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$^{240}Pu/^{239}Pu$ Atom Ratio as A Fingerprint of Local and Tropospheric Fallout Due to Events Involving Nuclear Weapons: A Review

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Abstract: A series of events involving nuclear weapons (atmospheric weapon tests, explosions in military use, aircraft accidents….) have generated the dissemination at local and/or regional scale of radionuclide debris characterized in each case by a characteristic $^{240}Pu/^{239}Pu$ atom ratio. This ratio could be used as a fingerprint to evaluate the weight of a particular event in the contamination of an environmental compartment.
In this paper is compiled a review of the $^{240}Pu/^{239}Pu$ atom ratios characterising the great majority of events involving nuclear weapons where dissemination of Pu isotopes in the environment were generated. Only data found in the open literature are included, although as a challenging issue some few events with no available public data are also presented.
Keywords: Plutonium, Nuclear weapons, Environmental impact.

1 Introduction

It is well known that the primary sources of anthropogenic radionuclides in the environment are the atmospheric nuclear tests that were carried out during the period from 1945 to 1980. A total of 543 atmospheric detonations were conducted with a total yield of 440 Mt. In addition in some other events where directly nuclear weapons were involved (military use in Japan, aircrafts accidents in Thule and Palomares,…) dispersion of anthropogenic radionuclides also occurs.

Atmospheric nuclear testing took place through four decades. Three periods can be easily identified. The first period (1945 to 1958) was dominated by US testing at equatorial sites (Bikini, Enewetak and Johnston Island). This period ended in 1958 with the implementation of a moratorium (1959-1961). In the second period (1961 and 1962) Soviet Union testing at Novaya Zelmya and Semipalatinsk dominated until the limited ban treated was implemented in 1963 finishing all the USA and USSR atmospheric tests. The third period included the 1964-1980 Chinese (Lop Nor) and French (Mururoa and Fangataufa) tests.
Debris from nuclear detonations may be divided into three impact classes: local, regional (tropospheric) and global (stratospheric), being their apportionment into the different atmospheric compartments dependent primarily on the yield of detonation, the height and the latitude at which the test took place. Debris from low yield detonations remained almost completely in the troposphere and deposited downwind of the detonations (the residence time in the troposphere is of several days), while high-yield detonations injected most of the debris into the lower or upper stratosphere. Debris inserted into the stratosphere was removed by atmospheric exchange processes and deposited essentially uniformly in the surface of the hemisphere on which the detonation took place.
The threshold yield needed for injection of debris in the stratosphere is latitude dependent because the thickness of the troposphere is clearly higher at low than at high latitudes. The troposphere covers 0-9 km width in the pole and 0-17 km in the equator, and consequently, while only equatorial tests with > 3 MT penetrates debris in the stratosphere, in the pole is only needed yields higher than 0.3 MT.

The apportionment between the different compartments was also dependent of the altitude at which the test was performed. Tests were conducted from a variety of platforms and at various heights above the ground. The

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platforms included wooden and steel towers ranging from a few meters to over 100 m high, balloons at heights of 100 to 400 m, drops from aircrafts, surface shots, underwater shots, shot on barges anchored in lagoon, shots at very high altitude using rockets and subsurface cratering shots.

In the great majority of tests performed involving fission, ^{239}Pu was the main constituent as fissile material, although some amounts of other Pu radio nuclides such as ^{238}Pu, ^{240}Pu and ^{241}Pu can be also present. Between these additional radio nuclides, ^{240}Pu has a relatively high rate of spontaneous fission that gives rise to neutron emission, fact that would make the nuclear weapon device hazardous and unpredictable because of the preinitiation of the trigger. ^{240}Pu is then a contaminant in nuclear weapons and its presence should be reduced at maximum. The Pu used in nuclear devices is for that reason classified as weapon-grade (^{240}Pu <7%), or super-grade Pu (^{240}Pu <3%).

The $^{240}Pu/^{239}Pu$ atom ratio existing in the undetonated material can experiment variation in the explosion, being the final value of this ratio dependent on the yield and design of the bomb. Low-yield detonations (0 – 300 kT) produce debris which differs little from undetonated weapon material, while very large detonations (above 5 MT) produce higher $^{240}Pu/^{239}Pu$ atom ratios.

The map of Pu contamination over the world due to the weapon tests is marked from an evenly distribution of this radionuclide coming from the stratosphere and originally formed in high or very high detonations, with some additional localized or regional spots associated to local and regional fallout due to tropospheric debris associated to all the weapons tests performed. The unevenly global fallout is characterized over the world with independence of latitude and hemisphere by a $^{240}Pu/^{239}Pu$ atom ratio of 0.18 which represents an integrated average value of these atom ratios in the different stratospheric inputs of this element. The local or regional spots associated to weapon tests events with enhanced concentrations of Pu will be characterized by $^{240}Pu/^{239}Pu$ atom ratios in general different that the observed one in areas affected only by global fallout. If the local or region spot has produced a severe Pu contamination, the contribution in this area of the Pu with origin in the global fallout will be negligible and the $^{240}Pu/^{239}Pu$ atom ratio will be the corresponding one to the local/regional fallout. If the Pu tropospheric contamination in the area analysed is comparable to the global fallout, in these areas the $^{240}Pu/^{239}Pu$ atom ratio will be a weighted average of the atom ratios corresponding to both contributing sources.

In the following sections, a review of the $^{240}Pu/^{239}Pu$ atom ratios determined at local (and in some cases at regional) level in places affected by nuclear weapon events will be performed. These events include mostly atmospheric weapon tests, but also weapon explosions in military combats and nuclear weapon accidents.

2 Events Involving Nuclear Weapons

The events analysed will be the following, detailed mostly in chronological order:

Atmospheric weapon tests and weapons military used

- Trinity Test (USA): 1945
- Nagasaki (USA): 1945
- Marshall Islands (USA): 1946-1958
- Semipalatinsk (USSR) : 1949- 1962
- Nevada Test Site (USA): 1951- 1962
- Australia (UK): 1952-1963
- Totsk (USSR): 1954
- Nova Zelmya (USSR): 1955-1962
- KasputinYar (USSR): 1958-1962
- Malden Island (UK) 1957
- South Atlantic (USA): 1958
- Christmas Island (UK, USA): 1958-1962
- Jhonston Island (USA): 1958-1962
- Algeria (France): 1960-1966
- Lop Nor (China): 1964 – 1975
- Mururoa and Fangataufa (France): 1966-1974

Weapons accidents

- Bomarc 1960
- Palomares 1966
- Thule 1968

Trinity

The first important event affecting dispersion of plutonium radionuclides in the environment and involving nuclear weapons was the nuclear weapon test conducted on July 16, 1945 by USA, near Alamogordo, New Mexico. This weapon test is colloquially known as Trinity test.

It was a low-yield explosion test of 21 kT, performed in a tower several meters over the surface. The core of the weapon was formed by super-grade weapon ^{239}Pu produced in Handford. At the moment of explosion, and due to the tremendous temperatures reached, the disseminated radioactive debris experimented its fusion with the local soil, being formed a glassy material that is known with the name of Trinitite.

Due to the low yield of the explosion, mostly local fallout was generated in the form of Trinitite material. This Trinitite includes fission products from the ^{239}Pu fission, and activation production from neutron reactions with nuclear fuel, with the soil and with the tower where it was the weapon placed. And also remain unburned nuclear fuel.

The low yield of the explosion did no alter appreciably the $^{240}Pu/^{239}Pu$ atom ratio in the debris in comparison with the originally existing in the nuclear fuel. In several Trinitite fragments the $^{240}Pu/^{239}Pu$ atom ratio value ranges between 0.015 and 0.025 [1].

Place	Years	Main tests	$^{240}Pu/^{239}Pu$ atom ratio
Weapon events (atmospheric and surface tests, safety trials)			
Trinity Test (USA)	1945	Low-yield explosion tests	0.015 - 0.025
Nagasaki (Japan)	1945	Low-yield explosion	0.032 - 0.037
Marshall Islands (USA)	1946 – 1958	67 tests, some with high yield	0.06 – 0.36
Semipalatinsk (USSR)	1949 – 1962	116 tests, most low-yield tests	0.04 – 0.07
Nevada Test Site (USA)	1951 – 1962	>100 tests, low yields	0.03 – 0.04
Australia (UK)	1952 – 1963	12 low yield tests	0.04 – 0.05
Totsk (USSR)	1954	1 low yield test	No data found
Novaya Zelmya (USSR)	1955 – 1962	130 tests, most high yields	< = 0.10-0.12
KasputinYar (USSR)	1956-1961	11 tests, high-altitude	No data found
Malden Island (UK)	1957	3 tests low yields	No data found
South Atlantic (USA)	1958	3 tests, high altitude	No data found
Christmas Islands (UK, USA)	1958 – 1962	30 weapon tests	No data found
Jhonston Island (USA)	1958 – 1962	12 weapon tests	No data found
Algerie (France)	1960 – 1966	4 low yield tests, safety tests	<= 0.06
Lop Nor (China)	1964 – 1975	21 tests, some high yields	0.06 – 0.22
Mururoa&Fangataufa (France)	1966 – 1974	41 atmospheric tests	0.03 – 0.05
Nuclear weapon accidents			
Palomares (Spain)	1966	2 weapons involved	0.064
Thule (Greenland)	1968	4 weapons involved	0.028 – 0.055
BOMARC (USA)	1960	1 weapon involved	0.056 – 0.059

Nagasaki

After the Trinity test, the following event involving nuclear weapons with the dispersion of radioactive debris in the environment containing plutonium, took place in Nagasaki (Japan) on August 9th, 1945 due to the military use of a Pu nuclear weapon. The previous military nuclear explosion in Hiroshima involved a ^{235}U enriched bomb and the contribution of the transuranic elements was found to be negligible small compared with the local fallout.

In spite of the tremendous destruction generated, the weapon explosion in Nagasaki can be considered a low-yield explosion of 22 kT, which provokes mostly local fallout. In the ground zero area $^{240}Pu/^{239}Pu$ atom ratios in the range 0.032-0.037 were found [2], indicating that the disseminated Pu mostly corresponds to unburned plutonium. Signals of the Pu disseminated due to explosion, is restricted to few km from the ground zero.

Marshall Islands

The first campaign of USA atmospheric nuclear weapon tests was performed in Marshall islands, Pacific ocean, from 1946 until 1958. A total of 67 tests were performed in the Enewetak and Bikini atolls, located in the northern hemisphere. From these tests, a total number of 23 were performed in the different islands forming part of the Bikini atoll.

The nuclear test program included air drops, barge detonations, tower detonations, surface detonations and underwater detonations. The total yield for all test conducted on Bikini and Enewetak atolls was 109 MT (75% in Bikini atoll) involving some high-yield thermonuclear explosions.

These nuclear explosions produced significant quantities of local fallout and in some cases, due to its high-yield, regional/ tropospheric and stratospheric fallout. Each event was characterized by a particular $^{240}Pu/^{239}Pu$ atom ratio value, which was affected by release conditions and the type/design of the device.

Although the number of weapon tests in the Bikini atoll was 23, the close-in fallout contamination in this atoll is dominated by the debris generated in 1954 by the so called BRAVO test, the explosion of a thermonuclear weapon with the higher yield in the Marshall Island (15 MT). The yield of this test was at least two times higher than expected; provoking the contamination of the Rongelap and Ultrik atolls located in the vicinity of the Bikini atoll, at the east. The soils of the islands forming the Bikini atoll are

characterised by a $^{240}Pu/^{239}Pu$ atom ratio of 0.30 [3], ratio that can be associated to the heavy influence of the BRAVO Test (the very high yield of this test induces the generation of ^{240}Pu in considerable amounts and a sharp increase of the $^{240}Pu/^{239}Pu$ in relation to the expected one in unburned fuel).

This ratio of 0.30 provides a characteristic marker for evaluating the range and extent of the BRAVO deposition over the region. In this sense, analysis of a composite sample soil sample collected in the Rongelap Island shows a $^{240}Pu/^{239}Pu$ atom ratio of 0.28 that is consistent with the knowledge than Rangelop Atoll was also contaminated by the BRAVO tests [3]..

Anecdotally, it can be indicated that in the crater generated by the BRAVO test were found $^{240}Pu/^{239}Pu$ atom ratios lower than 0.30, due to the simultaneous influence from other explosions performed in the area with much lower yield [4].

In the case of the Enewetak atoll (located at the west of the Bikini atoll), the $^{240}Pu/^{239}Pu$ atom ratios have a variety of values in their different islands showing the peculiarities of the different tests carried out there. In the Enewetak atoll, in 1952, was performed the first high-yield thermonuclear test by USA (MIKE test), being characterised its debris by a $^{240}Pu/^{239}Pu$ atom ratio of 0.36. After the MIKE test, and in order to be prepared for future explosions, the Runnit island (part of the Enewetak atoll) was cleaned, being the contaminated soils bulldozed and disposed.

Most of the contamination that they remain nowadays in the Runit Island was due to tests carried out after the MIKE test. It comprises a surface safety test (Quince safety test), an unsuccessful surface detonation which had no or minor fission yield, < 0.02 kT (called Fig test) and 17 nuclear atmospheric tests (13 of them with yields < 100 kT, and 4 with yields < 4 kT). The low yields of the tests carried out and the dispersion of unburned fuel in the safety test, is reflected in the fact that the soils of this island are characterized by a low $^{240}Pu/^{239}Pu$ ranging in the interval 0.06 – 0.10 [5].

The magnitude and yields of some of the tests performed in the Marshall Islands (sites called by the Americans as Pacific Proving grounds) have generated also a remarkable impact at regional level. In this sense:

a) the $^{240}Pu/^{239}Pu$ atom ratios in seawater samples on East China were found to be 0,21 – 0,25 due to the delivery in addition to the global fallout of close-in Pu from the Pacific Proving Grounds (PPG). This PPG contribution in East China sea was estimated to be on 40% on average [6]. Sediment cores collected in the cost of Japan shows also the PPG signal [7].
b) The PPG impact was also noted in central North Pacific were two major sources can be inferred: close-in (troposphere) fallout from nuclear weapons testing at the PPG in the Marshall Islands in the 1950s and global (stratospheric) fallout which peaked in 1962. Seawater and sediment samples from the North Pacific exhibit a wide range of $^{240}Pu/^{239}Pu$ values (0.19 – 0.34), with a trend towards higher ratios in the subsurface waters and sediments [8].

Semipalatinsk

The Semipalatinsk nuclear test site (STS) is located in the northern-east of present-day Kazakhstan and was the first and one of the main proving grounds for the testing of nuclear weapons by the former Soviet Union. From 1949 until 1962 a total of 116 atmospheric tests (30 surface tests and 86 air tests) were performed while after the signing of the limited ban treaty additional 340 underground tests were done until 1989.

The former USSR nuclear weapon testing programme was mainly carried out at two testing sites; one at the Novaya Zelmya test site in Russian Artic for large yields (40 MT atmospheric) and the other at Semipalatinsk for smaller yield tests (atmospheric 6.6 MT).

All the tests in Semipalatinsk were performed in four main technical areas known with the codes “Sh”, “B”, “G” and M. All the atmospheric nuclear tests (surface and atmospheric) were carried in the zone “Sh” referred as GROUND ZERO. In addition, to the mentioned atmospheric tests four cratering explosions (Chagan, Telkem 1, Telkem 2 and Savy-Uzen) were carried in the site in order to evaluate the potential of using nuclear explosions for civil engineering purposes. In particular, the Chagan explosion caused the so called “lake Balapan” or “Atomic lake”.

Associated mostly to the surface explosions, in the GROUND ZERO the generated radioactive debris was mainly fused with soils at very high temperatures, producing glassy materials containing fission and activation products and transuranic radio nuclides. This glassy material due to the low-yield explosions carried out in the analysed area is characterized by a low $^{240}Pu/^{239}Pu$ atom ratio. Measurements performed in some of these particles have shown ratios in the range 0.04 – 0.05 [9].

The cratering explosions were also low-yield detonations, being generated also local fallout characterized by low $^{240}Pu/^{239}Pu$ atom ratios. However, although low, these ratios are slightly higher than the found ones in GROUND ZERO. In this sense, atom ratios of 0.067-0.072 in soils from lake Balapan (atomic lake), of 0.051 in soils from Telkem 1 and of 0.054 in soils from Telkem 2 has been obtained [10].

The atmospheric weapon tests carried out in Semipalatinsk

has also produced Pu regional contamination either by regional tropospheric fallout immediately after the tests or either by the delayed transport of contaminated soil particles by winds. The signal of the STS tests can be detected in villages located in their vicinity (the $^{240}Pu/^{239}Pu$ atom ratios in the villages of Akhzar and Kainer were 0.068 and 0.103, respectively, [11]), and also in soils situated even 300 km downwind STS, where the $^{240}Pu/^{239}Pu$ atom ratios allows to discriminate between the % of global fallout and the % of global fallout. In the mentioned area, 30-60% was found to be due to local fallout [12].

We will finish this section by indicating that a combination of state-of-the-art isotopic fingerprinting techniques and atmospheric transport modelling using real-time historical meteorological data has even allowed demonstrating the direct tropospheric transport of radioactive debris from specific nuclear detonations at the Semipalatinsk test site in Kazakhstan to a remote place such as Norway, crossing large areas of Europe [13]. The study was based on the analysis of air filters collected at different sites of Norway at the beginning of the sixties

Nevada

In 1951, several years after the first USA weapon tests in the Marshall Islands, and due to logistic problems associated with the remote Pacific site, a decision was made by the US authorities to develop a test site near Las Vegas, Nevada, where the first test was carried out on 27th January 1951.

Between 1951 and 1958 about 100 aboveground atmospheric tests were performed with 21 additional atmospheric safety tests between 1953 and 1963. Mostly after the signing of the ban treaty, from 1961 until 1993, other 828 underground tests were carried out.

Most of the aboveground atmospheric tests in the Nevada Test Site (NTS) were relatively low yield devices designed to test various design concepts. The largest was a 74 kT test conducted from a balloon at a height of 400 m in July 1957.

About one half of the tests conducted in this area were tower or surface shots and the total yield of the atmospheric test conducted in NTS was about 1 MT, a very small fraction of the total yield of the USA tests [14].

Due to the low yields atmospheric and the safety tests, radionuclide debris was disseminated mostly at local level, although a fraction was also disseminated at regional level through the troposphere. Although some clean-up operations have been performed, some contamination remains specially in the north-east of the NTS, being this contamination characterised as expected, due to the low or null yield origin, by a quite low $^{240}Pu/^{239}Pu$ atom ratio of 0.032 [15].

Atmospheric weapon tests in NTS provoked regional fallout in western USA, with Pu levels higher and $^{240}Pu/^{239}Pu$ atom ratios lower than expected due to global fallout. In the affected soils collected from different states surrounding NTS it was possible to see that a fraction of the Pu was present in a quite refractory form, being this finding consistent with the presence of spurious, high-activity particles containing NTS-derived Pu in a fused silicate matrix that cannot be dissolved completely by acid leaching. The $^{240}Pu/^{239}Pu$ atom ratios in the soils should be measured after applying a total dissolution method.

In this way, the following atom ratios were found in the neighbouring states to NTS: 0.06 – 0.09 in Nevada, 0.10 – 0.11 in Utah, 0.14 – 0.17 in Arizona and 0.14-0.17 in Colorado [16]

There are at least four possible source of Pu in soils located downwind from the NTS, i.e., in the soils from neighbouring states:
a) Stratospheric fallout from atmospheric testing (global), b) tropospheric fallout from NTS atmospheric testing 1951-1958,
c) tropospheric fallout from NTS safety tests, 1953-1963, and d) wind-transported NTS surface soils containing radioactivity from the atmospheric and safety [15].

The influence of the atmospheric tests at Nevada Test site has been also detected in lichens collected in New Mexico in 2011-2013. This fact suggests continuous resuspension of fallout even 50 years after ratification of the Limited Test ban Treaty. The lichens reflect regional atmospheric transport (resuspension) rather that contamination from adjacent soils because the lichens have no roots and take the nutrients from the atmosphere. The $^{240}Pu/^{239}Pu$ atom ratios in the collected lichens ranged from 0.06 to 0.16 [17].

Some regional effect of the atmospheric nuclear weapon tests carried out in NTS has been detected in the North Atlantic Ocean. A systematic decrease in the $^{240}Pu/^{239}Pu$ atom ratio in marine sediments is found with increasing water depth in North Atlantic (transect from Woods Hole until Bermuda). The range of $^{240}Pu/^{239}Pu$ atom ratios was between 0.18 in shelf and 0.10 at 5000m depth [18]..

In the North Atlantic Ocean two major sources of fallout Pu can be inferred: a major source is the global stratospheric fallout (0.18 of $^{240}Pu/^{239}Pu$ atom ratio and a relatively long residence time in water), and a second source with clearly lower $^{240}Pu/^{239}Pu$ atom ratio and much more efficiently removed from the water column. The only source of low ratio fallout of Pu which could account for the timing, inventories and refractory characteristics of this second component is surface-based low yield testing at Nevada Test Site. Solubility and reactivity of Pu from different fallout sources are not necessarily identical.

Australia

In total, 12 atmospheric nuclear tests (known as major trials) were conducted during 1952-1957 at the three former British nuclear test sites in Australia which included Maralinga, Emu Plains and the Montebello Islands. In Montebello a total of 3 tests were performed (Hurrican, 1952; Mosaic One, 1956 and Mosaic two, 1956) in Emu were carried out two tests (Tottem One, 1953 and Tottem Two 1952) and in Maralinga the remaining seven, each one in one different firing site (one of these firing sites is. called Taranaki). The nuclear weapon tests were performed from 30 m towers, from air drop, balloon and ground surface, having in all the cases lower fission yields (< 100 kT). [19].

In addition, hundreds of "safety" tests were conducted in the test sites of Emu and Maralinga. These "safety" tests were reported to investigate the performance of various components of nuclear devices from a single point accidental detonation which might result from fire, transportation accident or plane crash. In these "safety" tests, Pu dispersion was not by nuclear explosion, but involved burning and high-explosive detonations, which released sufficient energy from a vertical jet firing upward from the test device to lift Pu-contaminated debris to heights of more than 750 m [20].

In the Taranaki firing site (Maralinga), in addition to a nuclear weapon test, 22 kg of Pu was dispersed in 12 "safety" tests carried out in 1961-1963 and known as Vixen B minor trials.

The low-yield of the nuclear tests, and the characteristics of the safety trials, allow to affirm that most of the generated radioactive debris in these events have been deposited as local fallout, being in addition the contamination characterized by a low $^{240}Pu/^{239}Pu$ atom ratio.

In this sense, analysis of soils collected in Montebello from the plume generated by Mosaic two test indicate $^{240}Pu/^{239}Pu$ atom ratios in the range 0.046- 0.054, while this ratio reach clearly lower values (0.009 – 0.028) in soils collected from the plume generated by the Totem-1 test in Emu Plain site [20].

In the Taranaki site the set of tests performed, resulted in four narrow plumes extending more than 25 km as it has been evidenced by ^{241}Am mapping. In these plumes the $^{240}Pu/^{239}Pu$ atom ratio is 0.06 with most of the Pu deposited in very fine particulate form [21].

In 2001, a part of the Taranaki site has been remediated by removal and reburial of the most contaminated soils immediately surrounding the test pad and out along the deposition plumes to distances of about 1.8 Km.

Totsk

On 14 September 1954 the U.S.S.R. conducted a military exercise incorporating a nuclear test detonation. The exercise involved 44,000 Soviet troops at the Totsk range in the Orenberg region. Troops were positioned in trenches as close as 5 km to the designated ground zero, with one group of troops in a forward post about 2.5 km from the designated ground zero. A Tu-4 "Bull" bomber was used to deliver an RDS-3 nuclear gravity bomb of 40 kT yield, an implosion device using a composite uranium-plutonium core. The soldiers involved in the exercise wore gas masks, protective suits and respirators, special gloves and capes, and they moved around the territory in armoured personnel carriers, holding the distance of 400-600 metres from the hypocenter and avoiding the most dangerous areas of the explosion site.

It was a low-yield detonation that provokes local fallout contamination in the area. No particular restrictions were imposed in the use of the affected agricultural fields and in the consume of water from the area after the detonation. No data have been found in the open literature about the levels of contamination in the affected area either just after the accident or nowadays.

Nova Zelmya

The Novaya Zelmya Test Site, located in the Russian Artic, was used historically by the Soviet Union for many different types of nuclear weapon tests and nuclear effects tests. It was the site where USSR carried out the tests with higher yields. A total of 130 tests were carried out high in the atmosphere, at low levels above water, at the water/air interface, below water and underground. These 130 tests entailed 224 separate explosive devices, including by far the largest atmospheric and underground tests of the Soviet Union. The underwater, above-water and surface tests were performed between 1955 and 1962, the atmospheric tests between 1955 and 1962 and the underground tests between 1955 and 1990.

The high fission yields of most of the atmospheric tests induce the introduction of the majority of the generated debris in the stratosphere. However, also tropospheric fallout was disseminated by atmospheric testing at Novaya Zelmya, being recorded worldwide and specifically within the Soviet Union by the network of more than 500 stations equipped with standard instruments for monitoring of radionuclides. From observations made in 1958-1962 and reported daily to Moscow, two main trajectories of significant radioactive fallout were observed: a) directly to the south as far as the Caspian Sea, b) towards the sea of Okhotsk stretching several thousands of km to the south-east of Novaya Zelmya [22] On the other hand, direct atmospheric transport from Novaya Zelmya to Norway has been investigated at the northernmost Norwegian sampling

site, Vadso, located only 800 km west of Novaya Zelmya being no clear evidence of such transport [23]. In the same way, transport to finish Lapland could not be detected [24].

A little bit surprising is the very little information existing about the magnitude of the contamination associated to local fallout in Novaya Zelmya. Only information exists about a surface nuclear test of 32 kT carried out in the south of the island in 1957, which provokes a crater of 80 m diameter, 15 m depth and significant local contamination. And a second local terrestrial contaminated area was the result from fallout of an above water explosion performed in the Chernaya Bay in 1961. The two areas are still contaminated (100 Km^2) and access is prohibited, although no public data about the $^{240}Pu/^{239}Pu$ atom ratios are available.

High Pu concentrations (in relation to global fallout) were determined in sediment samples from south-west of the Kara Gate. A probable source of Pu at this site is underwater nuclear weapon tests carried out in Southern Novaya Zelmya. Sediments from the nearby Chernaya Bay are known to be highly contaminated (up to 15 Bq/g) and $^{240}Pu/^{239}Pu$ atom ratios of 0.030 have been reported indicating low yield tests. In Kara Gate $^{240}Pu/^{239}Pu$ atom ratios of 0.15 are reported, which is compatible with inputs up to 40% originated from a source having a $^{240}Pu/^{239}Pu$ atom ratio of 0.03 [25].

Due to the high yields of most of the tests performed in Novaya Zelmya could be theoretically expected to find areas with signals of these events characterized by high $^{240}Pu/^{239}Pu$ atom ratios. This is not the case. Even, debris captured in the lower stratosphere above Sweden in 1958 and originated in two large detonations above Nova Zelmya had a reported $^{240}Pu/^{239}Pu$ atom ratio only of 0.10.

Several authors have explained this discrepancy indicating that in contrast to the early USA thermonuclear detonations carried out in the Marshall Islands, the majority of the high yields USSR detonations were conducted without the presence of a fissionable natural or depleted U tamper. The fissionable tamper constitutes an important source of Pu isotopes trough single or multiple neutron captures in ^{238}U. Such detonations when conducted at sufficient height above ground, would not only produce less radioactive fallout, the production of Pu isotopes heavier than ^{239}Pu would also be lower, resulting in lower $^{240}Pu/^{239}Pu$ atom ratios {26].

The conclusions reflected in the previous paragraph, can explain the integrated value of 0.18 for the $^{240}Pu/^{239}Pu$ atom ratio corresponding to stratospheric (global) fallout. The stratospheric fallout disseminated over the world until the end of the 50s was dominated by the inputs of the high yields detonations conducted by USA and characterized by a $^{240}Pu/^{239}Pu$ atom ratio of 0.25 – 0.30. After that, the contribution in the 60s to the stratospheric fallout mostly due to the high yield detonations of USSR in Nova Zelmya, was characterized by a clearly lower $^{240}Pu/^{239}Pu$ atom ratio (about 0.10), giving as a result a total smoothed integrated $^{240}Pu/^{239}Pu$ atom ratio of 0.18, representative of the integral stratospheric global fallout nowadays.

Kasputin Yar

The KasputinYar, located in South-West Russia is a rocket launch from where between 1956 and 1961 were launched to the upper layers of the atmosphere a total of 11 nuclear weapons for testing. All the detonations were conducted at high altitude (30 km-80 km) and were provoked after travelling over the space to different parts of the former USSR (Volvograd, North and centre of Kazakhstan, Uzbekistan, etc). Due to the high-altitude detonations, no signals of local fallout were detected, being all the radioactive debris generated incorporated to the stratosphere.

High-altitude weapon detonations were provoked also over Nova Zelmya from weapons launched in the Kola Peninsula. These last detonations are included in the number of tests assigned to Nova Zelmya in a previous section.

Malden Island

The British Government, after the tests performed in the Australian tests sites (Maralinga, Emu and Montebello) and in part due to the social pressure, looked for remote places at the Pacific Ocean to start their tests involving hydrogen bombs. The first place elected was the Island of Malden, in the southern hemisphere, uninhabited, and joined administratively by UK and USA.

In this island, a total of 3 atmospheric nuclear weapon tests were performed in May-June 1957. These tests were air drops detonations conducted at 2000-2500 meters and with yields of 200, 300 and 720 kT. The altitude of the detonation avoid that the fireballs touch the ground, be disminished in this way considerably the local fallout. Nevertheless, tropospheric fallout is expected in the area, affecting the South Pacific Ocean.

No data about $^{240}Pu/^{239}Pu$ atom ratios associated to these events have been found in the open literature.

South Atlantic

The United States conducted also in 1958 a total of three high-altitude nuclear weapon tests at the South Atlantic, 1500 km south of Cape Town in South-Africa. The three detonations were low yield (less than 2 KT each), and no impact in the local environment is expected.

These tests were performed because high-altitude nuclear detonations would create a radiation belt in the extreme

upper regions of the Earth's atmosphere that can have possible tactical use in war, including degradation of radio and radar transmissions and damage or destruction of the arming and fuzing mechanisms of some warheads.

Christmas Island

In this island, located in the vicinity of the Malden Island, and also joined administratively by UK and USA, two different nuclear weapon tests series have been historically performed. A first series formed by 6 tests were conducted in 1958 by UK, and afterwards, in 1962, a second series formed by 24 detonations was conducted by USA.

The 6 tests conducted by UK were carried out from November 1957 until September 1958. Two of them were balloon detonations with low-yield (24 and 25 kT) while the other four have higher yield (0.8, 1.0, 1.8 and 3.0 MT) and were air drop detonations.

The balloon detonations were performed at a height of 450 m, while the air-drop were carried out at near 3000 m. For that reason, it is expected limited local fallout due to no strong interaction with the ground surface.

The 24 tests conducted by USA in this island were carried out along 1962 and account for a total yield detonation of 30 MT. 10 test were balloon and 14 air drops detonations, being performed all of them at heights higher than 1000 m. No surface detonations were performed. From the 24 tests, 9 have yields lower than 100 kT, 8 between 100 kT and 1 MT, and 7 high than 1 MT. The higher yield corresponds to an air drop detonation of 7.7 MT.

No data about local fallout contamination in the island have been found in the open literature. Consequently, no $^{240}Pu/^{239}Pu$ atom ratio data are available.

Jhonston Island

Also in the Pacific, in the Jhonston Island, the United States conducted a total of 12 atmospheric tests in the interval 1958-1962, following the detonations performed in the Marshall Islands. 5 of these tests were air-drops detonations being performed the majority of them at 4000 m, while the other 7 corresponds to high-altitude nuclear explosions, being conducted in the outer space at 30-80 km after transportation in rockets. These 12 detonations contributed appreciably to the global stratospheric fallout, being not expected elevated levels in local fallout due to their characteristics.

In addition to the 12 commented tests, in the Jhonston atoll four failures occurs, all of which were deliberately disrupted by range safety officers when the missiles' systems failed during launch. Three of these failures, caused serious contamination to the island and surrounding areas with weapons-grade plutonium and americium that remains an issue to this day.

In 1962 Plutonium pollution following the three failed nuclear missile launches was heaviest near the launch emplacement. This contaminated launch site was stripped, the debris gathered and buried in the island's 1962 expansion. In addition, a comprehensive radiological survey was completed in 1980 to record transuranic contamination remaining from the 1962. Unfortunately, no data have been found in the open literature about the remaining contamination in the area.

Algeria

In the early 60s, between 1960 and 1966, France conducted a series of nuclear weapon test in three sites in South Algeria. In the first site, called Reganne (dessert area, 1500 km south of Algiers) 4 low-yield atmospheric nuclear weapon test were performed: three on a tower and the four on the ground. In a survey performed in the 90s only some remaining contamination was detected in the firing site where the surface test was carried out. In this location, most of the contamination was found in black, vitreous and porous material (sand melted at the time of the explosion and then solidified). This type of contamination has been also found in places like in Semipalatinsk Ground Zero and in Trinity. No data exist about the $^{240}Pu/^{239}Pu$ atom ratio, although the type and origin of the contamination induce to think that contaminated Pu will be weapon-grade plutonium.

In a second place, called Taourit tan Afella, a total of 13 underground tests were conducted in tunnels. No signals of contamination in most firing sites were found because the contamination was confined. Only in one test, 5-10% of the debris escapes as lava and aerosols. This lava can be found nowadays in the place of formation with high radionuclide content but with a quite refractory behaviour. No radionuclide leaching, and consequently no contamination of waters, exists in the area.

Finally in a third place called Addar Tikertine, safety experiments on the dispersion of plutonium in air (called “pollen” experiments) were conducted. In these pollen experiments were simulated accidents involving plutonium dispersion (pyrotechnical dispersal of 20-200g of Pu), analysing its consequences. After each “pollen” experiment, the most contaminated area was covered by asphalt to limit/avoid the resuspension of plutonium. In a survey performed in the 90s in this area no Pu signals were found. Due to the nature of the “pollen” experiments, it would be expected the dispersion of some active particles in the area, with the larger and heavier ones settling closest to the point of origin of the dispersion experiment. However, no evidence of such particles was found in the survey conducted at the 90s. The particles have been

probably widely dispersed by the wind in the intervening years [27].

Lop Nor

China has conducted a total of 45 nuclear weapon tests in one test site: Lop Nor, which is a former salt lake now largely dried-up and located in the north-western part of China. From the 45 tests, a total of 21 were atmospheric, conducted mostly at towers and air drops. The 21 atmospheric nuclear tests were conducted between 1964 and 1980, being the first eight ^{235}U devices. The first plutonium weapon was tested in 1968.

In all the atmospheric tests a total of 25 MT yield was involved with the maximum yield individual detonation being 4MT. 6 of the 21 detonations have associated more than 90% of the yield explosions.

The higher-yield explosions have introduced radionuclide debris in the stratosphere. In fact, a high $^{240}Pu/^{239}Pu$ atom ratio of 0.224 was found in some radioactive debris collected at 10 km height in the atmosphere after the largest test of 4 MT conducted in 1976. This high $^{240}Pu/^{239}Pu$ atom ratio of 0.22 was observed in 1976 in grass samples from the Rothemsted archive (Great Britain) and the source of plutonium that was suggested was the mentioned Chinese test.

In addition to stratospheric fallout, low and medium yield Chinese detonations have produced tropospheric fallout, dispersed and deposited regionally. In soils and freshwaters lake sediments downwind the test site, has been measured $^{240}Pu/^{239}Pu$ atom ratios lower than 0.18, because in the tropospheric fallout the Pu isotopic composition has changed only slightly in relation with the found in weapon-grade unburned fuel. In sediments collected from freshwater lakes collected around the Test site, it was possible to observe $^{240}Pu/^{239}Pu$ atom ratios as low as 0.08, while in soils in the neighbouring Junquian region atom ratios from 0.06 until 0.19 were measured [28]. On the contrary, in studies performed analysing soils in Central China, it was possible to conclude the negligible close-in fallout contribution from the test site located at 2500 km. The $^{240}Pu/^{239}Pu$ atom ratio ranges from 0.17 to 0.21, with an average value of 0.186 [29].

Anecdotally, we can indicate that studying sediment cores collected in lake Qinghai, it was possible to observe low $^{240}Pu/^{239}Pu$ atom ratios in layers older than 1964. In layers formed in the 50s, ratios in the range 0.04-0.16 were found that cannot be attributed to the Lop Nor tests that were conducted in the 60s. In the surroundings of the lake and in the 50s was constructed a nuclear research centre where the first atomic and hydrogen Chinese nuclear weapons were designed and developed. It is suspected that the low Pu ratios found in the layers formed in the 50s corresponds to uncontrolled waste discharges conducted by the research centre in the watershed [30]).

Mururoa and Fangataufa

The atoll of Mururoa and its sister Fangataufa were the sites in French Polynesia where from 1966 until 1974 atmospheric nuclear weapon tests and several safety tests were conducted by the French authorities. The atolls are located in the central part of the South Pacific Ocean.

A total of 41 atmospheric tests were carried mostly in the atoll of Mururoa and additionally several safety tests that produced variable, in magnitude, local contamination. The three tests conducted in barges above the Mururoa lagoon, a few meters above the surface provokes the deposition of fissile material and fission products in the lagoon bottom sediments, while the safety tests performed in the north of the atoll disseminate also contamination in the lagoon and in the terrestrial soils of the islands forming the atoll. In the case of Fangataufa atoll the great majority of the contamination was due to a barge test conducted in 1966. The majority of tests were performed at relatively high altitude suspended in balloons.

The debris disseminated in both atolls has characteristics $^{240}Pu/^{239}Pu$ atom ratios. In Mururoa the safety tests in the north yielded $^{240}Pu/^{239}Pu$ atom ratios lower than 0.03, while the barge tests yield atom ratios of 0.035 in the centre of the lagoon and 0.05 in the south. In the case of Fangataufa the ratio found is 0.05 and can be considered uniform over the atoll because the origin is unique [31].

The atmospheric nuclear tests can be classified in Mururoa in the majority of cases as low yield detonations, with some few exceptions where the yield reach even 3 MT. It can be expected in addition to the local fallout the tropospheric dissemination of radioactive debris affecting regional areas in South Pacific. In fact, determinations performed analysing soils along the coast of Chile shows $^{240}Pu/^{239}Pu$ atom ratios lower than the expected average worldwide fallout value of 0.18, indicating the presence of Pu released in the tests performed in Mururoa. Atom ratios in the range 0.05 – 0.18 were found with an average of 0.15. The majority of the tests at Mururoa were conducted hundreds of meters above sea level, so the corresponding fallout, mostly fine-size particles, may have travelled thousands of kilometres in the troposphere reaching South-America [32].

Palomares

Broken arrow is a euphemism that describes an accidental event associated with a nuclear weapon, warhead, or component that may involve a non-nuclear explosion or fire resulting in the spread of radioactive contamination on the environment. The release of nuclear and radiological material into the environment creates a potential health risk.

One of the well-known Broken arrow incident is the known as the Palomares accident.

On 17 January 1966, an accident during a fuel-feeding operation above the small village of Palomares (Cuevas de Almazora, Almeria, Spain) resulted in the destruction of a US Air Force refueling KC-135 tanker and a B-52 bomber carrying four thermonuclear weapons. Two of these bombs were mechanically destroyed with some burning and release of the nuclear fuel at impact on the ground. The fuel was disseminated locally in an area of about 2 km^2 and although the affected area was immediately cleaned up as a remedial action, some contamination of the surface layer soils with plutonium still remains nowadays.

The Pu contaminating the area is weapon-grade Pu, in correspondence with the content of the involved weapons (the fuel of the two bombs was a mixture of weapon-grade plutonium and enriched uranium). AMS determinations in heavily contaminated soils from the area show an average $^{240}Pu/^{239}Pu$ atom ratio of 0.064 [33].

One peculiarity of the contamination generated by the accident is the presence of the contamination mostly in particulate form. Micrometer particles with a quite refractory behavior contain the great majority of the remaining contamination in the area. Some of these particles have been isolated and fully characterized by applying solid-speciation techniques [34].

In the analysis of an isolated hot particle, the $^{240}Pu/^{239}Pu$ atom ratio was determined as 0.060 and 0.068 by SIMS, values compatible with the found ones in heavily contaminated soils. There are quite uniform values. Until now, no evidences of significative differences between the two affected bombs have been found. Only one group for the $^{240}Pu/^{239}Pu$ atom ratio can be assigned to the remaining contamination in spite of the fact that two different weapons were involved in the accident dispersing their fuel [35].

$^{240}Pu/^{239}Pu$ activity ratios have been also determined in soils contaminated by Palomares after $^{239+240}Pu$ analysis by alpha-particle spectrometry prior purification on anion-exchange resins and alpha-spectra deconvolution. The obtained values were coherent with the obtained ones by other more sophisticated techniques [36].

On the other hand, it has been proved that a fraction of the Palomares terrestrial contamination has been transported to the nearby seacoast via mainly river flooding and also by airborne relocation. One sediment core collected in the Aguas river submarine canyon shows enhanced Pu values indicating the impact of the Palomares accident. The sediment core has 20 cm, and the $^{240}Pu/^{239}Pu$ atom ratio profile evidences the influence of the Palomares with values in the upper 18 cm lower than 0.13. The lowest ratios are found in the first 9 cm of the core suggesting that 47% to 95% of the total plutonium activity originated from the Palomares accident, whereas from 9 to 18 cm depth the influence is continuously decreasing until 0% Palomares contribution [37]. This marine influence has been also evidenced by the values obtained by the $^{240}Pu/^{239}Pu$ atom ratio measured in some seaweed samples collected in the coast located in the vicinity of the contaminated terrestrial zone.

Thule

In January 1968, a B-52 airplane crashed on the sea ice about 12 km west of the Thule air base, NW Greenland. The aircraft was carrying four hydrogen nuclear weapons. The impact triggered the conventional explosives within the weapons and the explosion pulverized the fissile material in the bombs. The debris was scattered around the point of impact and some square kilometers of the ice was contaminated. The benthic marine environment received the fraction of the weapon material that was not recovered from the ice during the cleanup operation following the accident and probably also a fraction injected during the accident, as the impact caused part of the ice to break-up. In addition, a small terrestrial area, covered by ice and snow at the moment of the accident was affected [38].

The radioactive contamination generated by the Thule incident has several characteristics totally similar to the found ones in the Palomares accidents. The contamination is formed by a mixture of enriched uranium and weapon-grade plutonium, components of the weapons involved in the accident. In addition, most of the contamination is in the form of quite refractory particles that forty years after the accident shows the same characteristics that the Palomares particles in spite of the very different scenario in which were disseminated (artic vs semi-dessertic ecosystems) [34].

For the characterization of the $^{240}Pu/^{239}Pu$ atom ratio a total of five hot particles and several heavy contaminated sediment samples were initially analysed. In the five hot particles the obtained $^{240}Pu/^{239}Pu$ atom ratio was 0.055 + 0.002, while the results from the sediment samples showed the presence of more than one Pu source involved in the accident. The $^{240}Pu/^{239}Pu$ atom ratio was divided into two Pu-isotopic ratios: one with identical ratios as the five hot particles and the other group with a value of 0.03 [39]. The two sources are characterized by low ratios but shows different degree of weapon grade purification

In an independent study were analysed 3 additional Thule hot particles all showing equal $^{240}Pu/^{239}Pu$ atom ratios of 0.055 [34] but in another work, additional eight isolated Thule hot particles were studied by SIMS, being found four different $^{240}Pu/^{239}Pu$ atom ratios. Five of them as in previous studies show a Pu ratio of 0.058 and the other 3

had the Pu ratios of 0.042, 0.036 and 0.028. Conclusion: 4 groups are found with the majority having a Pu ratio of 0.058 [40]. It is clear to conclude that the Thule weapons involved in the accident contains Pu with different weapon-grade degree. These Pu ratios are, on other hand, in all the cases lower than the determined one in the similar Palomares incident.

Bomarc

The characteristics of particles released as a result of the two previously well-known broken arrow incidents, Palomares, Spain and Thule, Greenland, have been well documented in the open literature. In opposition, a third broken arrow incident, the BOMARC incident, in which uranium and plutonium particles were disseminated to the environment, has not been investigated as thoroughly [41].

In 1960, a military accident occurred at McGuire Air Force Base in New Yersey (USA) when a Boeing Michigan aeronautical research centre (BOMARC) missile caught fire and the warhead was partially melted by the fire. While the warhead did not explode, heat from the fire and fire-suppression activities together with the turbulent local atmospheric conditions during the fire contributed to the dispersion of weapons grade plutonium (WGP) into the local environment. As a result, a significant quantity of Pu particles with varying sizes (submicrometer-sized hot particles or single large particles) containing $^{239+240}$Pu and ^{241}Am were inhomogeneously dispersed.

A total area of 7 acres was contaminated, being this zone immediately remediated by the removal of the contaminated soil. Although the cleaning was exhaustive, still some contamination remains in the affected area.

The atom ratio of ^{240}Pu/^{239}Pu in the BOMARC soil still contaminated was remarkably lower than the fallout value. The atomic ratio of ^{240}Pu/^{239}Pu was very close to the value of the weapons-grade Pu detected for the Thule accident. In fact, determinations performed by high-resolution ICP-MS gives Pu atom ratios in the range 0.056- 0.059, and quite uniform with grain size: 75- 150 μm 0.057 ; 150-250 μm 0.059 ; 250-350 μm 0.057 ; 350-425 μm 0.056 and 450-600 μm 0.059 [42].

3 Conclusions

In this paper a compilation of the great majority of nuclear weapon events generating historically the dissemination of nuclear debris in the environment have been detailed. These events can be classified in two groups: nuclear weapon tests and broken arrows incidents. Special emphasis has been put in to show associated to each event the information available in the literature about the values of the ^{240}Pu/^{239}Pu atom ratio, which acts as a fingerprint of the type, magnitude and characteristics of the incident analyzed. In the compilation conducted there are some nuclear tests series where the author has not found in the open literature representative ^{240}Pu/^{239}Pu atom ratios. It is a challenge for the scientific community to cover these gaps, in some cases by doing available unpublished and/or no public information, and in other by performing research in the not public explored areas.

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