise missile of the Burevestnik type (NATO reporting name: SSC-W-9 Skyfall).^[7] The testing of this type of nuclear-propelled engine may be in direct context with Russian Federation President Vladimir V. Putin's 2018 announcement of the on-going development of nuclear-powered engines^[8] with "basically unlimited range, unpredictable trajectory and ability to bypass interception boundaries."^[9]

Although early reports indicated no release of dangerous substances,^[10] NGO reports and governmental measurements indicated a short (and innocuous) spike of radiation levels (from a $\sim 0.11 \mu\text{S h}^{-1}$ background level to $\sim 2 \mu\text{S h}^{-1}$) in Severodvinsk, about 30 km east-southeast from Nenoksa.^[11]

Rosatom reported that the test had involved a "radio-isotope propellant source,"^[11] or "isotope power source" according to different reports.^[12] Any nuclear-propelled missile requires a nuclear heat source, which would be used to heat and expand a propellant (most likely air) which would then be emitted from a jet, thus providing thrust for the missile. The heat source could be either a small nuclear reactor or a powerful radionuclide source. The energy needed for propelling the missile would hence come from nuclear fission or radioactive decay. The explosion of the missile triggered concern about the release of radionuclides (fission products or radionuclides released from the radionuclide source) into the environment. Without further specifications, the Rosatom statement could be interpreted as either of the two options.

Atmospheric transport modelling conducted by the CTBTO indicated that the plume would travel toward the southeast and remain over Russian territory for a considerable time.^[13]

According to P. Seibert,^[14] the plume from the explosion would have reached IMS station RUP61 Dubna (south of Nenoksa) on August 10, 2019; station RUP54 Kirov (south-southeast of Nenoksa) on August 11, 2019; station RUP59 Zalesovo on August 13, and later possibly also the stations at Norilsk (RUP55; under construction) and Bilibino (RUP57). However, radionuclide data from these stations were never received by the CTBTO's International Data Centre in Vienna. On August 18, 2019, the Executive Secretary of the CTBTO, Lassina Zerbo, tweeted that the CTBTO was working with station operators to solve "technical problems" that had been reported on the IMS stations Dubna and Kirov after they had stopped transmitting data previously. Later, stations Zalesovo, Peleduy (RUP56), and Bilibino (RUP57) also stopped transmission of data.^[14]

On August 20, 2019, in an interview with Interfax, Deputy Foreign Minister of the Russian Federation Sergei Alexeyevich Ryabkov, argued that "Russia's transmission of data from radiation stations to the Vienna-based CTBTO was voluntary, and in any case was not subject to the organization's consideration,"^[12] thereby indirectly admitting that the cessation of data transmission was not due to technical problems.

The Russian Federal Service for Hydrometeorology and Environmental Monitoring (Rosgidromet) reported on its website that, from August 8 to 23, 2019, it found short-lived fission products ^{91}Sr , ^{139}Ba , ^{140}Ba , and ^{140}La (for half-lives and nuclear properties see Experimental Section) in aerosols and deposition samples from the Arkhangelsk region.^[15]

The objective of this paper is to summarize our efforts to trace radioactive contaminations that may explain some circumstances of the release. In other words, the objective of this study was finding a nuclear "smoking gun" of this release. Both of the aforementioned scenarios were considered for this study: radionuclides released from a damaged nuclear reactor as well as radionuclides released from a powerful source. In any case, we limited our investigation to gamma-emitting radionuclides; other radionuclides such as alpha-emitting actinides (e.g., ^{238}Pu) or pure beta-emitters (e.g., ^{90}Sr) were not investigated because it was deemed unlikely to find any indicative traces given their ubiquitous abundance in the environment and also their low volatility, which would have caused only a marginal release. Also, the spike in dose rate in neighboring Severodvinsk points at a release of gamma emitters. In addition to particulate radionuclides which would likely be deposited in air filters, we investigated a possible release of the noble gas ^{42}Ar , thus testing a hypothesis that was stipulated recently.^[8] The ^{42}Ar would be the result of a damaged ^{42}Ar - ^{42}K source (see discussion below).

Results and Discussion

Various samples were investigated in this study. For basic radionuclide data, see Experimental Section.

Vessel air filters and cargo paper cover. A total of two air filters were retrieved from two vessels that were in the Severodvinsk/Arkhangelsk region of the White Sea at the time of the accident. They were analyzed by gamma spectrometry, targeting short- and medium lived fission products. Filter 1 (F1) (from vessel no. 1) was a commercial plastic fleece from an air condition inlet. In addition to this filter, also a paper cover (PC1) covering the cargo was retrieved for analysis from vessel no. 1. Filter 2 (F2) (from vessel no. 2) was the inlet air filter of the ship's engine.

In various consecutive measurements at the University of Bremen, low-level laboratory VKTA Felsenkeller, and the Advanced Radionuclide Gamma-spectrometer (ARGO) system at Pacific Northwest National Laboratory (PNNL) (in this order), no traces of short-/medium-lived anthropogenic radionuclides were found (Table 1). Find descriptions of the labs' instrumentation below.

Of all possible fission products, ^{131}I , ^{140}La (daughter of ^{140}Ba), $^{129\text{m}}\text{Te}$, ^{103}Ru , and ^{137}Cs were selected for Table 1. No detections of other fission products were observed either. Some inconsistencies were found for the ^{137}Cs activities found in F1 between Felsenkeller and PNNL-ARGO, which is likely due to inhomogeneities within the sample.

For the air filters, presence of natural atmospheric ^7Be can be viewed as quality control indicating the airflow through the filter. Presence of ^{137}Cs in air is due to the normal radiocesium background in northern Europe (resulting from Chernobyl fallout and atmospheric nuclear weapons testing from the 20th century).

Since all filters were taken from ships north of the blast site, whereas the plume was carried in southward direction, the negative result can be explained by the specific meteorological conditions in the area.

Ro5 air filters. Although the plume from the detonation was carried inland into the Russian Federation for several days, two Ro5 stations in northern and southern Europe were possibly affected days after the accident by a very diluted plume. The Norwegian Radiation and Nuclear Safety Authority (DSA) air filter station at Svanhovd, according to meteorological models published by Environment and Climate Change Canada,^[16] would have been hit by a diluted plume around August 12/13, 2019 (Figure 1). During the sampling period from

August 9–12, 2019, this station reported minute concentrations of particulate ^{131}I in the low $\mu\text{Bq}/\text{m}^3$ range.^[17] It shall be noted that ^{131}I detections are occasionally (6–8 times a year) reported in the very north of Europe.^[17] The detection, therefore, cannot be linked unequivocally to the Nenoksa incident.

CTBTO's atmospheric transport models^[13] indicated that the plume might have reached Greece on August 16–17, 2020 (Figure 2). In order to investigate any radioactive traces in this area, an atmospheric aerosol sample on a filter (F3) collected at the Ro5/Global Atmosphere Watch (GAW) station operated by the Environmental Radioactivity Lab, Institute of Nuclear & Radiological Sciences and Technology, Energy & Safety (IN-RASTES) in Athens, Greece was included in this study. The filter was taken in a high volume impactor from August 13–20, 2019.

The measurement of the high-volume sampler in Greece, however, did not reveal any detectable traces of anthropogenic radionuclides other than ^{137}Cs in the air filter. The ^{137}Cs activity concentration was in the normal range and can be attributed to the typical background.

It is likely that the air-masses passing over the test site did not reach the vessels' location because the plume was carried first in eastern and then in southern direction (see HYSPLIT plot in Figure 3).

In conclusion, no "smoking gun" was identified in our independent air filter measurement. When considering the reports from Rosgidromet,^[15] it is obvious that the list of fission products detected after the release (^{91}Sr , ^{139}Ba , ^{140}Ba , ^{140}La) excludes ^{131}I , which, due to its volatility, is considered one of the most prominent (and radiotoxic) fission products in reactor accident scenarios. A similar pattern (most notably the absence of radioiodine) has been observed previously at a non-seismic event in the Democratic People's Republic of Korea in 2010.^[18] That release marked a unique event in which numerous detections of ^{133}Xe , ^{135}Xe , ^{140}Ba , and ^{140}La were reported from the IMS during the time span of May 13 to 22, 2010 with remarkable spatial and temporal coincidence. It was concluded then that the event (possibly a low-yield or sub-critical nuclear test) most likely only released radioactive noble gases but held back other elements. Beta-minus emitting radioiodine nuclides have no noble gas precursor, but radiostrontium fission products have a radiokrypton precursor, and both radiobarium and radiolanthanum have radioxenon precursors. It could then

Table 1. Tabulated results of the gamma spectrometry (in mBq) or limits of detection (LoD) of radionuclides in vessel air filters no.1 and 2, and in cargo paper cover. Note that short-lived radionuclides are mentioned in early measurements only, whereas long-lived radionuclides are relevant for later measurements only. Activities and LoDs are given for the time of measurement.

Sample	Laboratory	Days past Aug. 08, 2019	^7Be [a]	^{131}I	^{140}La	$^{129\text{m}}\text{Te}$	^{103}Ru	^{137}Cs
F1	U. Bremen	11	6200 ± 400	< 12	< 23	< 370	< 10	< 13
F1	Felsenkeller	30	5100 ± 500	< 1.5	< 2.0	N/A	N/A	2.2 ± 0.5
F1	PNNL-ARGO	91	5330 ± 50	N/A	< 18	< 140	< 5	11.3 ± 2.7
F2	PNNL-ARGO	61	50100 ± 200	N/A	< 16	< 170	< 6	36.7 ± 3.8
PC1	U. Bremen	11	< 480	< 66	< 110	< 1820	< 54	< 100
PC1	Felsenkeller	30	< 26	< 2.2	< 2.9	N/A	N/A	29 ± 3
F3	PNNL-ARGO	70	41000 ± 100	N/A	< 27	< 130	< 4	6.0 ± 1.0

[a] Natural atmospheric radionuclide – N/A: Not applicable.

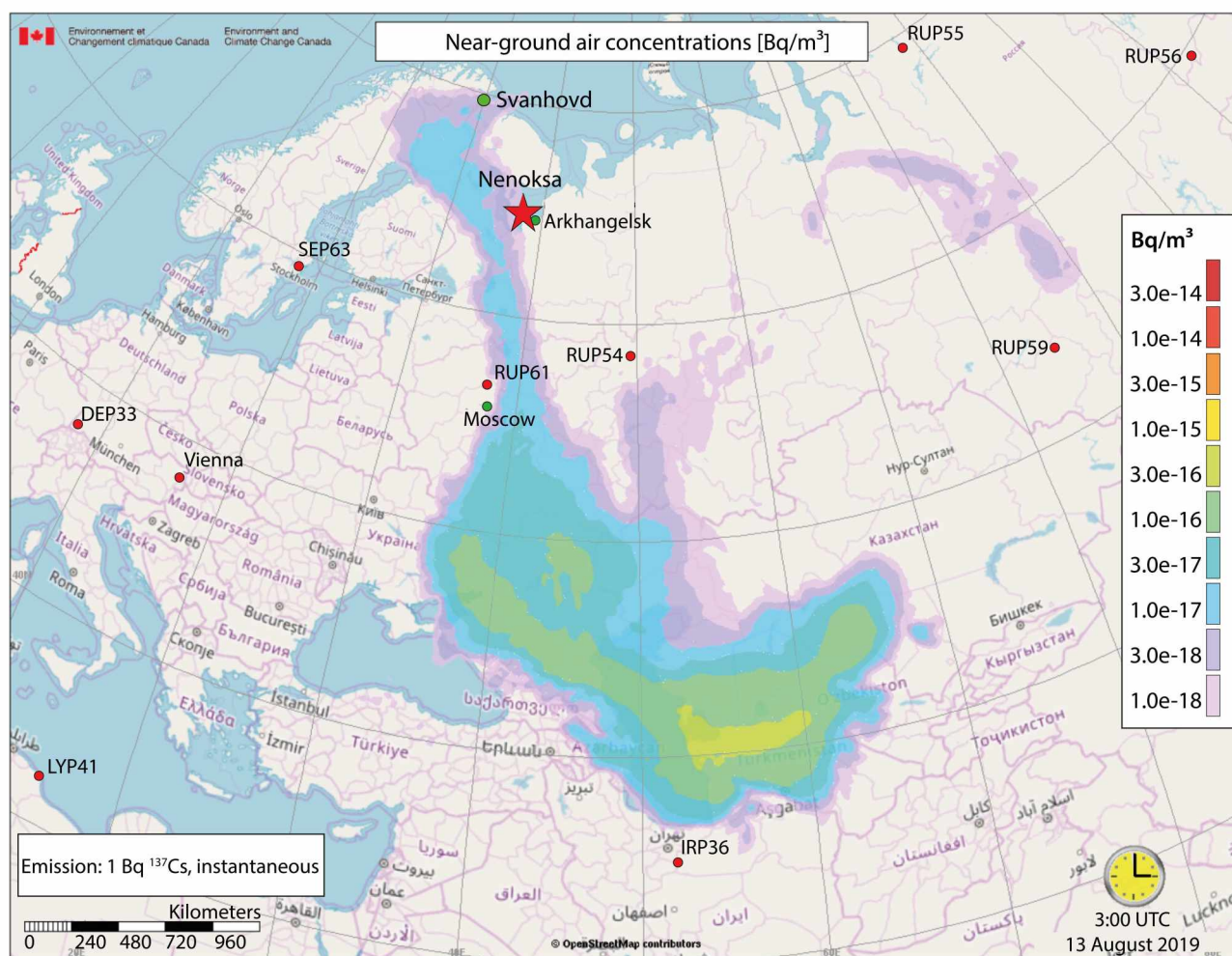


Figure 1. Environment and Climate Change Canada simulation of the plume finally reaching Northern Norway (Svanhovd monitoring station) by August 12/13, 2020. The Nenoksa test site is indicated as the red star. Radionuclide stations of the IMS are also shown. Modified after.^[16]

be concluded that the release only included noble gas emissions (followed by radioactive decay to other elements). This characteristic is a familiar pattern for underwater releases (in particular underwater nuclear tests), which usually scavenge metal ions, but mainly release noble gases. A confirmation of this hypothesis would have been possible by IMS monitoring data from the Russian (noble gas) stations, but no radioxenon data were transmitted either. If a release to the atmosphere only included noble gases, then the detection of ^{131}I in northern Norway was indeed the result of an independent event and uncorrelated to the Nenoksa incident.

Radioargon. Another hypothesis about the circumstances of release was put forward by Mietelski and Povinec,^[8] who suggested that the nuclear jet-engine may have been based on ^{42}Ar - ^{42}K . Argon-42 is a long-lived pure beta emitter with a short-lived daughter (^{42}K) that emits highly energetic beta particles (3.5 MeV max.) and characteristic gamma-rays. In an nuclear heat source, ^{42}Ar acts as a generator nuclide that constantly produces ^{42}K , which would be trapped in the heat exchanger and heat the surrounding air through its highly energetic beta

particles. Argon-42 is produced by rare double neutron capture in ^{40}Ar (especially by nuclear explosions), and thus has been present in the atmosphere only in ultra-trace levels ($< 0.7 \text{ nBq m}^{-3}$)^[19] since World War II. Here, we wanted to test if these levels have suddenly increased since the Nenoksa incident, which would be indicative that the nuclear missile had used an ^{42}Ar - ^{42}K generator as a heat source. Measuring ^{42}Ar directly at such low activities is impossible, but even measuring the gamma emission from its daughter nuclide ^{42}K diluted in the atmosphere would be extremely challenging. However, by measuring ^{42}K gamma emissions from a commercial tank with liquefied argon, the radionuclide would be accumulated, which would allow detection of a release of ^{42}Ar . In general, argon is produced from atmospheric argon through the Hampson-Linde cycle. It is assumed that in the Hampson-Linde cycle, the isotopic ratios remain unaffected, thus measuring liquefied argon does not involve the risk of isotopic fractionations or shifts (through which ^{42}Ar might be depleted). Similarly, ^{85}Kr (dispersed globally by nuclear fuel reprocessing) can be found

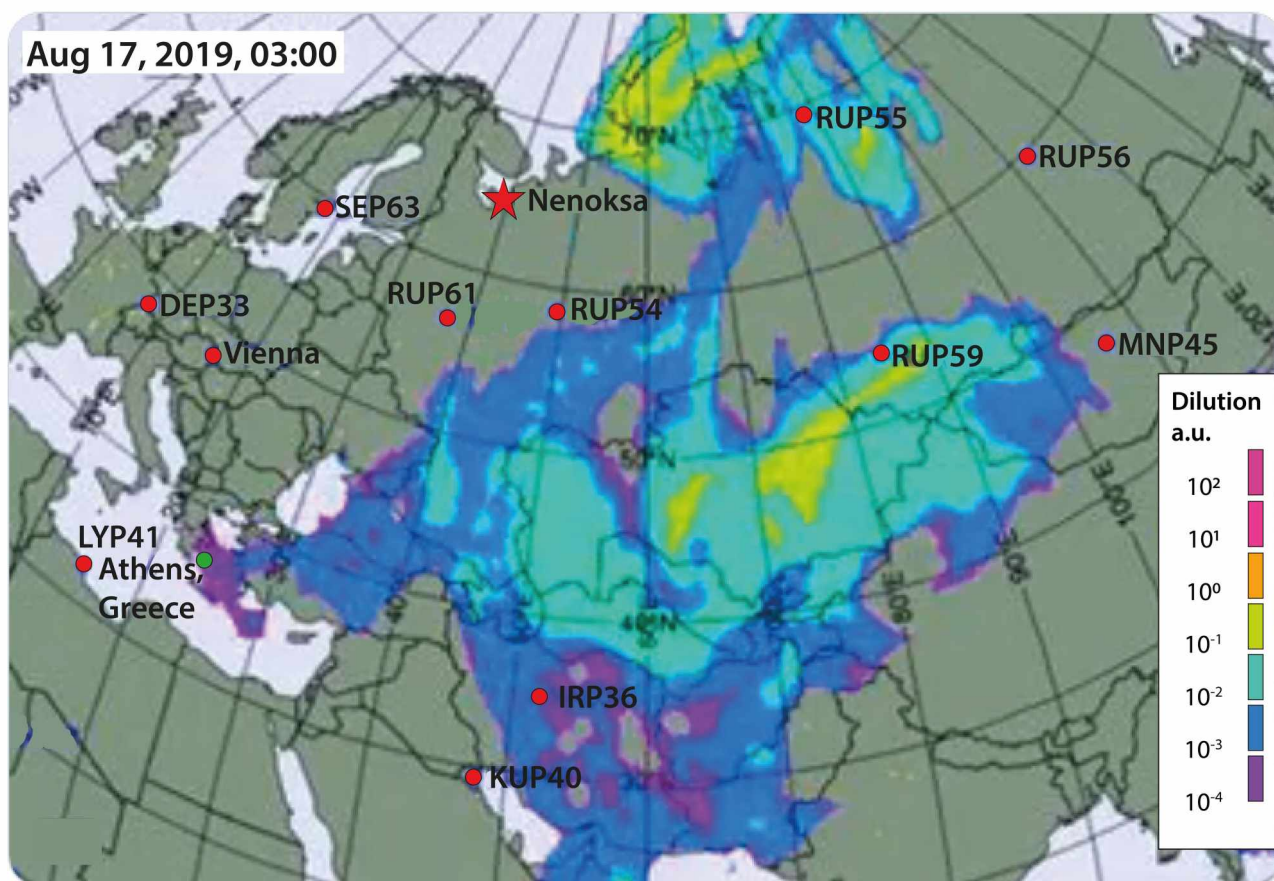


Figure 2. CTBTO dispersion simulation of the (highly diluted) plume reaching Greece by August 17, 2020. The Nenoksa test site is indicated as the red star. Radionuclide stations of the IMS are also shown. Please note that color scale refers to relative concentration values. Modified after.^[13]

in liquefied krypton gas, in considerable activity concentrations.^[20]

Mietelski and Povinec proposed an activity $> 1 \text{ PBq } ^{42}\text{Ar}$ for this type of propulsion. This activity may be split into several gas tanks, though. If we assume 1 PBq of ^{42}Ar injected into the atmosphere, we would expect a count rate at the ^{42}K 1525 keV photopeak of 0.1 cts/s in our measurement setup at the argon tank, summing up to 2370 cts over the measurement period. However, the LoD of 41 cts over the period of the measurement (December 12, 2019) was not exceeded (see Figure 4). Assuming complete and homogeneous mixing and assuming that 100% of the argon contained in the tank was collected from the atmosphere after the release (August 8, 2019), any release of ^{42}Ar above $1.7 \cdot 10^{13} \text{ Bq}$ would have exceeded our LoD.

We thus may conclude that no trace of ^{42}Ar was found, hence indicating that the circumstances of the release did not yet result in homogeneous mixing, did not involve ^{42}Ar , or involved a much smaller amount than proposed by Mietelski and Povinec.^[8]

Conclusions

In all the measurements conducted in this study, no traces of radionuclides were found that would provide any clues about the nature of the release. It is likely that the two vessels whose air filters were part of this study were located north of the plume and were not reached by it. For the Greek air filter investigated in this study, the plume was likely too diluted to yield detectable traces (provided that the plume reached Greece within a reasonable timeframe). If we consider detections of radiostrontium, radiobarium, and radiolanthanum isotopes reported by Rosgidromet, a possible explanation would be that only radioactive Kr and Xe noble gases were released from an underwater detonation. These detections would be in line with the observation of the radiobarium and radiolanthanum isotopes after a nuclear event in the Democratic People's Republic of Korea in 2010,^[18] which were explained with a release of noble gases only, which then decayed to the Ba and La radionuclides. Furthermore, no traces indicating a release of radioactive Ar were found. Therefore, we cannot confirm that the release involved radioargon from a ^{42}Ar -fueled radionuclide source.

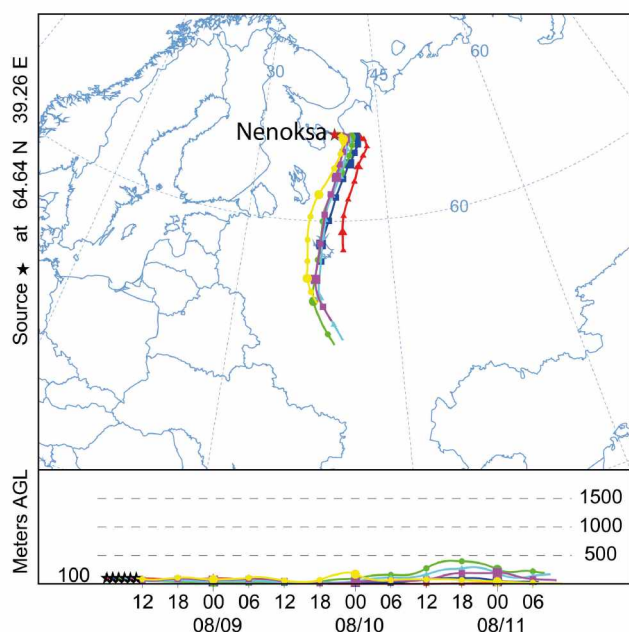


Figure 3. 72 h forward atmospheric transport simulation (HYSPLIT forward modelling) from the Nenoksa test site (indicated as the red star) beginning on August 8, 2019, 06:00 UTC. Altitude dispersion is given in meters above ground level (AGL).

Experimental Section

Nuclides of interest. For a list of relevant radionuclides and their nuclear characteristics, see Table 2.

Monitoring stations. One Greek Ro5 station participated in this study, in particular, the Environmental Radioactivity Lab at INRASTES belonging to the National Centre for Scientific Research (NCSR) "Demokritos".^[21] The monitoring station is located at N37°59', E23°49'. The cellulose air filters (quantitative Whatman41) were inserted on August 13, 2019 at 12:40 (local time) and exchanged on August 20, 2019 at 12:35 (local time) (total sampling duration 167.6 h, total volume 5738 m³). The sampling station employed a high volume impactor with two stages and a backup

Table 2. Nuclear data of relevant radionuclides.

Radionuclide	Decay type	Half-life	Dominant γ -photo peak (keV)	Gamma emission probability (%)
⁷ Be [a]	ec/ β^+ , γ	53.22 d	477.6035	10.44
⁴² Ar	β^-	32.9 y		
⁴² K	β^- , CE, γ	12.360 h	1524.67	17.9
⁹¹ Sr	β^- , CE, γ	9.63 h	1024.3	33.5
¹⁰³ Ru	β^- , CE, γ	39.247 d	497.085	91.0
^{106m} Rh	β^- , CE, γ	131 m	511.7	85
			621.93	9.93
¹⁰⁶ Ru	β^-	371.8 d		
^{129m} Te	β^- , CE, γ	33.6 d	695.88	3.1
¹³¹ I	β^- , CE, γ	8.0252 d	364.489	81.5
^{137m} Ba	CE, γ	2.552 m	661.657	90.1
¹³⁷ Cs	β^- , (γ)	30.08 y	(283.5)	5.8E-4
¹³⁹ Ba	β^- , CE, γ	83.06 m	165.864	24
¹⁴⁰ La	β^- , CE, γ	1.67855 d	1596.21	95.4
¹⁴⁰ Ba	β^- , CE, γ	12.7527 d	537.311	24.0

[a] Cosmogenic radionuclide – ec: electron capture – CE: conversion electron emission.

filter however, all fractions were measured together to assess the accumulation of the entire particulate matter suspended in air.

Vessel air filters. Air filters for this study were provided from the captains of their vessels under the condition that the names and positions of the ships would remain undisclosed, unless a positive result was found in the investigation. This was not the case. Both vessels were cruising in the White Sea not very far from Severodvinsk at the time of the accident. Vessel air filter F1 was collected from the air condition inlet on August 19, 2019 and weighed 7.7 g. It was made of a commercial plastic fleece. In addition to the air filter, a paper cover (PC1) protecting the cargo was provided for this study as well. The paper cover was exposed to the atmosphere and covered approx. 1 m² and weighed about 200 g. The second vessel air filter F2 originated from a different ship and was used to clean the incoming air to the engine. Although this filter was oily and dirty, presence of natural

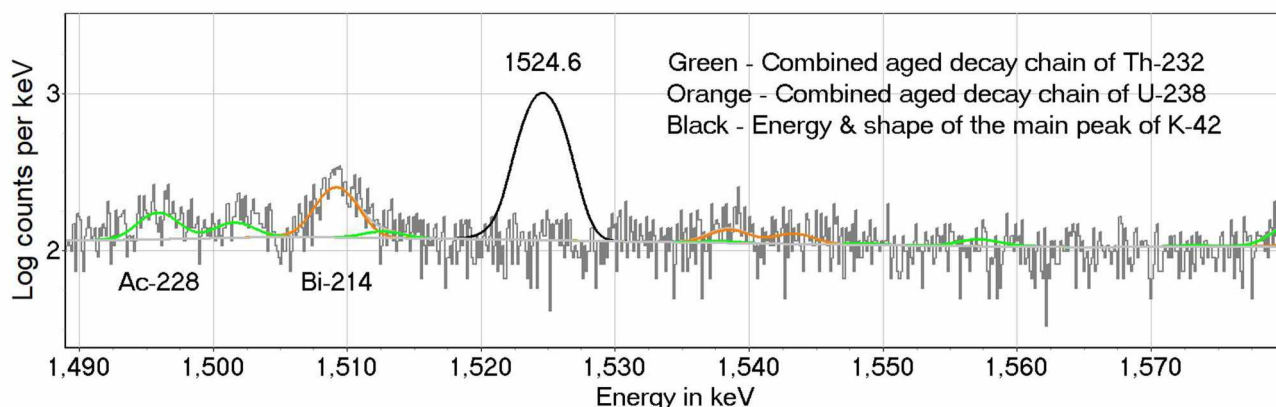


Figure 4. Section of the gamma spectrum showing the lack of a peak at 1524.6 keV. The VRF software from Snakedance Scientific identified neighboring peaks belonging to ²²⁸Ac and ²¹⁴Bi. The position and shape of a 1524.6 keV peak of ⁴²K (if it were present) is also shown. The hypothetical peak area of the simulated peak at 1524.6 keV is about 3590 cts. This would correspond to a release into the atmosphere of about 1.5 PBq of ⁴²Ar in the course of the incident.

atmospheric radionuclides indicated a reasonable airflow and contact to the surrounding atmosphere.

Gamma-ray spectroscopy. Various gamma-ray spectrometers were used in this study. The first measurements (of air filter F1 and cargo paper cover PC1 only) were conducted at University of Bremen, followed by the VKTA lab Felsenkeller (F1 and PC1 only), the ARGO system at PNNL (F1, F2, and the Greek air filter F3), and Leibniz University of Hannover (liquid argon tank measurements).

University of Bremen laboratory. Two high-purity germanium (HPGe) gamma-ray spectrometers with similar characteristics were used for the measurements of F1 and PC1. One detector had a volume of 213 cm³, relative efficiency of 50% at the 1332 keV peak of ⁶⁰Co, and a resolution (full width at half maximum, FWHM) of 1.9 keV at the 1332 keV peak of ⁶⁰Co. The second detector had a volume of 193 cm³, relative efficiency of 50%, and FWHM of 2.1 keV at the 1332 keV peak of ⁶⁰Co.

VKTA Felsenkeller. The low level underground laboratory Felsenkeller^[22] of VKTA provides geological shielding equivalent to the shielding of 138 m of water.^[23] Gamma measurements of F1 and PC1 were conducted with a HPGe p-type detector with enhanced front-side sensitivity (sensitive volume 361 cm³, relative efficiency 92%, FWHM at 1.3 MeV: 1.9 keV) installed inside a graded shield and with nitrogen flushing of the interior.^[24]

PNNL-ARGO. The ARGO system is located in in the Shallow Underground Laboratory at PNNL (30 m water equivalent).^[25] The ARGO includes two Broad Energy Germanium Detectors (BEGe; relative efficiency 60%) with a germanium crystal with 5000 mm² active area. The BEGe detectors are surrounded by 12 NaI detectors (10,000 cm³) for Compton suppression purposes. The ARGO system is located in a low-background graded shield consisting of 200 mm Pb, 12.5 mm 30% borated polyethylene (PE), 1 mm Cd, 50 mm Pb, 1 mm Sn, 1 mm Cu. The interior of the system is flushed with N₂ for removal of Rn and its progeny. As a cosmic radiation veto system, the shield is surrounded by 6 Eijen Technology polyvinyltoluene (PVT) plastic scintillator plates, each with two photomultipliers installed.^[26] The ARGO system is optimized for the gamma energy range between 3 and 3000 keV. Samples (F1, F2, and F3) were installed in a calibrated geometry between the BEGe detectors.

Leibniz University Hannover. For the investigations of gamma emission from the argon tank at Leibniz University Hannover, a HPGe detector was installed at half filling level of the tank at a distance of less than 3 cm (Figure 5). The HPGe detector was a horizontally mounted coaxial Ge crystal with a volume of 252 cm³ and a relative efficiency of 65%, and a FWHM of 1.8 keV at the 1332 keV peak of ⁶⁰Co.

Radioargon measurements. For testing of the ⁴²Ar-⁴²K hypothesis,^[8] several assumptions had to be made. The total mass of the atmosphere is 5,1441 · 10¹⁸ kg. Natural Ar constitutes an abundance



Figure 5. Measurement setting of gamma emission from the argon tank.

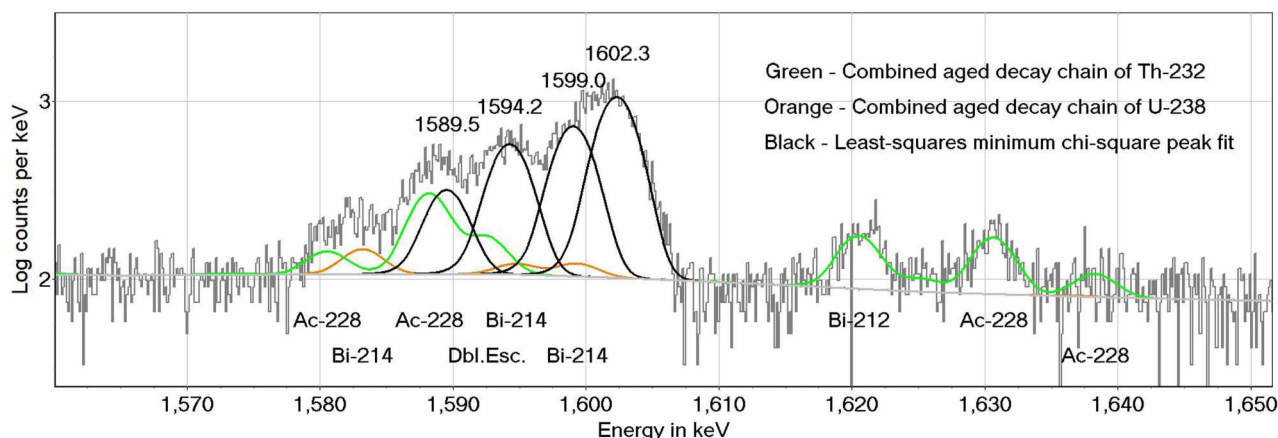


Figure 6. The anomalous quadruple peak in the gamma spectrum of a tank of liquid argon as analyzed with VRF software from Snakedance Scientific. The software suggests partial contribution to the peaks from naturally occurring radionuclides. However, not all peaks can be assigned fully.

of 1.288 mass %. The liquid argon tank at Leibniz University Hannover had dimensions of 7.375 m in height and 2 m in diameter. The tank was refilled with argon in November 2019, and had a filling level of 45 % (of a maximum content of 21198 kg) at the time of the measurement (December 12, 2019). By this date, we assume a homogeneous atmospheric mixing of any radioargon with the atmosphere of the northern hemisphere only.

Measurements were conducted with the aforementioned gamma-ray spectrometer and lasted 23192 s. The peak efficiency of this cylindrical source was calculated according to Aguiar.^[27] The gamma spectrum was evaluated using the Snakedance Scientific, LLC, gamma spectrum software VRF.

In the relevant energy range around 1525 keV (characteristic for ⁴²K), no signs of a peak were detected in the spectrum (see Figure 4). However, a rather anomalous quadruple peak was observed between 1578 and 1607 keV, as shown in the portion of the spectrum in Figure 6. A least-squares best fit decomposed this feature with minimal error into four peaks centered at 1589.5, 1594.2, 1599.0, and 1602.3 keV. No source of these peaks could be identified. Until further investigation is possible, it is presumed that the source of these peaks is either as-yet unexplained natural causes or possible problems with instrumental processing.

Atmospheric transport modelling. HYSPLIT (online version) was used to model the early atmospheric transport of the plume.^[28]

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