Long-range tropospheric transport of uranium and plutonium weapons fallout from Semipalatinsk nuclear test site to Norway

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Abstract

A combination of state-of-the-art isotopic fingerprinting techniques and atmospheric transport modelling using real-time historical meteorological data have been used to demonstrate direct tropospheric transport of radioactive debris from specific nuclear detonations at the Semipalatinsk test site in Kazakhstan to Norway via large areas of Europe. A selection of archived air filters collected at ground level at 9 stations in Norway during the most intensive atmospheric nuclear weapon testing periods (1957 – 1958 and 1961 – 1962) have been screened for radioactive particles and analysed with respect to the concentrations and atom ratios of plutonium (Pu) and uranium (U) using accelerator mass spectrometry (AMS). Digital autoradiography screening demonstrated the presence of radioactive particles in the filters. Concentrations of ^{236}U (1.0 – 22.6 nBq m $^{-3}$) and $^{239+240}$Pu (0.7 – 782 µBq m^{-3}) as well as the atom ratios ^{240}Pu/^{239}Pu (0.052 – 0.237) and ^{236}U/^{239}Pu (0.019 – 0.68) varied widely indicating several different sources. Filter samples from autumn and winter tended to have lower atom ratios than those sampled in spring and summer, and this likely reflects a tropospheric influence in months with little stratospheric fallout. Very high ^{236}U, $^{239+240}$Pu and gross beta activity concentrations as well as low ^{240}Pu/^{239}Pu (0.052 – 0.077), ^{241}Pu/^{239}Pu (0.00025 – 0.00062) and ^{236}U/^{239}Pu (0.0193 – 0.046) atom ratios, characteristic of close-in and tropospheric fallout, were observed in filters collected at all stations in Nov 1962, 7 – 12 days after three low-yield detonations at Semipalatinsk (Kazakhstan). Atmospheric transport modelling (NOAA HYSPLIT_4) using real-time meteorological data confirmed that long range transport of radionuclides, and possibly radioactive particles, from Semipalatinsk to

Norway during this period was plausible. The present work shows that direct tropospheric transport of fallout from atmospheric nuclear detonations periodically may have had much larger influence on radionuclide air concentrations and deposition than previously anticipated.

Key Words: Atmospheric dispersion modelling; Source identification; Plutonium; Uranium-236; Atom ratio

1. Introduction

The primary sources of anthropogenic radionuclides in the environment are the atmospheric nuclear tests that were carried out during the period 1945 to 1980. A total of 543 atmospheric detonations were conducted worldwide, with a total yield of 440 Mt (megatons TNT equivalents) distributed as local (~15%), tropospheric (~8.5%) and stratospheric (~76%) fallout (UNSCEAR 2000a). The apportionment of debris into the different atmospheric compartments depended primarily on the yield of the detonation, the detonation height and the latitude at which the tests took place. Debris from low-yield detonations remained almost completely in the troposphere and deposited downwind of the detonations, 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 on the surface of the hemisphere on which the detonation took place.

The most intensive period was 1951 – 1962 interrupted by a moratorium between the United States, the Soviet Union and the United Kingdom banning testing between November 1958 and September 1961. During the period 1951 – 1958, 252 tests were conducted, with a total yield of 152 Mt and a calculated distribution of 35 %, 9 % and 56 % between local, tropospheric and stratospheric compartments, respectively. From 1960 to 1962, 180 tests were conducted with a total yield of 257 Mt and a distribution of 0.1 %, 7 % and 93 % between the abovementioned compartments (UNSCEAR 2000a).

Four factors impact on the Pu and U fallout signal from a nuclear weapon: the fissile material, the type of device, the tamper, and the fission yield. Nuclear weapons material is assumed to consist in general of high purity ^{235}U and / or ^{239}Pu (Bukharin 1998; Choppin et al. 2002; Forsberg et al. 1998). The $^{240}Pu/^{239}Pu$ atom ratio is primarily affected by the yield of the detonation, where a higher yield gives higher atom ratios. There is an important difference between nuclear weapons and reactors in the way heavier isotopes are formed. In UO_2 reactors Pu isotopes are formed from neutron irradiation of ^{238}U yielding ^{239}U followed by double beta decay to ^{239}Pu. Heavier isotopes are formed from continued neutron capture in the ^{239}Pu, and the proportion of heavier Pu isotopes formed increase with irradiation time and neutron flux (burn up). Since the half-life of ^{243}Pu (4.956 h) is short, the production of ^{244}Pu is insignificant in reactors (Bodansky 2004; Winkler 2007). In thermonuclear devices on the other hand, irradiation times are short and neutron fluxes are high, and U-isotopes up to ^{255}U are generated before beta decay (Diamond et al. 1960). Thus, the presence of ^{244}Pu above the environmental background can be regarded as a clear signal of debris from thermonuclear detonations. An overview of the abundances of ^{236}U, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{244}Pu relative to ^{239}Pu associated with different sources are summarized in table 1.

During the period 1957 – 1982 the Norwegian Defence Research Establishment (FFI) measured gross beta activities in ground level air on a daily basis. There were 2 - 11 air filter stations in operation nationwide (Njølstad 2006; Sæbø et al. 1998). While the collected gross beta data show clear indications of influences from nuclear weapons tests, they do not provide information on the specific source of the fallout.

Tropospherically transported debris from atmospheric nuclear detonations has been assumed to follow the prevailing wind directions at the test site latitude, and air concentrations and deposition to be strongest downwind of the test site. Deposition of debris at areas distant from the test sites is thus assumed to be dominated by stratospheric fallout. The prevailing wind

direction at the Semipalatinsk test site in Kazakhstan is westerly carrying debris westwards over Asia and USA before reaching Western Europe. In addition it is known that Former Soviet Union atmospheric nuclear tests were carried out under wind directions that would lead the debris cloud over Former Soviet Union territories (Khalturin *et al.*, 2005). The long transit caused by the eastward route would cause dilution of the debris, and substantial sedimentation of large refractory particles. However, particles from detonations at both Semipalatinsk, Novaya Zemlya and the Chinese test site Lop Nor has been detected in Sweden, e.g. Sisefsky (1961), Sisefsky (1964), Sisefsky (1967). We here hypothesize that tropospheric direct transport of debris from Former Soviet Union test sites Novaya Zemlya and Semipalatinsk to Western Europe and Norway has occurred on several occasions. Such transport would be recognized by characteristic perturbations in the $^{240}Pu/^{239}Pu$ atom ratios reflecting the yield of the detonations carried out at the respective test sites. The aim of this study has been to determine $^{240}Pu/^{239}Pu$ and $^{236}U/^{239}Pu$ atom ratios in debris from atmospheric nuclear detonations captured in surveillance air filters from the period 1957 – 1963 by accelerator mass spectrometry (AMS). Furthermore, to use the obtained results in combination with atmospheric dispersion modelling, NOAA HYSPLIT and NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder Colorado (www.esrl.noaa.gov/psd/), to determine the main sources of radioactive debris in ground level air in Western Europe in this time period.

2. Materials and methods

2.1. Archive air filters

The Norwegian air monitoring programme covered the time period of 1956 – 1982. During this period air filters were sampled daily at 2 – 10 stations (Fig. 1) (Njølstad 2006). At the air filter stations, 430 m^3 of air was pumped through cellulose asbestos filters (Draeger,

Draegerwerk, Lübeck Germany) over a period of 24 hrs (Small 1959). The filters were specified to retain more than 99.9 % of particles larges than 0.3 µm at the flow rates used (Lockhart et al. 1964). The air intake of the filter stations were positioned 2 m above the ground, and the filters were changed every 24 hours. After sampling, the filters were sent for analysis at a central lab, and the gross beta activity was determined 48 – 72 hrs after the collection (Bergan personal communication; Hvinden 1958).

Air filters from 1957 and onwards were available for this work. The air filters selected were collected from the time before and after the moratorium, from 9 October 1957 (Sola) through 8 June 1963 (Røros). The samples were collected at seven coastal locations (Bergen, Sola, Ålesund, Værnes, Bodø, Tromsø and Vadsø) and two inland locations (Gardermoen and Røros). Vadsø was the northernmost sampling site, situated some 850 km southwest of Chernaya Bay, which was the southernmost testing site for nuclear devices at Novaya Zemlya. Air sampling performed in this way offers an exact geographical location and unique time resolution that is ideal for the purpose of this work.

Selection of filters

Filters were primarily chosen from three locations:

- Bergen representing a southern coastal site,
- Røros representing an inland site and
- Vadsø for its proximity to Novaya Zemlya and being the northernmost site.

In addition, filters from the other sites (Fig. 1) were chosen based on high gross beta activities. An episode with particularly high gross beta activities occurred in November 1962. It has previously been assumed that the high activities registered in this episode originated from a detonation at Novaya Zemlya in October / November 1962 (Hvinden et al. 1964).

Hence, filters from all stations during this episode were selected for analysis. Finally, four filters from 1957 (October) and three from 1958 (June) with relatively high gross beta activities were chosen to represent the activity concentrations and atom ratios associated with pre-moratorium tests.

2.2. Autoradiography

In order to identify radioactive heterogeneities on air filters, archived filters were subjected to digital autoradiography (Molecular Dynamics storage phosphor screen). The storage phosphor screens were exposed to filters for 30 days in a (low activity) lead chamber, whereupon the plates were scanned within 1 hour with a Typhoon 8600 digital image scanner (resolution 100 µm).

2.3. Sample dissolution and radiochemical separations

Unless stated otherwise all chemicals used were of analytical grade. Ultra-pure HNO_3 and HCl were produced in the laboratory using a sub-boiling distillation unit (Milestone SubPure), and used for digestion, ion separation and final sample preparation. Deionised water (18Ω) was obtained from a MilliQ apparatus (Barnstead B-pure).

Approximately 0.4 – 0.7 g of each filter (corresponding to 1/4 – 1/2 of each filter) was weighed directly into PTFE ultraclave tubes, and for most samples high-purity tracers (20 pg ^{233}U and 17 pg ^{242}Pu) were added as yield monitors prior to digestion. Three samples were prepared with filter material from ten consecutive days at filter stations Bergen (1957) and Røros (1962 and 1963). These samples were prepared without yield monitor for determination of $^{242}Pu/^{239}Pu$ and $^{244}Pu/^{239}Pu$ atom ratios. All samples were digested under high pressure and temperature in nitric acid (conc.) in an ultraclave (UltraCLAVE 3, Milestone Ltd). After digestion of the samples the acid concentrations were diluted to 8 M and actinides were

separated by anion exchange (DOWEX 1x8) (Clacher 1995). The U eluate was further purified by UTEVA extraction chromatography according to the procedure suggested by Wilcken (2006). After separation the eluates were evaporated to dryness, taken up in 2 ml HNO_3 (conc.) with 2 mg Fe as $Fe(NO_3)_3$ and evaporated to dryness again before being ashed at 500 °C to give final preparates for AMS analysis of Pu and U.

2.4. Determination of Pu and U

Concentration and atom ratios of U and Pu were determined by accelerator mass spectrometry using the 14 UD pelletron tandem accelerator at the Australian National University (ANU), Canberra as described in detail by Fifield (2008).

2.4.1. Plutonium analysis

Three plutonium isotopes (^{242}Pu (yield monitor), ^{240}Pu and ^{239}Pu or ^{241}Pu and ^{239}Pu) were counted sequentially with counting times ranging from one to three minutes according to their abundance. The sequence was repeated twice with a third ^{242}Pu count at the end. Concentrations of ^{239}Pu, ^{240}Pu and ^{241}Pu were calculated from the measured ^{239}Pu/^{242}Pu, ^{240}Pu/^{242}Pu and ^{241}Pu/^{242}Pu atom ratios. For the samples without added yield monitor, Pu isotopes ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{244}Pu were measured in sequence, with counting times of three and five minutes being used for the least abundant isotopes ^{241}Pu and ^{244}Pu, respectively.

Analytical blanks spiked with 17 pg ^{242}Pu gave less than 4 and 2 counts of ^{239}Pu and ^{240}Pu for 2 and 3 minutes counting times, respectively. With count rates for ^{242}Pu (yield monitor) of 8000 counts in 1 minute this corresponds to procedural detection limits of 4.2 and 1.3 fg for ^{239}Pu and ^{240}Pu respectively. Blank calculations for ^{239}Pu and ^{240}Pu were based on both chemical blanks and air filters through which very little air had passed due to technical

difficulties. Even though it cannot be ruled out that the blank filters did capture a little fallout during their short operating time, they showed very low Pu concentrations and were assumed to be representative of the upper limit of the background levels of Pu and U in the air filters. Blanks without yield monitor were run for ^{241}Pu, ^{242}Pu and ^{244}Pu. A count rate corresponding to 0, 0.48 and 0.1 counts per minutes were measured for the three isotopes respectively. A certified reference material for Pu atom ratios (UKAEA No. UK Pu 5/92138) was measured repeatedly in each run. The ^{242}Pu/^{239}Pu atom ratio precision for the reference material was 2.7 % (standard deviation, n=13).

2.4.2. Uranium analysis

Three U isotopes (^{233}U, ^{234}U and ^{236}U) were measured sequentially similarly to the Pu isotopes and the concentrations of ^{234}U and ^{236}U were calculated based on the measured ^{234}U/^{233}U and ^{236}U/^{233}U atom ratios. Analytical blanks spiked with 20 pg ^{233}U gave 115 and 17.5 counts of ^{234}U and ^{236}U for counting times of 0.5 and 3 minutes respectively. With count rates of 5000 counts in 1 minute for ^{233}U (yield monitor) this corresponds to a detection limit of 24 fg ^{236}U and 0.9 pg of ^{234}U. Due to the higher blank levels in the U samples, it was necessary to apply a blank correction based on tracer count rates and blank levels of ^{234}U and ^{236}U.

2.5. HYSPLIT air transport modelling

The use of the HYSPLIT model in simulating dispersion of debris from nuclear detonations and accidents has been well documented (e.g. Draxler and Hess (1997), Draxler and Hess (1998), Kinser (2001), Moroz et al. (2010)). HYSPLIT does not simulate the dynamics of the debris cloud prior to stabilisation; therefore releases were simulated from the vertical axis of the stabilized mushroom cloud at several release heights following the work by Moroz et al.

(2010). Several particle classes and vertical velocities were incorporated into the model (Saltbones et al. 2003). The vertical extent of the stabilized mushroom cloud cap was estimated according to Peterson (1970). Concentrations of post detonation debris in the layers 0 – 75 m above ground level were simulated throughout the transport simulation.

3. Results and discussion

3.1. Gross beta activities

The mean gross beta activities (3 – 9 stations) obtained in air filters collected in the period 1956 – 1964 are given in Fig. 2. Furthermore, atmospheric detonations larger than 1 kt performed within the northern hemisphere are shown. The most important test sites in the northern hemisphere during the sampling period were the former Soviet Union test sites Novaya Zemlya and Semipalatinsk and the US test sites in the Pacific (Bikini, Eniwetok, Johnston Island and Christmas island), and Nevada. In addition, four small atmospheric nuclear detonations took place in the Reggane desert in Algeria (France). The largest detonations took place at Novaya Zemlya with a total yield of 255 Mt between 1957 and 1962. The total yield at Semipalatinsk was 6.6 Mt between 1949 and 1962. US testing sites in the Pacific had a total yield of 149 Mt between 1946 and 1962 in 94 underwater, surface and atmospheric bursts. Finally there were 101 detonations with a total yield of 1.1 Mt at the Nevada test site in the period 1951 – 1962 (Björklund and Goliath 2009; UNSCEAR 2000a).

The gross beta activities in air filters were generally higher in spring and early summer. After the passage of the spring maximum in 1959 (early May), the gross beta activities decreased rapidly and remained consistently low during the remaining moratorium period (1959 – 1962). Following the resumption of Former Soviet Union nuclear testing in 1961, a

substantial increase is observed 12 – 14 days later. An event with particularly high gross beta activity concentrations is seen in November 1962, and will be discussed in section 3.6.1.

3.2. Radioactive heterogeneities in filters-Autoradiography

Radioactive heterogeneities indicating the presence of particles were observed as hotspots in digital autoradiographs in filters from periods with atmospheric nuclear detonations. Digital autoradiographs of selected filters are shown in Fig 3. Even though the largest numbers of particles were indicated in filters from November 1962, associated with the highest gross beta activities, there was no general correlation between gross beta activity concentrations and the number of hotspots. The lack of correlation between the presence of particles and Pu and U concentrations is corroborated by the fact that particle indications are absent or at best ambiguous in filters from the spring. The concentration of radionuclides is normally higher in the spring due to the arrival of the spring peak deposition. Since the absence of particles in debris from nuclear detonations is unlikely, it must be assumed that particles are either too small to be detected by digital autoradiography, or embedded too deeply in the air filter material. Considerable efforts were made to localize Pu or U containing particles by ESEM-XRMA and synchrotron; however, concentrations proved to be too low for detection by these methods.

3.3. Activity concentrations of Pu and ^{236}U in filters

The activity concentration of Pu, ^{236}U, the atom ratios and gross beta activities for all stations are presented in table 2 and supplementary materials Table S1. The activity concentrations of $^{239+240}$Pu and ^{236}U (μBq m^{-3} and nBq m^{-3}) have been derived from the atom ratios to facilitate comparison with other publications.

3.3.1. Plutonium activity concentrations

The $^{239+240}$Pu activities varied widely (1.3 – 782 Bq μm^{-3}) reflecting input of Pu from individual tests as well as stratospheric / tropospheric exchange which reaches its maximum in spring (Bennett 2002). The activity concentrations for $^{239+240}$Pu were found to increase from 1957 to 1958 (table 2). However, the filters from 1957 (October) and 1958 (June) are from different seasons. The 1958 filters were from a season associated with stratospheric / tropospheric exchange processes in which debris from the lower stratosphere is transferred to the troposphere. This increases the tropospheric inventory of radioactive debris independent of atmospheric testing. Testing in 1957 and 1958 took place at all former Soviet Union test sites, Christmas Island (UK) and US Pacific and continental sites. Large tests (>500 kt) took place at Christmas Island, Novaya Zemlya and Semipalatinsk. Since the air sampling in 1957 was performed in a period associated with little stratospheric / tropospheric exchange, Pu concentrations in these filters are likely to be primarily affected by tropospheric fallout. The sampling in 1958 on the other hand would have received Pu from both tropospheric and stratospheric fallout.

The lowest activity concentration were observed in filters from 1957 and 1961. Two orders of magnitude higher activity concentrations in early November 1962 were likely associated with tropospheric fallout from detonations in Semipalatinsk (Cf. section 3.6.1). Gross beta activities in filters increased relatively rapidly (14 days) after the resumption of testing at Semipalatinsk and Novaya Zemlya in September 1961. In contrast, Pu concentrations increased slowly, and a substantial and stable increase did not take place until January 1962 after the large former Soviet Union test series in September through November 1961. Pu activity concentrations in air continued to increase steadily towards the summer of 1963. The Pu concentrations were generally within the same range as previously reported for air during the same period (Harley 1980; Osborne 1963; Salminen and Paatero 2009).

3.3.2. Uranium-236 activity concentrations

Concentrations of ^{236}U were found to vary in the range 1 to 22.6 nBq m^{-3}. In contrast to $^{239+240}Pu$ the highest concentrations were found in June 1963 rather than in November 1962 (table 2 and Supplementary materials, Table S1). Minimum concentrations of ^{236}U were observed in the autumn of 1957 and 1961, periods associated with little stratospheric fallout.

3.4. Pu atom ratios

The $^{240}Pu/^{239}Pu$ atom ratios ranged widely from a minimum of 0.052 (Ålesund 11 November 1962) to 0.237 (Bergen 03 June 1963). The $^{240}Pu/^{239}Pu$ atom ratios were generally lower in autumn and winter samples (September through January) than in spring and summer samples (March through July); this applies in particular to periods with atmospheric nuclear testing. Although the time series are by no way complete, we believe that this reflects influence of tropospherically transported debris from the northern hemisphere test sites. Previously reported $^{240}Pu/^{239}Pu$ atom ratios associated with low-yield detonations at Novaya Zemlya, Semipalatinsk and Nevada test sites are low (0.03 – 0.085) (Hicks and Barr 1984; Smith et al. 1995; Smith et al. 2000; Yamamoto et al. 2004).

We believe that atom ratios obtained in our October 1957 samples (0.09 – 0.15) originate from two large detonations above Novaya Zemlya the 24.09 and 06.10.1957. We base this on the following argumentation. Debris collected in an air filter in the stratosphere in October 1958 originated from comparable detonations above Novaya Zemlya the 30 September 1958 (900 and 1200 kt) (Sisefsky (1961), and references therein). Thus, we infer that the $^{240}Pu/^{239}Pu$ atom ratio (0.101) obtained for the same filter by (Warneke 2002), is representative of debris from detonations of approximately 1 Mt. Accordingly, the $^{240}Pu/^{239}Pu$ atom ratios obtained for our October 1957 samples (0.09 – 0.15) should originate from

detonations of similar size. The Pu atom ratios of debris from two somewhat larger detonations that took place above Novaya Zemlya the 24 September, and 6 October 1957 (1600 and 2900 kt respectively), are expected to be similar to or somewhat higher than the ones associated with the above mentioned detonations. All other northern hemisphere detonations during August – October 1957 had considerably lower yields (<520 kt) and $^{240}Pu/^{239}Pu$ atom ratios in debris from these detonations should be significantly lower.

The three samples from June 1958 were found to have $^{240}Pu/^{239}Pu$ atom ratios (0.162 – 0.170) which are in accordance with previously published values in samples from the same period (Warneke et al. 2002). It is unlikely that tropospheric debris from the detonations in February and March 1958 should still reside in the troposphere at appreciable concentrations in the beginning of June. The values obtained are therefore likely to be representative of contemporary global fallout.

After the resumption of atmospheric testing in September 1961, the measured $^{240}Pu/^{239}Pu$ atom ratios were rather low (0.067 – 0.10), indicating strong influence of tropospherically transported debris. The ratios are generally much lower than the 0.198 observed by Warneke et al. (2002). However, the samples used by Warneke et al. (2002) were from an herbage archive harvested in June 1961, well before the resumption of atmospheric testing, thus representing stratospheric fallout. All atmospheric testing in the second half of 1961 was performed by the former Soviet Union; 26 and 28 tests were carried out over Novaya Zemlya and Semipalatinsk, respectively. By this time all former Soviet Union thermonuclear tests were carried out above Novaya Zemlya, and 6 detonations larger than 2500 kt were detonated above at this site in 1961. These detonations are likely to have produced debris with high $^{240}Pu/^{239}Pu$ atom ratios; however, this is not reflected in our results. According to Peterson (1970) the debris cloud from detonations of this size and at this latitude would rise to be completely injected into the polar stratosphere. Accordingly the debris would not be available

to the filters before it is brought down to tropospheric levels after a considerable stratospheric residence time (0.5 – 2 years). In periods with frequent atmospheric testing the debris composition in air would be dominated by tests which inject the majority of their debris into the troposphere.

The samples from January through March 1962 generally exhibit lower $^{240}Pu/^{239}Pu$ atom ratios (0.127 – 0.156) than contemporary samples, e.g. Warneke et al. (2002). In 1962 there was no former Soviet Union atmospheric nuclear testing until August. There were numerous tests at the US Pacific test sites from March through July. However, tropospheric debris from these tests is likely to be retained at low latitudes due to atmospheric circulation patterns (UNSCEAR 2000a). It is therefore likely that the debris captured by these filters originate from the former Soviet Union tests in 1961. The lower $^{240}Pu/^{239}Pu$ atom ratios observed indicates that this debris was originally injected into the lower polar stratosphere by relatively large detonations (400 – 3000 kt). Debris from very large detonations (<5000 kt) should produce substantially higher atom ratios, e.g. Diamond et al. (1960), Yamamoto et al. (1996). However, this debris would predominately be injected into the upper polar stratosphere. The residence time in the lower polar stratosphere (9 – 17 km a.s.l) is estimated to 5 months and in the upper polar stratosphere (17 – 50 km a.s.l.) the residence time is estimated to 2 years (Peterson 1970). Thus debris injected into the lower polar stratosphere should be expected to be deposited before the debris injected into the upper polar stratosphere. The first deposition of debris injected into the lower polar stratosphere should be expected the first spring after the detonations (Peterson 1970). The filters from March were chosen based on the peaks in gross beta activities indicating the arrival of the spring peak. Nevertheless, it cannot be ruled out that the maximum deposition of stratospheric debris occurs later in the summer. Salminen-Paatero et al. (2012) observed prolonged periods with high $^{240}Pu/^{239}Pu$ atom ratio debris in

Finland in June through December 1963. Filters from November 1962 were also analysed, however, these results are discussed in section 3.6.1.

Atmospheric nuclear testing by the former Soviet Union and the USA had effectively ceased by the end of December 1962 and no sources of direct tropospheric Pu were present. The $^{240}Pu/^{239}Pu$ atom ratios measured for the 1963-samples, after the cessation of nuclear testing (0.190 – 0.237) are in accordance with published values for the same period (Leifner and Chan 1997; Olivier et al. 2004; Salminen-Paatero et al. 2012; Warneke et al. 2002).

3.4.1. Heavier plutonium isotopes: *$^{241}Pu/^{239}Pu$, $^{242}Pu/^{239}Pu$ and $^{244}Pu/^{239}Pu$ atom ratios*

Atom ratios of $^{241}Pu/^{239}Pu$, $^{242}Pu/^{239}Pu$ and $^{244}Pu/^{239}Pu$ relative to $^{240}Pu/^{239}Pu$ obtained in this work are presented in table 2 and 4, Supplementary materials Table S1 and Fig. 4. The filters from November 1962 were chosen from an episode of particularly high gross beta activities in the air filters. Pooled air filter samples (10 filters from Bergen October 1957, Røros March 1962 and June 1963) were chosen from a period of high gross beta activities in October 1957, and the spring peaks of 1961 and 1962.

The pooled air filter samples from April 1962 had significantly lower atom ratios (0.145 ± 0.003, 0.00038 ± 0.00008 and 0.0013 ± 0.0001) than those published for global fallout (0.182 ± 0.005, 0.0011 ± 0.0009 and 0.0037 ± 0.0003) with respect to all three $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$ and $^{242}Pu/^{239}Pu$ atom ratios, respectively. Twenty-five detonations took place above Novaya Zemlya in 1961, 11 of these detonations had a yield exceeding 500 kt. Debris from these detonations could potentially enhance the Pu atom ratios in air in 1962; however, this is not reflected in the $^{240}Pu/^{239}Pu$ $^{241}Pu/^{239}Pu$ or $^{242}Pu/^{239}Pu$ atom ratios in the pooled filters from 1962. A possible explanation for this could be suppression of the signal by tropospheric sources with lower atom ratios as discussed in section 3.4. The Tsar Bomba detonated above Novaya Zemlya 31 October 1961 (50 Mt) would have had very high neutron

fluxes, potentially producing massive amounts of heavier Pu isotopes. However, it has been indicated that the device was downscaled from its original 100 Mt to ~50 Mt by replacing the ^{238}U tamper with lead (Lapp 1970). This would reduce the production of higher Pu-isotopes significantly.

The pooled filter sample from June 1963 is indistinguishable from global fallout with respect to the ^{242}Pu/^{239}Pu atom ratio, albeit slightly higher with respect to the ^{240}Pu/^{239}Pu ratio. This is reasonable since there was no atmospheric testing between December 1962 and the sampling in June 1963. Thus Northern hemisphere tropospheric Pu by this time should be dominated by stratospheric fallout from the large Soviet testing series in 1961 and 62. The ^{244}Pu/^{239}Pu atom ratios measured in this work $(1.7 \pm 0.5) \times 10^{-4}$ agrees well with the value (1.21×10^{-4}) reported by Winkler (2007) (cf. Fig. 4 and table 1). To the knowledge of the authors the present results are the first ^{244}Pu/^{239}Pu results published in air filters.

3.5. Uranium atom ratios

3.5.1. ^{236}U/^{238}U atom ratios

^{236}U/^{238}U atom ratios 0.0000034 – 0.00011 were calculated from the measured ^{236}U/^{234}U atom ratio assuming a ^{234}U/^{238}U natural ratio of 0.000055. As for the Pu atom ratios, the highest values were found in June 1963, in accordance with peak stratospheric deposition following the large atmospheric tests in 1961 and 1962. All values were substantially higher than values stated for global fallout by Sakaguchi et al. (2009), and more in accordance with results from soils contaminated by local fallout as reported by Ketterer et al. (2003).

3.5.2. ^{236}U/^{239}Pu atom ratios

Literature data on ^{236}U/^{239}Pu are scarce. Ratios measured in the present work ranged from 0.0188 – 0.7 with a mean value of 0.24 (table 2), this is within the range of ratios for global

fallout (Ketterer et al. 2003; Sakaguchi et al. 2009; Srncik et al. 2011). The highest $^{236}U/^{239}Pu$ ratios were found in the October 1957 and December 1961, which are also the periods with lowest concentration of both ^{236}U and ^{239}Pu in air. The lower ratios were associated with the episode of tropospheric deposition in November 1962 noted above for which $^{240}Pu/^{239}Pu$ ratios were similarly low.

3.6. Source identification

The high and accurate time resolution of the air filters permits identification of debris from test series, and in some cases individual tests. As mentioned above, there were three dominant test sites in the Northern hemisphere in the time period 1956 – 1962 (table 3). The dominant Northern hemisphere test site in this time period by far was Novaya Zemlya, followed by Semipalatinsk and Nevada test site. It has been indicated that fallout from low-yield tests at the Nevada test site in 1952 has reached England (Warneke et al. 2002), and that this might also affect other Northern hemisphere sites. However, the close relationship between atmospheric testing at the two Soviet testing sites and the gross beta activities in air in Norway (cf. Fig. 2) indicate that testing at Novaya Zemlya and Semipalatinsk were the major contributors to the radioactivity collected in these air filters.

3.6.1. Transport of tropospheric fallout from Semipalatinsk to Norway

In November 1962 gross beta activities at all air filter stations in Norway increased abruptly up to a factor of 200 of the average recorded for the entire year 1962. The highest activity was reached in Ålesund on 11 November 1962 with 15 Bq m^{-3} (table 4, cf. also fig 2). Increased activities were first observed at all stations in southern Norway; however, the peak activities arrived first at the mid-East (Værnes) and Northern (Bodø) stations. Peak concentrations occurred with a time lag of up to three days between the different stations. Pu concentrations

and atom ratios were determined in 11 filters from this episode. The $^{240}Pu/^{239}Pu$ atom ratios (0.052 – 0.07), $^{241}Pu/^{239}Pu$ (0.00025 – 0.00046) and $^{236}U/^{239}Pu$ (0.02 – 0.05) deviated from published global fallout values (cf. Fig. 4, table 4), but were generally in agreement with values published for soil (Beasley et al. 1998) and a radioactive particle (Lind 2006) from Ground Zero in Semipalatinsk.

Assuming a $^{240}Pu/^{239}Pu$ atom ratio of 0.0438 to be representative of bomb debris from Semipalatinsk (Beasley et al. 1998), and an atom ratio of 0.162 to be representative for global fallout in 1962 (Warneke et al. 2002), the following mixing equation can be set up to estimate the fraction of Pu derived from Semipalatinsk.

$$X=(R_{Sample}-R_{Global})/(R_{Semipalatinsk}-R_{Global})*100$$

where X is the relative (%) contribution from Semipalatinsk, and R_{Sample} is the atom ratio of the sample, $R_{Semipalatinsk}$ is the atom ratio measured at ground zero in Semipalatinsk (0.0438) and R_{Global} is the contemporary global fallout (0.162). The results and estimated Semipalatinsk contribution in the samples is given in table 4.

This high gross beta activity incident has previously been suggested by other authors to be associated with an atmospheric detonation at Novaya Zemlya on the 30 October – 01 November 1962 (Hvinden et al. 1964; Peirson and Cambray 1965). It appears that these authors did not possess a complete detonation record, and were unaware of testing activities in Semipalatinsk at the time. Winds over Kazakhstan in the period 30 October – 5 November 1962 were easterly or north-easterly. A high pressure area (1035 hPa) North of Kazakhstan, forced debris towards the Black and Baltic Seas where a variable low pressure (995 – 1015 hPa) forced southerly and later east-south-easterly winds across Scandinavia (Personal communication).

In Fig. 5 mean gross beta activities are plotted together with mean precipitation (mm water equivalent) at all air sampling stations. A similar plot can be made with temperatures, which starts to decrease after 6 November 1962 (not shown). The high gross beta activities of this specific incident seem to be associated with dry and cold air from continental Eastern Europe.

There were five northern hemisphere detonations in the time interval suggested by Hvinden *et al.* (1964) that could have generated the debris, two over Novaya Zemlya and three over Semipalatinsk. Debris from the high altitude detonation above the Kapustin Yar (Former Soviet Union) and Johnston Island (US) is unlikely to be present in surface air at this time. The detonations at Novaya Zemlya were fusion devices or boosted fission devices of relatively low-yield (240 – 280 kt). While we have not found any good correlation between fusion yield and $^{240}Pu/^{239}Pu$ atom ratios at low yields, it is reasonable to assume that both thermonuclear and / or boosted fission devices would produce higher atom ratios than those observed in the filters. The detonations over Semipalatinsk on the other hand were smaller, (1.2, 3 and 10 kt) and are more likely to produce the near weapon grade $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$ and $^{236}U/^{239}Pu$ atom ratios observed in the filters. The detonation height of the Novaya Zemlya detonations are unclear, UNSCEAR (2000a) and Björklund and Goliath (2009) list these as air burst, while Grønhaug (2001) questions whether the detonation height of one of the detonations could be low. The tests at Semipalatinsk on the other hand are listed as surface (30 October 1962) and air detonations (31 and 01 November 1962) respectively (Björklund and Goliath 2009; Grønhaug 2001; UNSCEAR 2000a).

3.6.2. HYSPLIT simulation

Air concentration patterns calculated in HYPSLIT indicated that debris from detonations in the period 29 October 1962 – 2 November 1962 over Semipalatinsk were moving towards the West affecting ground level air in Western Europe and Norway. Fig. 6 shows a simulation of

concentrations from a release over Semipalatinsk 31 October 1962 (10 kt bomb, height of burst 700 m and plume rise up to 8 km, Peterson (1970), and references therein. Debris from detonations at the Semipalatinsk testing site (50.1167 °N, 78.71667 °E, 1750 – 5000 m a.g.l.) in the period 30 October 1962 – 1 November 1962 could well explain the increasing activity concentrations seen in ground level air at all air filter stations in Norway. The simulated arrival of debris in eastern Norway the 7 November 1962 fits well with the first appearance of elevated gross beta activities at stations Gardermoen, Kjeller, Røros and Værnes. It is also in fine agreement with the calculations made by Hvinden et al. (1964). This indicates a transit time of debris from the Semipalatinsk test site to Norway of about 7 – 10 days under suitable weather conditions.

Debris from a simulated release above Semipalatinsk the 31 October 1962 first appears in the 0 – 75 m layer in Belorussia 5 November 62, thereafter moving towards Lithuania, Poland Latvia and Estonia. Sweden and southern Finland is reached the 6 November 62, and the plume reaches Eastern Norway in the evening the same day. By this time the plume has stretched to a band covering western Russia south of Lake Ladoga, Southern Finland, Northern Estonia, Central Sweden and South Norway with the strongest concentrations appearing over Finland / Estonia. The plume then moves Northwards with the areas of highest concentration moving eastwards crossing Sweden and Norway. The plume moves northwards to reach Svalbard and the Fram Strait by the 8 November 1962. A simulated release from 5000 – 8000 m above ground level from Semipalatinsk the 31 October 1962 gives a plume reaching England as described by Peirson and Cambray (1965). Simulations were also performed for detonations performed at Novaya Zemlya the 30 October 1962 and 01 November 1962 (not shown) demonstrating that direct transport of debris from these tests to Norway was not plausible.

The highest concentrations over Norway were measured in an area spanned by Røros and Gardermoen in East and Ålesund in West (cf. Fig. 1). Gross beta activities and $^{239+240}$Pu activity concentrations were well correlated (R^2=0.92, P=0.000075) during the incident, suggesting that these signals originate from the same source.

4. Conclusions

A series of ground level air filters from the period 1957 – 1963 have been examined. Activity concentrations $^{239+240}$Pu and ^{236}U as well as atom ratios (^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu, ^{242}Pu/^{239}Pu, ^{244}Pu/^{239}Pu and ^{236}U/^{239}Pu were determined. Results show substantial variations in both ^{240}Pu/^{239}Pu atom ratios (0.0517 – 0.237) and $^{239+240}$Pu activity concentrations (1.3 – 782 mBq m $^{-3}$). The ^{240}Pu/^{239}Pu atom ratios during and after periods of intensive testing were substantially lower than contemporary global fallout, indicating significant tropospheric fallout, most likely from USSR testing sites in Semipalatinsk or Novaya Zemlya. An episode with elevated ^{236}U (4.6 – 20.1 nBq m^{-3}) and $^{239+240}$Pu (11.7 – 782 µBq m $^{-3}$) activity concentrations, and atom ratios characteristic of the Semipalatinsk test site (^{236}U/^{239}Pu=0.0188 – 0.046, and ^{240}Pu/^{239}Pu=0.0517 – 0.077) were observed in November 1962. A HYSPLIT simulation using real time meteorological data of this episode shows direct tropospheric deposition of debris from a detonation at Semipalatinsk to have arrived from East with the highest impact in Mid-east – Mid west Norway. The fine time resolution of the air filters permitted day by day fluctuations in both atom ratio and concentrations of anthropogenic U and Pu to be seen.

Acknowledgements

The authors gratefully acknowledge the support provided by the Research Council of Norway (Project no. 421048). We are thankful to the Norwegian Defence Research Establishment (FFI) for giving us access to their collection of air filters and letting us (destructively) analyse some of them. We are grateful to Dag Roger Kristoffersen at the Norwegian meteorological institute for sharing his insight in the meteorological conditions of central Asia. We are indebted to Signe Dahl for kind assistance with the layout.

Captions:

Table 1. Atom ratios from various sources. Reference date: 01.01.2012.

Table 2. Mean, median, minimum and maximum values of $^{239+240}$Pu, ^{236}U activity concentrations and ^{240}Pu/^{239}Pu, ^{236}U/^{234}U and ^{236}U/^{239}Pu atom ratios for air filter samples, presented by year and for the entire period of 1957 – 1963.

Table 3. Northern hemisphere atmospheric nuclear testing by site and time period

Table 4. Results from measurements of samples from 9 November 1962 – 13 November 1962. Reference date: 01 January 2012. † ^{236}U/^{238}U atom ratios calculated from the measured ^{236}U/^{234}U and the ^{234}U/^{238}U atom ratio of natural uranium (0,000055). Literature values brought in for comparison, a – Kelley et al. (1999); b – Ketterer et al. (2007); C – Beasley et al. (1998).

Figure 1. The Air filter stations in Norway, 1956 – 1982, and the approximate location of the Novaya Zemlya test site C.

Figure 2. Mean gross beta activities (N=3 – 11 stations) and northern hemisphere detonations larger than 1 kt in the period 1956 – 1964. Gross beta activities from FFI and Bergan (Personal communication)), detonation data from Björklund and Goliath (2009). Sampling periods are indicated by black ellipses. North equatorial test sites are US test sites Bikini, Eniwetok, Johnston Island and Christmas Island.

Figure 3. Digital autoradiographs of selected filters from November 1962. Left hand side figure depicts the autoradiograph of two filters halves from Værnes and Ålesund the 11.11.1962, while the right hand side figure depicts a filter from Bodø the 13.11.1962. The filter areas inscribed in rectangles are shown below the figure at a higher magnification.

Figure 4. Atom ratios of Pu (^{241}Pu/^{239}Pu, ^{242}Pu/^{239}Pu and ^{244}Pu/^{239}Pu (vertical axes) plotted against ^{240}Pu/^{239}Pu (horizontal axis). Reference date 01 January 2012. The November 1962 samples were individually analysed, while the autumn and spring samples from Bergen and Røros in 1957, 1962 and 1963 respectively were based on pooled samples from each period and site. Literature data from * Beasley et al. (1998) based on measurements of soils at ground zero in Semipalatinsk; ** Kelley et al. (1999) soil samples collected worldwide; †

Winkler (2007) sediment profile from Lake Erie; ‡ Lachner et al. (2010), mixed high yield weapon debris from the Bikini atoll.

Figure 5 Mean gross beta activity concentrations (open circles black line) and mean precipitation (filled circles grey line) at all stations in October / November 1962. Precipitation data from www.eklima.no.

Figure 6. Summary of HYSPLIT simulations of the release of radionuclides from a detonation of yield 10 kt which took place over Semipalatinsk testing site 31 October 1962. Simulated release heights were between 1750 and 5000 m, and post detonation concentrations between 0 and 75 m above ground were simulated. The plume position and air concentrations of debris from 7 November 1962 with 12 h increments is shown in the figure. The release location (Semipalatinsk) is indicated as a star in the lower right hand side of each map.

Table 7. (Supplementary) Activity concentrations, atom ratios and atom / activity ratios of ^{234}U, ^{236}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{244}Pu all air filter samples analysed in the present work.

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135 Table 1

Source	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{236}U/^{239}Pu$	$^{244}Pu/^{239}Pu$
Undetonated weapons plutonium	0.01 – 0.07 [a]	-	-	-	-
Low-yield detonations U-based [b]	0.00015 – 0.053 [b]	$(0.2 – 2.3)\times10^{-4}$ [b]	-	-	-
Low-yield detonations Pu based [b]	0.01 – 0.08 [b]	$(0.2 – 6.7)\times10^{-4}$ [b]	-	-	-
Low-yield detonations GZ Semipalatinsk	0.0438 ± 0.0001 [c]	$(2.21 \pm 0.035)\times10^{-4}$ [c]	$(7.89 \pm 0.26)\times 10^{-5}$ [c]	0.0244 ± 0.001 [c]	-
Global fallout N. hemisphere	0.182 ± 0.005 [d]	$(1.14 \pm 0.85)\times 10^{-3}$ [d]	$(3.71 \pm 0.3)\times10^{-3}$ [d]	0.235 ± 0.014 [e]	1.44×10^{-4} [i]
Castle Bravo, Ivy Mike	0.32 – 0.367 [f]	$(2.27 \pm 0.029)\times10^{-3}$ [f]	0.019 ± 0.003 [f]	-	$(1.18 \pm 0.07) \times10^{-4}$ [f]
Bikini atoll Various	0.06 – 0.32 [g]	$(0.7 – 5.5)\times 10^{-3}$[g]	$(2.5 – 5.7)\times10^{-4}$ [g]	-	$(4.18 \pm 1.25) \times10^{-4}$ [g]
Reactor debris Chernobyl	0.43[h]	0.12[h]	0.047[h]	8.53[h]	-

a) Warneke *et al.* (2002), Rokop *et al.* (1995), Eriksson *et al.* (2008)
b) Hicks and Barr (1984), Hansen (1995), Oughton *et al.* (2000), Smith *et al.* (2000)
c) Beasley *et al.* (1998)
d) Kelley *et al.* (1999)
e) Sakaguchi *et al.* (2009)
f) Yamamoto *et al.* (1996), Diamond *et al.* (1960)
g) Lachner *et al.* (2010)
h) UNSCEAR (2000)
i) Based on Winkler (2007) results from Lake Erie, years 1963 and 1964

136 Table 2

Year		$^{240}Pu/^{239}Pu$	$^{236}U/^{239}Pu$	$^{239+240}Pu$ µBq m^{-3}	^{236}U nBq m^{-3}	$^{236}U/^{234}U$
1957 – 1963	Min	0.0517 ± 0.0009	0.0188 ± 0.0009	1.3 ± 0.1	0.17 ± 0.06	0.06 ± 0.01
	Max	0.237 ± 0.005	0.7 ± 0.2	782 ± 7	23 ± 4	2.0 ± 0.8
	Median	0.137	0.22	20.8	2.8	0.31
	Mean	0.13	0.24	83.9	5.5	0.54
		(n=55)	(n=13)	(n=52)	(n=16)	(n=14)
1957	Min	0.09 ± 0.02		2.2 ± 0.1		
	Max	0.15 ± 0.02		7.4 ± 0.2		
	Median	0.12		3.3		
	Mean	0.12	0.7 ± 0.2	4.0	1.7 ± 0.4	0.12 ± 0.03
		(n=5)	(n=1)	(n=4)	(n=1)	(n=1)
1958	Min	0.162 ± 0.007		22.1 ± 0.5		
	Max	0.170 ± 0.007		28.4 ± 0.6		
	Median	0.17		24.9		
	Mean	0.17	0.15 ± 0.03	25.1	2.2 ± 0.4	2.1 ± 0.8
		(n=3)	(n=1)	(n=3)	(n=1)	(n=1)
1961	Min	0.067 ± 0.006	0.12 ± 0.02	1.3 ± 0.1	0.17 ± 0.06	0.06 ± 0.01
	Max	0.15 ± 0.02	0.6 ± 0.1	16.3 ± 0.5	2.1 ± 0.4	0.5 ± 0.2
	Median	0.09	0.40	2.9	0.60	0.29
	Mean	0.1	0.40	5.2	0.90	0.29
		(n=11)	(n=3)	(n=11)	(n=6)	(n=4)
1962 excl.	Min	0.110 ± 0.007		2.25 ± 0.09		
November	Max	0.21 ± 0.01		36.0 ± 0.5		
episode	Median	0.14		21.4		
	Mean	0.14	0.24 ± 0.05	20.4	5 ± 1	0.20 ± 0.04
		(n=16)	(n=1)	(n=15)	(n=1)	(n=1)
1962	Min	0.052 ± 0.001	0.0188 ± 0.0009	11.7 ± 0.2	4.6 ± 0.2	0.20 ± 0.01
November	Max	0.077 ± 0.004	0.046 ± 0.002	782 ± 7	20.1 ± 0.6	0.34 ± 0.02
episode	Median	0.06	0.027	267	9.6	0.30
	Mean	0.06	0.030	323	11.1	0.29
		(n=9)	(n=4)	(n=11)	(n=4)	(n=4)
1963	Min	0.190 ± 0.006	0.22 ± 0.04	18.2 ± 0.4	3.5 ± 0.7	0.3 ± 0.06
	Max	0.237 ± 0.005	0.28 ± 0.05	157 ± 2	23 ± 4	1.5 ± 0.3
	Median	0.22	0.27	26.7	3.5	1.1
	Mean	0.22	0.26	44.7	10.1	0.96
		(n=9)	(n=3)	(n=8)	(n=3)	(n=3)

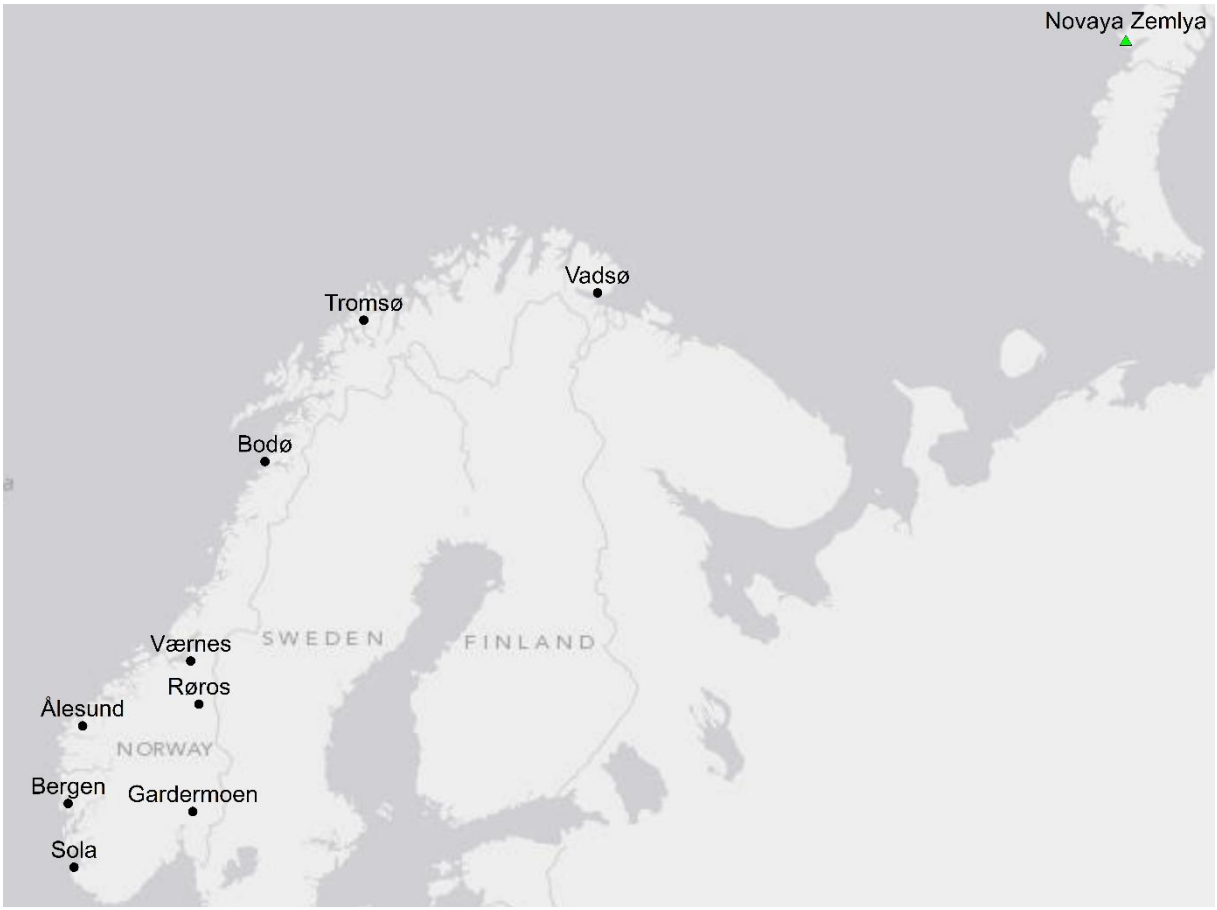
137 Table 3

Test site	Period	Sum yield (Mt)	Mean yield (kt)	Number of tests
Nevada test site	1945 – 1958	1	10	100
(USA)	1962	0.15	11	14
Pacific proving grounds	1946 – 1958	108	1721	63
(USA)	1961 - 1962	35	1154	30
Novaya Zemlya	1957 – 1958	21	766	27
(Former Soviet Union)	1961 – 1962	235	386	61
Semipalatinsk	1949 – 1958	6	127	49
(Former Soviet Union)	1961 – 1962	0.64	10	67

138 Table 4

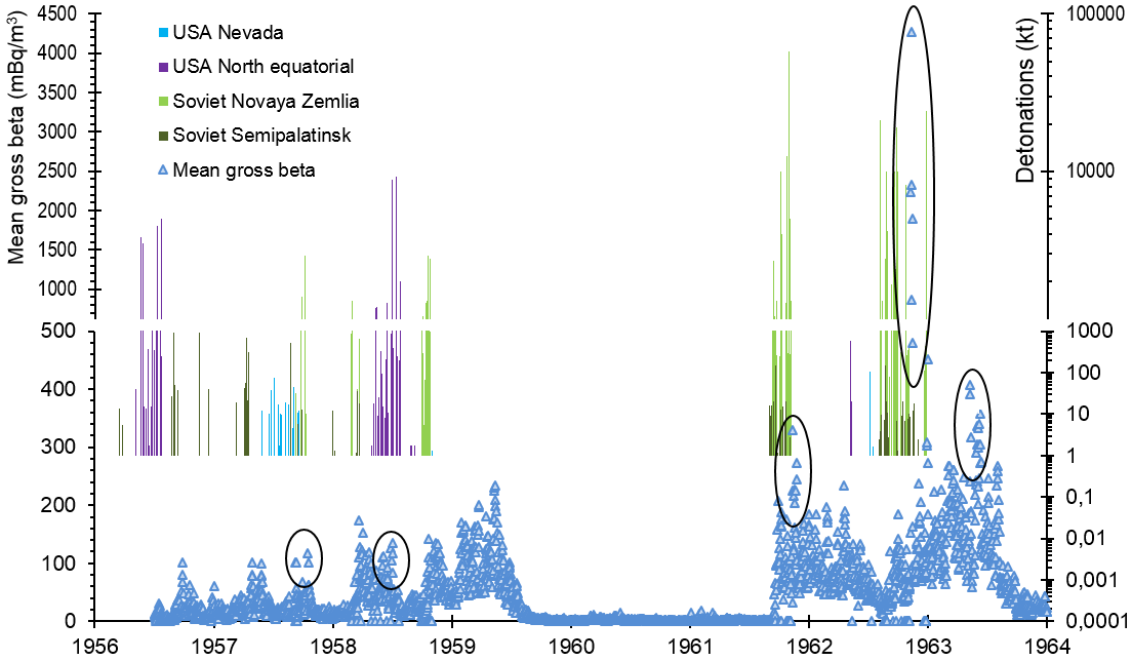
Station	Date 1962	Gross beta mBq m^{-3}	$^{239+240}Pu$ µBq m^{-3}	% contribution Semipalatinsk	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$ $\times10^{-4}$	$^{236}U/^{238}U$† $\times10^{-6}$	$^{236}U/^{239}Pu$
Sola	9.11	3142	169 ±2	86	0.060 ± 0.001			
Gardermoen	9.11	9092	575 ± 5	85	0.0614 ± 0.0007		17 ± 1	0.0188 ± 0.0009
Røros	9.11	11526	487 ± 4	89	0.0574 ± 0.0006	4.6 ± 0.7	15.9 ± 0.9	0.0246 ± 0.0009
Bodø	9.11	8132	301 ± 3	91	0.054 ± 0.001			
Tromsø	10.11	2050	116 ± 2	84	0.063 ± 0.003			
Bergen	11.11	6327	267 ± 2	87	0.059 ± 0.001	6.2 ± 0.9		
Ålesund	11.11	15467	782 ± 7	93	0.0517 ± 0.0009	3.8 ± 0.5	27.4 ± 0.8	0.0297 ± 0.0009
Værnes	11.11	2362	121 ± 1	85	0.062 ± 0.002		41 ± 2	0.046 ± 0.002
Ålesund	12.11	8781	631 ± 5	86	0.0605 ± 0.0008	2.5 ± 0.5		
Vadsø	12.11	464	11.7 ± 0.2	72	0.077 ± 0.004			
Ålesund	13.11	1762	87 ± 1	83	0.064 ± 0.003			
Global fallout	-	-	-	-	0.182 ± 0.005[a]	11.2 ± 0.85[a]	-	0.05 – 0.5[b]
Semipalatinsk ground Zero	-	-	-	-	0.0438 ± 0.0001[c]	2.21 ± 0.035[c]		0.024 ± 0.0001[c]

139 Figure 1



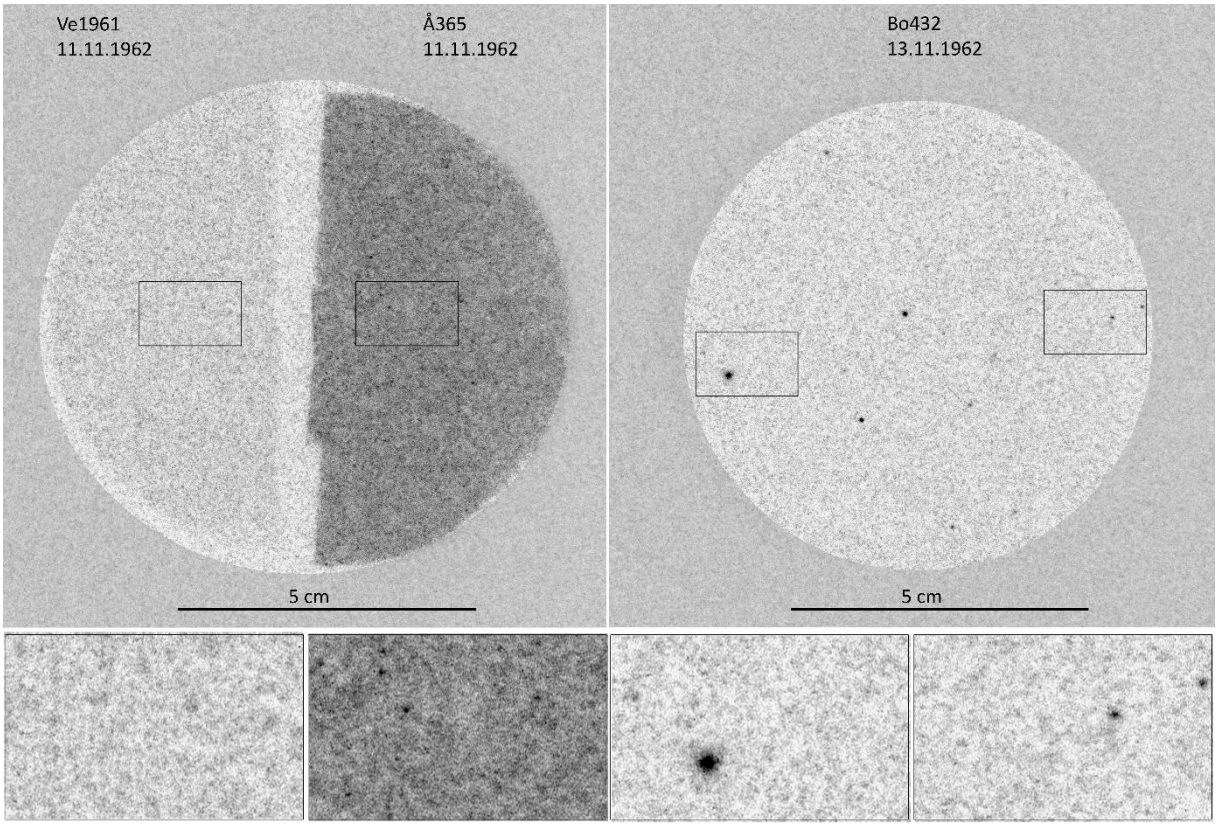
140

141 Figure 2



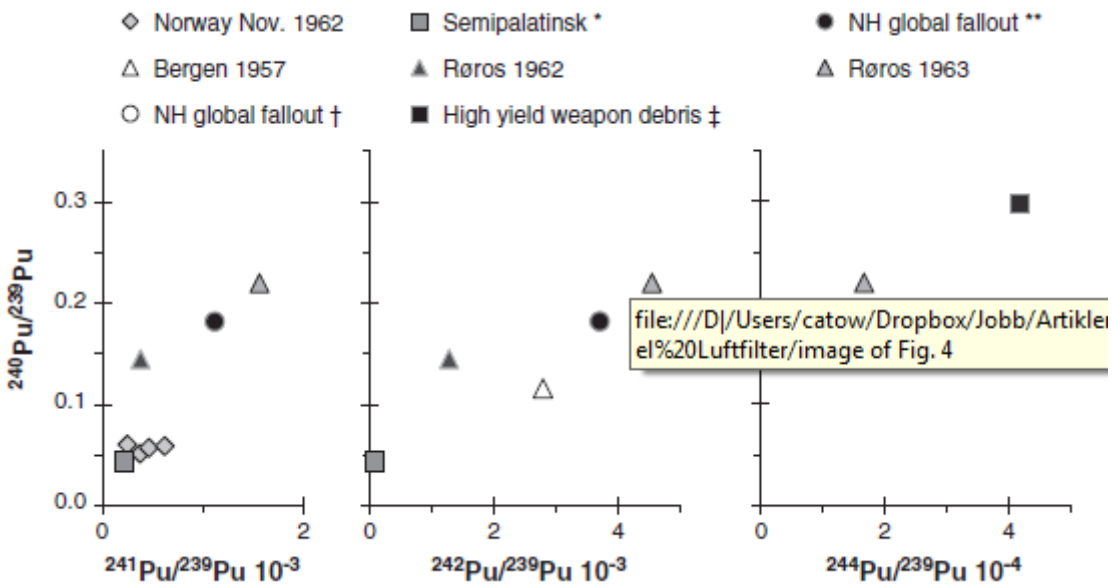
142

143 Figure 3



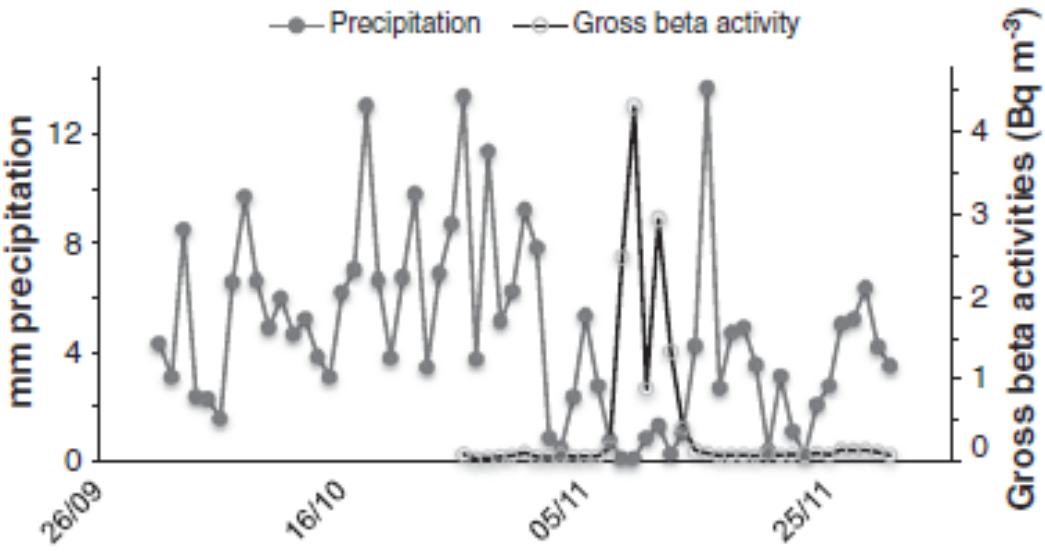
144

145 Figure 4



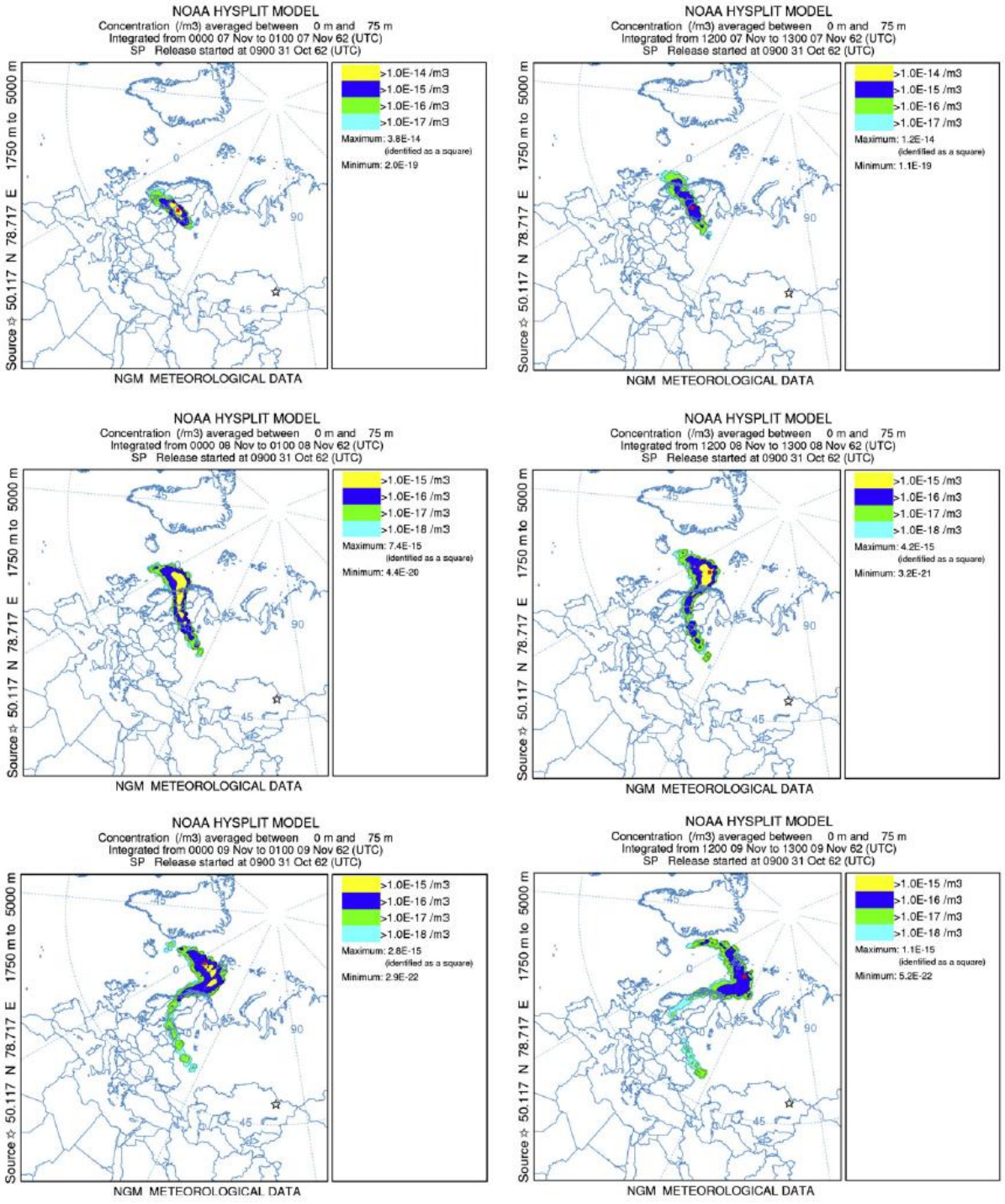
146

147 Figure 5



148

149 Figure 6



150

151 Supplementary materials, table 1

Site	Date	Gross B mBq m^{-3}	$^{239+240}$Pu µBq m^{-3}	±	^{236}U nBq m^{-3}	±	^{240}Pu/^{239}Pu atom ratio	±	^{241}Pu/^{239}Pu atom ratio ×10^{-4}	±	^{242}Pu/^{239}Pu atom ratio ×10^{-3}	±	^{244}Pu/^{239}Pu atom ratio ×10^{-4}	±	^{236}U/^{239}Pu atom ratio	±	^{236}U/$^{239+240}$Pu atoms Bq^{-1}×10^{-9}	±	^{236}U/^{238}U atom ratio×10^{-6}	±
Sola	09.10.1957	93	7.4	0.2			0.13	0.01												
Bergen	08 - 20.10.1957	941	n.a.		n.a.		0.116	0.007	n.d.		2.8	0.5	b.d.							
Sola	16.10.1957	321	3.0	0.2	1.7	0.4	0.09	0.02							0.7	0.2	613	134	6	2
Sola	17.10.1957	141	3.5	0.2			0.115	0.006												
Sola	22.10.1957	41	2.2	0.1			0.15	0.02												
Gardermoen	01.06.1958	127	22.1	0.5	2.2	0.4	0.169	0.008							0.15	0.03	115	11	115	45
Gardermoen	02.06.1958	152	24.9	0.5			0.170	0.007												
Gardermoen	03.06.1958	155	28.4	0.6			0.162	0.007												
Sola	27.09.1961	235	6.9	0.2	2.1	0.4	0.090	0.008							0.40	0.08	327	64	10	2
Gardermoen	28.09.1961	327	1.96	0.08			0.09	0.01												
Bergen	19.10.1961	428	3.2	0.1			0.086	0.009												
Røros	19.10.1961	98	15	3	0.17	0.06	0.20	0.07							0.019	0.008	12	5	b.d.	
Vadsø	19.10.1961	130	1.3	0.1			0.14	0.03												
Bergen	27.10.1961	65	1.7	0.2			0.09	0.02												
Røros	27.10.1961	35	b.d.		0.32	0.09	b.d.													
Vadsø	27.10.1961	83	1.73	0.09			0.15	0.02												
Bodø	10.11.1961	1093	14.9	0.7			0.09	0.01												
Tromsø	11.11.1961	888	1.7	0.1	0.9	0.2	0.07	0.02							0.35	0.09	521	105	30	10
Tromsø	12.11.1961	1747	16.3	0.5	1.6	0.1	0.067	0.006							0.12	0.02	105	21	21	5
Ålesund	17.11.1961	305	4.3	0.1			0.100	0.007												
Røros	11.01.1962	199	2.25	0.09			0.13	0.01												
Bergen	17.01.1962	297	18.1	0.5			0.110	0.007												
Røros	04.03.1962	58	12.3	0.2			0.127	0.005												
Bergen	15.04.1962	209	26.7	0.5			0.138	0.006												
Bergen	16.04.1962	205	27.8	0.4			0.137	0.005												
Bergen	17.04.1962	263	30.3	0.7	5.0	0.96	0.142	0.003							0.24	0.05	176	34	11	2
Bergen	18.04.1962	104	15.4	0.3			0.148	0.006												
Bergen	19.04.1962	157	21.4	0.5			0.138	0.007												
Bergen	20.04.1962	252	36.0	0.5			0.141	0.004												
Bergen	21.04.1962	219	29.1	0.4			0.152	0.005												
Bergen	22.04.1962	207	27.8	0.4			0.140	0.005												
Bergen	23.04.1962	222	34.0	0.5			0.156	0.005												
Bergen	24.04.1962	154	19.7	0.4			0.152	0.006												
Bergen	15.04.1962	78	3.2	0.1			0.143	0.001												
Røros	15 - 25.04.1962	1992	n.a.		n.a.		0.145	0.003	4	1	1.3	0.1	b.d.							
Ålesund	17.11.1962	48	2.48	0.09			0.21	0.01												
Bergen	24.04.1962	154	19.7	0.4			0.152	0.006												
Vadsø	25.04.1962	78	3.2	0.1			0.14	0.01												
Gardermoen	09.11.1962	9092	575	5	9	1	0.061	0.001							0.0193	0.0009	17.3	0.8	0.23	0.03
Røros	09.11.1962	11526	487	4	10	1	0.057	0.001	4.6	0.6					0.0247	0.0009	22.4	0.8	0.26	0.03
Bodø	09.11.1962	8132	301	3			0.054	0.001												
Sola	09.11.1962	3142	169	2			0.060	0.001												
Tromsø	10.11.1962	2050	116	2			0.063	0.003												
Ålesund	11.11.1962	15467	782	7	20	2	0.052	0.001	3.7	0.4					0.0298	0.0009	27.5	0.8	0.30	0.03

Værnes	11.11.1962	2362	121	1	4.7	0.5	0.062	0.002							0.046	0.002	41	2	0.18	0.02
Vadsø	11.11.1962	464	23.3	0.4			0.077	0.004												
Bergen	11.11.1962	6327	267	2			0.059	0.001	6.2	0.9										
Ålesund	12.11.1962	8781	631	5			0.061	0.001	2.5	0.5										
Ålesund	13.11.1962	1762	87	1			0.064	0.003												
Bergen	28.05.1963	162	20.1	0.7	2.6	0.1	0.190	0.006							0.156	0.007	122	6		
Bergen	29.05.1963	130	36.0	0.6			0.195	0.007												
Bergen	30.05.1963	306	27.1	0.5			0.23	0.01												
Bergen	31.05.1963	499	26.3	0.8	2.8	0.2	0.204	0.007							0.140	0.009	104	6		
Bergen	01.06.1963	393	62.8	0.7			0.237	0.005												
Røros	31.05-10.06.1963	4680	n.a.		n.a.		0.227	0.007	16	2	4.5	0.2	1.7	0.5						
Bergen	03.06.1963	332	35.3	0.6			0.221	0.005												
Bergen	06.06.1963	376	20.3	0.6	1.5	0.1	0.221	0.005							0.106	0.008	80	6		
Røros	08.06.1963	817	157	1	22.6	0.5	0.225	0.004							0.225	0.004	150	3		