6
Radioactive Aerosols – Chernobyl Nuclear Power Plant Case Study

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6.1
Introduction

On the night of 26 April 1986, the greatest accident in nuclear-power engineering took place at the fourth power-generating unit of the Chernobyl Nuclear Power Plant (ChNPP). The active zone and the upper part of the reactor building were completely destroyed as a result of the explosion. The safety barriers and systems protecting the environment against the *radionuclide*s produced in the irradiated nuclear fuel (*uranium dioxide*) over the previous two years were also destroyed.

The scientific literature contains many versions of the event: how the accident developed and how the active zone was destroyed [1]. The amount of nuclear fuel and its *fission product*s released outside the power-generating unit have been discussed. However, as for any explosion, the destroyed building constructions, fragments of the reactor's active zone, and finely dispersed uranium dioxide (the ''hot'' fuel particles) remain as silent witnesses to the accident. Condensed micrometer and submicrometer aerosols were spread over the whole Northern Hemisphere.

All the fuel assemblies (FAs) were destroyed, and most fuel element cans (FECs) proved to be empty. Mechanical or thermal degradation of the cans resulted in the formation of fuel dust. According to expert estimates, the total amount of uranium dioxide in the form of fine dust in the upper parts of the fourth power-generating unit make up about 30 tonnes [2].

To prevent further contamination of the region around the ChNPP and the environment as a result of any migration of radionuclides, a shelter of concrete and metal construction was built on top of the ruins of the reactor building; it was brought into commission on 30 November 1986. This construction object was named the ''Shelter.'' Observation of *radioactive aerosol*s of Chernobyl genesis both in the vicinity of the ChNPP and inside the destroyed unit itself remains a priority for the provision of radiation safety and understanding of the processes going on in the ''Shelter,'' including the evaluation of the state of the remaining nuclear fuel

and the lava-like fuel-containing materials (LFCMs) formed from molten nuclear fuel and construction materials.

To reduce the irradiation of building workers, all the works were performed by a remote method, which did not allow the creation of a leakproof construction. There were very many cracks in the upper part of the construction. Besides, operational openings and hatches were made in the roof to insert gage heads into the central hall above the reactor ''ruins.'' Finally, there is designed exhaust ventilation through the high-rise pipe HRP-2. Thus, intensive air exchange with the environment takes place in the ''Shelter,'' and external meteorological conditions influence the state of the air (the air temperature, humidity, direction, and rate of movement, and so on) indoors.

At present, radioactive aerosols from two sources can be emitted from the ''Shelter'': (i) those formed during the accident and being inside the ''Shelter'' in the form of dust; and (ii) new ones generated in the process of physico-chemical degradation of fuel-containing materials (FCMs), including the remaining uranium dioxide. The processes of FCM degradation occur under the influence of natural (radiation fields, air humidity, temperature variation, and so on) and man-made factors (any works performed in the ''Shelter''). These processes include mechanical embrittlement of FCM resulting in dust formation, radionuclide leaching from uranium dioxide matrix, and so on.

The radiological danger of aerosol products of the Chernobyl accident relates to the presence of highly toxic and long-lived fission and trans-uranium isotopes, in particular, plutonium isotopes. It is necessary to track the lowest concentrations of radioactive substances. Therefore, all the used methods of *aerosol radiometry* are based on their preliminary extraction from the air by any method followed by measurement in concentrated form. This is achieved either by forced pumping of air through filters or its expulsion through traps of different designs due to wind dynamic pressure or by gravitational settling on the surface (plates).

Pollution of air masses inside and outside the ''Shelter'' can be the result of a number of processes:

1) rise of dust from the soil, main traffic arteries, and vegetation in the vicinity of the ChNPP;
2) rise of dust from the surface of the ''ruins'' in the central hall, the reactor space, and other premises of the ''Shelter'';
3) dust formation in the process of construction and erection works;
4) dust formation and rise as a result of falling structural components in the ''Shelter'';
5) degradation of FCM as a result of radioactive processes and aging of materials;
6) leaching of radioactive substances, drying of solutions, formation and dusting of salt sediments.

The evaluation of the effect of the ''Shelter'' on the environment is a complex and multi-factor problem. As before, one of the main sources of potential

radioecological danger from the destroyed fourth power generating unit (hereafter reactor 4) of ChNPP is the process of air migration of radionuclides. It is important to know the *radionuclide composition* of aerosols, their concentration, the size distribution of particles (dispersity), the place of and reasons for the generation of aerosols, their methods of transport, deposition, and dissolution in the human respiratory system, and the efficiency of use of individual and collective protective devices.

Before we start to examine the aerosols related to the "Shelter," let us explain what radioactive aerosols are and describe some of their specific properties. Radioactive aerosols are aerodisperse systems of solid or liquid particles suspended in the air or in other gaseous media and consisting wholly or partially of radioactive substances. The radioactive component can be evenly distributed within the particle volume, can be on its surface, or can occupy a certain permanent or non-permanent place inside and/or on the surface.

Radioactive aerosols are characterized by all the features typical of common aerodisperse systems. However, the presence of *ionizing radiation* imparts a number of specific properties. These include, first of all, the presence of *electrical charge*s on the particles and ionization of both the material of the particles and the gaseous medium itself. Secondly, there is the possibility for spontaneous formation of nanoparticles (clusters) at radioactive decay of gaseous substances, for example, radon, thoron, xenon, krypton, and so on, as well as its daughter products. At radioactive alpha decay, for example, transuranium isotopes may generate recoil atoms. Thirdly, there is the possibility of detecting and studying aerosol particles as a result of their radioactive emission.

Radioactive aerosols are generated during various processes in the treatment of materials containing radioactive substances: fragmentation, grinding, bolting, pouring into another container, heating of solids, boiling, evaporation, pouring and bubbling of fluids, chemical interaction of substances in the gas phase, and so on. In the atomic industry, the formation of radioactive aerosols occurs mainly in the following situations: in the mechanical, metallurgical, and chemical treatment of ores (in particular, uranium ones), in the production of uranium hexafluoride, in the gas flows used to cool nuclear reactors, during radiochemical isolation of plutonium, at incineration and wet treatment of radioactive waste, and in the apparatus and plants of research laboratories. Large amounts of radioactive aerosols are generated in nuclear weapons trials, especially during explosions in the atmosphere. Accidents at nuclear power plants result in serious consequences and environmental contamination with radioactive aerosols.

All the above mechanisms of radioactive aerosol formation are special cases of dispersion and condensation processes causing the generation of common aerosols. Consequently, they have been sufficiently well investigated. However, there is a specific mechanism of generation of radioactive aerosols requiring special attention, as can be observed in the case of products of the Chernobyl disaster, that is, the appearance of radioactive recoil atoms or clusters after alpha decay in air. This mechanism was discovered at the beginning of the twentieth

century soon after the discovery of radioactivity. In the case of aerosols of Chernobyl genesis, this mechanism can be observed near surfaces of lava-like materials, which appeared in 1986 in the lower part of the destroyed reactor and in the sub-reactor premises as a result of fusion and spread of the remaining nuclear fuel. Almost a quarter of a century after the accident, these lava-like materials contain, besides uranium, also alpha-emitting radioisotopes of plutonium (^{238,239,240}Pu), americium (^{241}Am), and curium (^{244}Cm).

A nuclear explosion in the atmosphere when the fireball does not touch the ground surface is an example of the condensation mechanism for generation of radioactive aerosols. In the epicenter of the explosion, the temperature goes up to 1 000 000 °C. As the result, all nuclear reaction products and the components of mechanical constructions are immediately vaporized. In 0.01 s the temperature goes down to several thousand degrees. The formation of condensation aerosols of submicrometer size begins. If the explosion was near the ground surface, the rising and expanding fireball draws in a considerable amount of soil. This soil partially vaporizes and then condenses together with the products of the fired object. Some soil particles, for example, in the base of the ''atomic cloud,'' become radioactive because of the deposition of explosion products on them. A number of chemical elements present in the soil become radioactive as a consequence of neutron irradiation. The study of aerosols generated in atmospheric nuclear explosions performed in 1950–1960 detected individual highly active particles named ''hot'' particles. Similar particles formed in the Chernobyl accident were detected both in the surroundings and at long distances, including the countries of continental Europe and Scandinavia.

Since the cessation of nuclear weapons trials in the atmosphere (the last such explosion was performed in China in 1980), radioactive products have been continually settling on the ground surface. By the end of the twentieth century, the atmosphere had practically cleared itself of the radioactive aerosols generated during nuclear trials. Against this background, radioactive aerosols formed in the Chernobyl accident and released into the *troposphere* and the *stratosphere* were distinctively detected in the Northern Hemisphere in 1986 and later. No aerosols of Chernobyl genesis have been found in the Southern Hemisphere. This is caused by the specificity of air exchange between the hemispheres of the Earth, which was revealed in the observations of radioactive products of nuclear trials already by the middle of the twentieth century.

Carriers of natural cosmogenic radionuclides occupy a special place among radioactive aerosols, which can be classified under the category of condensation ones. They are ^{3}H (tritium), ^{7}Be, ^{14}C, ^{22}Na, ^{24}Na, ^{32}P, ^{33}P, ^{35}S, and so on. They appear as a result of nuclear reactions in the upper atmospheric layers under the influence of neutrons and protons of cosmic radiation. The radioactive atoms formed settle on small atmospheric particles and then slowly get into the lower tropospheric layers. As the above-listed radionuclides have half-lives from several hours to several years, they can be used successfully to study atmospheric processes. They figure as tracers in many works on *nuclear meteorology*.

During the last decades of the twentieth century, the attention of radioecologists was drawn to radon gas and its daughter products (DPs). This was preceded by the discovery of an association between radon concentration and cancers of the lungs and upper respiratory tract in uranium miners and other underground workers. High radon concentrations can also be observed in cellars and on the first floors of dwellings and offices, where radon penetrates from the ground through cracks, poorly sealed joints, and leakage. This gas is known to appear as a result of the radioactive decay of ^{238}U. It was shown that the inert gas itself is not as dangerous as its short-lived DPs, especially alpha-emitting ones. As they appear in radon decay in the form of polonium, bismuth, and lead atoms, and have high diffusion coefficients, they deposit in the upper respiratory tract. If the air is dusty, the formed atoms settle on small atmospheric particles and penetrate into deep sections of the lungs (alveoli and primary bronchi).

The problem of radon and its DPs exists also in the ''Shelter'' of the ChNPP. Its sources are the huge masses of concrete and the sandy ground in which the foundations of the destroyed reactor 4 are embedded. The ''Shelter'' has no forced ventilation. Besides radon, the premises of the ''Shelter'' also emanate thoron, which is partially generated in the decay of the ^{232}U formed in the nuclear fuel of the reactor during its almost 850 days of working before the accident.

Some radioactive substances can exist in the atmosphere and in production equipment in the forms of aerosols and gases. These include mercury, polonium, iodine, ruthenium, tellurium, and so on. After the Chernobyl accident, one of the most biologically dangerous radionuclides, ^{131}I, was observed in the atmosphere in both phases for two months. The portion of radioiodine in aerosol form varied from 10% to 100%. In the gas phase, this radionuclide was present in the form of molecular iodine (I_2) and methyl iodide (CH_3I) vapors. In this connection, sorption-filtering materials entrapping both phases should be used for protection against radioiodine and for analytical purposes.

Radioactive aerosols are used for medical purposes and in scientific research, for example, in the above-mentioned nuclear meteorology. Their main advantage over other methods is the possibility of exact measurement using different radiometers. Specific methods of measurement of radioactive substances, especially alpha-emitting ones, include autoradiography and solid-state detection. One of advantages of autoradiography is the possibility of visualizing, isolating, and analyzing individual particles. This was done with aerosol samples collected at air nuclear trials for the purpose of isolation of ''hot'' particles and with samples collected after the Chernobyl accident.

Below are the results of observations and studies of aerosols in the ''Shelter'' and its vicinity performed in 1986–2008. *Petryanov filter*s, which were specially developed for radioactive aerosol studies (for a detailed description of these filters, see Part 3 of this book), were used for collection and analysis of aerosol samples throughout the entire duration of this study [3, 4].

6.2
Environmental Aerosols

6.2.1
Dynamics of Release of Radioactive Aerosols from Chernobyl

Estimates of the daily release of radioactive substances (Figure 6.1) are included in ''Information on the accident at the Chernobyl NPP and its consequences'' [1] presented by the USSR State Committee on the Use of Atomic Energy for the Meeting of IAEA experts, which took place on 25–29 August 1986 in Vienna.

The dynamics of the release has been widely commented upon in the literature [5–9]. Here is, for example, an extract from [8]:

Radioactive substances were released into the atmosphere during the first ten days after the accident before the release was stopped. The heat of the fire increased the levels of released radioactive iodine (^{131}I and ^{133}I), a considerable portion of volatile elements from the group of metals including radioactive cesium (^{134}Cs and ^{137}Cs) as well as a somewhat smaller amount of other radionuclides that are usually present in the fuel of a reactor, which has been working for several years.

This release was not a one-time event. On the contrary, only 25% of material was released on the first day of the accident, the remaining amount was released during the subsequent nine days (Figure 6.1). The curve of the release intensity depending on the time can be divided into four areas:
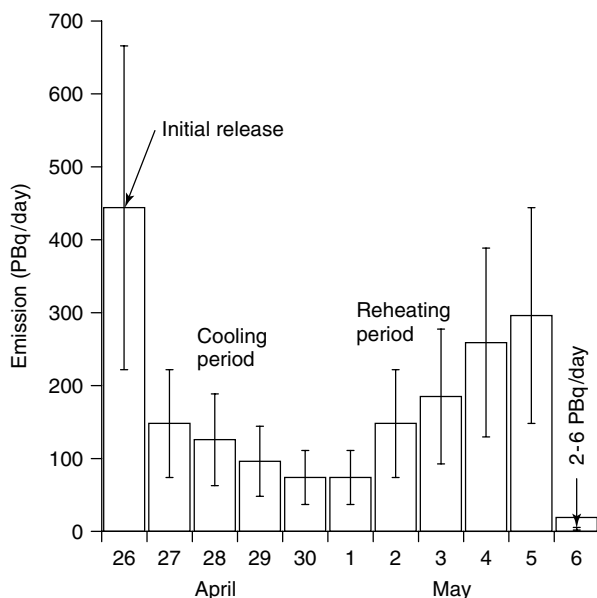

Figure 6.1 Emission of radioactive aerosols to the atmosphere during the 1986 Chernobyl disaster (24 h average with radioactive decay taken into account).

1) The first release took place on the first day of the accident. During this period, the physical release of radioactive materials was the result of explosion in the reactor and the subsequent heating as a result of the fire and the effect of the active zone.

2) During the subsequent five days, the release intensity decreased to a minimum approximately six times smaller as compared with the initial level. At this stage, the decrease in the release intensity was achieved due to measures to control graphite combustion and due to the reactor cooling. These measures, including the casting of 5000 t of boron carbide, dolomite, clay and lead from helicopters to the active zone resulted in the filtration of radioactive substances released from the active zone. At this moment, a release of fine-dispersed fuel took place immediately together with the flow of hot air and smoke from burning graphite.

3) After that there was a period of four days during which the release intensity increased again up to 70% of the initial level. First the release of volatile components, especially iodine was observed; the subsequent composition of radionuclides reminded a typical composition of spent fuel. This phenomenon was attributed to the fuel heating in the active zone to 2000 °C, which was caused by residual heat generation and isolation with materials cast down to the active zone.

4) Ten days after accident there was a sharp reduction in the release intensity to less than 1% of the initial one. Subsequently, the release intensity was constantly decreasing. The last stage, which started on May 6, was characterized by a sharp decrease in fission yield and gradual cessation of release. These events were the consequence of special measures, which resulted in binding of fission products into more stable chemical compounds [4, 10].

The dynamics of the release of radioactive substances from the destroyed reactor 4 is one of the key aspects of the acute phase of the accident. A review of the initial materials [4] used to evaluate the radioactive release in the 1986 accident report presented by Soviet experts for IAEA was published by Yu.V. Sivintsev and A.A. Khrulev (two researchers of the Kurchatov Institute – Russian National Center) in the journal *Atomic Energy* in 1995 [11].

Many authors have tried to reconstruct the *release dynamics* and to calculate the amount of released radioactive aerosols, including ^{137}Cs and especially radioiodine. Models of *atmospheric transport* that were available before the Chernobyl accident, for example, the ARAC system in the USA [12], were used, and new ones were created for different spatial and time scales. A review of the comparison of results obtained with the models used is presented in [13]. The authors of the review state that the results of reconstruction of the total amount of ^{137}Cs released with the method of solution of the inverse problem of atmospheric transport coincide quite well with each other in spite of the differences between the models. They also coincide with the results of similar evaluations performed with other methods. The differences in the reconstruction of the release dynamics are more noticeable. This is associated with the fact that it is much more difficult to calculate the spatial

features of the formation of radioactive contamination fields in individual periods at the initial stage of the accident. That is why the quality of the model – that is, the detailed description of the radionuclide dispersion and deposition processes as well as the completeness and reliability of meteorological data – plays the decisive role in the reconstruction of the dynamics of nuclide activity in a given territory.

6.2.2
Transport of Radioactive Clouds in the Northern Hemisphere

The data presented in Figure 6.1 have been used in many works, for example, for the calculation of the transport of gas–aerosol discharge from the ChNPP in the Northern Hemisphere [14–16]. The heights to which they rise in the atmosphere are discussed. The results of sampling carried out from an airplane above Poland show that a portion of the radioactive aerosols was in the stratosphere [17]. As a result of the large thickness of the cloud, transport of the initial products of the explosion at different heights occurred in both the westerly and easterly directions. The monograph [6] presents the trajectories of air-mass transfer at different heights for a week after the accident.

The winds carried the torn-apart initial radioactive cloud from above the ChNPP at comparatively low height (about 3–4 km) through northwest Europe. From [6, 18–21] it follows that two or three days after the accident this part of the cloud passed above Belarus, Lithuania, Poland, the Baltic Sea, Sweden, and Finland. According to [22], air masses reached the southwestern part of Finland at about noon on 27 April 1986 (at heights of 1500 and 2000 m) and three hours later at lower heights (1000 and 1500 m). The next day, daily aerosol samplings from an airplane were started at these heights [23]. According to the data of the Finnish Center for Radiation and Nuclear Safety [24, 25], the maximal surface concentration of ^{137}Cs of about 10 Bq/m^3 was recorded on 28 April in the south of the country in Nurmiyarvi region. No other large amounts of Chernobyl aerosols coming to Finnish territory were observed.

In the report [26] it was noted that, after leaving Scandinavia, the radioactive cloud reached the North Atlantic on 2 May. At the monitoring point in Resolute in the far north of Canada, the first increase of air *activity concentration* was recorded on 2–3 May. In the sample collected here over 24 hours the aerosol concentration already exceeded the background level by five times. Some 4000 km to the southeast, on the east coast of Canada, the first noticeable increase in the air activity concentration took place in Greenwood, Halifax, Fredericton, and Digby with the filters exposed on 4–5 May. As follows from Figure 6.2, the maximal concentrations of radioactive aerosols were recorded on 10–12 May.

Other winds carried radioactive substances, which rose at the reactor explosion to heights of 5–7 km and even into the stratosphere, above Siberia. For instance, in Novosibirsk, a sharp activity increase was recorded on the night of 30 April–1 May [27]. Radioisotopes of both volatile substances (^{134}Cs, ^{137}Cs, ^{103}Ru, ^{106}Ru) and substances of low volatility (^{95}Zr, ^{95}Nb, ^{141}Ce, ^{144}Ce, ^{90}Sr) were found in the aerosols. Then these winds transported radioactive clouds above China and Japan
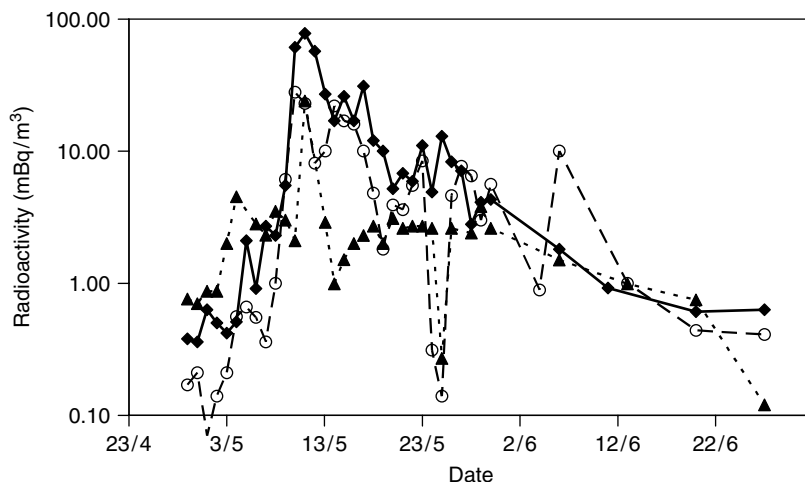
Figure 6.2 Radioactivity of beta-emitting mixtures in aerosol samples in Canada in April–June 1986. The results were obtained at Digby (open circles), Resolute (full triangles), and Edmonton (full diamonds) monitoring stations.

and brought them across the Pacific Ocean to the west coast of the USA and Canada. The authors of the report [26] think that the head of the radioactive cloud reached the west coast of Canada on 7 May. In any case, in Vancouver (the most westerly point of the Canadian monitoring network), an increased activity concentration of aerosols was observed in the sample exposed on 5–6 May, and quantitatively ^{131}I, [134]Cs, and ^{137}Cs were detected in the sample collected on 8–9 May.

However, in the southwestern part of Canada, that is, in Winnipeg, Saxatoon, Calgary, and Edmonton, the products of the accident were recorded for the first time somewhat earlier (4–5 May). Most probably, they were brought there through the northwestern part of the USA.

Thus, Canada turned out to be a region of the Northern Hemisphere where radioactive clouds of the first release from the destroyed reactor 4 torn apart by winds above ChNPP came together and mixed. This meeting and mixing of radioactive clouds above Canada, clouds which after the reactor explosion spread in opposite directions round the globe, can be considered one of the peculiarities of Chernobyl aerosol release. Another peculiarity consisted in the fact that the analysis of samples collected in Canada did not reveal detectable quantities of the low-volatility radionuclides [95]Zr, [95]Nb, [140]Ba, [140]La, [141]Ce, and [144]Ce, though the volatile [103]Ru, ^{131}I, [132]Te, [134]Cs, [136]Cs, and ^{137}Cs were detected. [131]I was in both aerosol and gaseous forms. The portion of the latter varied from 20% to 100%.

One can judge the significance of aerosol fallout from the atmosphere and its contribution to environmental pollution by the following extract from the monograph [28]: "Ingress of ^{137}Cs with waters of the Dnepr and the Danube to the Black Sea in 1986–2000 was extremely insignificant and made up approximately 1% of its atmospheric fallouts." The authors of [29–32] came to the same conclusion.

No radioactive aerosol products of the Chernobyl accident were found in the Southern Hemisphere.

6.2.3
Observation of Radioactive Aerosols above Chernobyl

Sampling of gas−aerosol radioactive substances in the vicinity of the ChNPP and along transfer routes was started the day after the accident. In [33, 34] it is reported that on the night of 27−28 April 1986 the first operative sample was collected above the ruins of reactor 4 of ChNPP from an An-24rr airplane laboratory. Then helicopters began to work together with airplanes.

However, the first (and we can say random) aerosol sample from the plume of the radioactive cloud was collected on the day of the accident (26 April) during the duty flight of an An-30rr airplane of the USSR State Hydrometeorological Committee above the western part of the European territory of the country. Having landed on one of the aerodromes, the operators discovered that the gondola with FPP-15-1.5 filtering material, which had been working during the flight, had a strong radiation background. The intensity of the exposure dose amounted to several R-units per hour. This was reported to the leadership of the USSR State Hydrometeorological Committee. The sample was promptly taken to Kiev and measured on a gamma-spectrometer. The results obtained were included in the report presented to IAEA [9].

The results of measurements of isotope composition and concentrations of radioactive aerosols collected from 28 April to 19 May 1986 with filters installed on helicopters are presented in Figure 6.3. The flights were usually performed at an altitude of about 200 m within a radius of 0.5−1 km from the ruins of the reactor building. Sampling from An-24rr airplane laboratories equipped with filtering gondolas [34−37] was performed at altitudes of 200−1200 m within a radius of 2−5 km from the ChNPP. The concentrations of radioactive aerosol products of the Chernobyl accident are presented in Figure 6.4 [35]. It can be seen that the concentrations vary considerably. In part, this is explained by differences in sampling places and times for airplanes and helicopters. Nevertheless, the comparison of Figures 6.3 and 6.4 allows us to observe the synchronism of changes in the concentrations of radioactive aerosols. High values were recorded on 28 and 29 April and on 4 and 16 May. The maximal values were observed not on 5 May, as follows from Figure 6.1, but on 4 May. On that day, the total beta activity at aircraft sampling made up about 55 000 Bq/m^3, and at helicopter sampling the total activity of nine gamma-emitting nuclides was 6900 Bq/m^3. From Figure 6.4 it is clearly seen that an increase in the concentrations of radioactive aerosols above the ruins of the reactor building was taking place with time. However, within only three months of observations, but not in the second half of May, as reported in [5], the concentrations decreased by six orders of magnitude. In Figure 6.4 we can distinguish four areas: 28 April−4 May, 5−23 May, 24 May−6 July, and 7 July−6 August.

On the whole, the first area agrees with Figure 6.1. In the second area, the maxima are observed on 7−9 May and 16−21 May. The concentration of 16 May
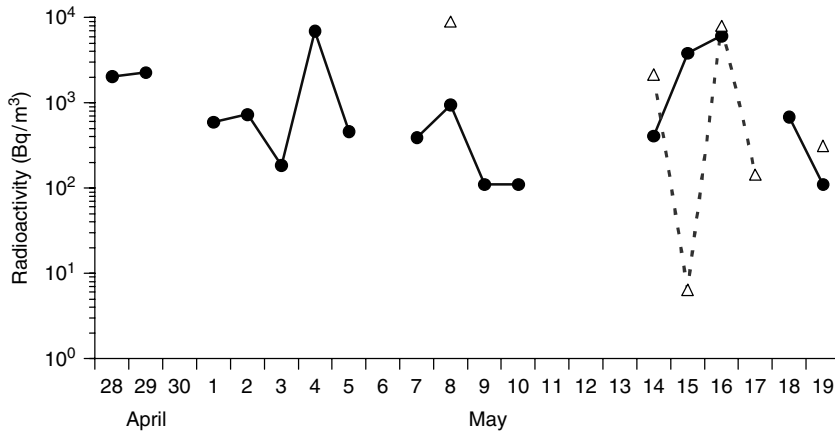
Figure 6.3 Radioactivity of nine gamma-emitting nuclides in aerosols above the ruins of the fourth block of the Chernobyl NPP in April–May 1986. Samples were obtained by using helicopter (full circles) and aircraft (open triangles).
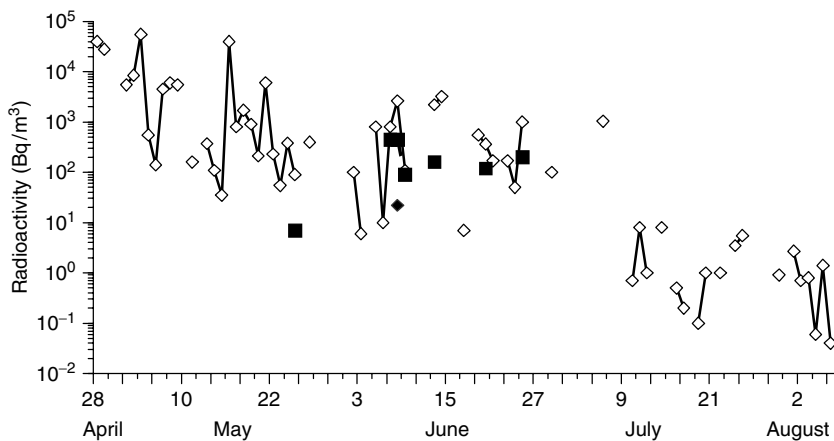

Figure 6.4 Concentration of beta-active aerosols above the Chernobyl NPP in April–August 1986 (collected from aircraft).

was conditioned mainly by refractory elements and was only ~1.5 times lower than the value obtained on 4 May. In the third area, the practically constant concentration of radionuclides remained at a level approximately 100 times lower than on 4 May. Four values exceeding 1000 Bq/m^3 were recorded in June, and one more such value was recorded early in July. The fourth (also practically horizontal area) is characterized by a sharp decrease in radioactive aerosols on average by two more orders of magnitude. In two cases, the concentration was lower than 0.1 Bq/m^3. Pronounced maxima are absent.

Thus, the results of aircraft and helicopter sampling did not confirm such a sharp decrease in the concentration radionuclide concentration of radioactive substances above the reactor as described in [4–8]. Moreover, the studies performed in the early fall of 1986 showed that the release from the destroyed reactor 4 still remained at a high level. In those experiments, in late August and early September, aerosols were collected on filters placed on a rope thrown over from the eastern to the western wall of the reactor at a height of 20–30 m from the surface of the destroyed reactor, and on 1–2 October they were collected from the arm of a construction crane at the erection of the roof of the ''Shelter.'' It turned out that the aerosol concentrations in the flow coming out of the ruins of the reactor building made up $10^2 - 10^4$ Bq/m^3, that is, they were several orders of magnitude larger than those recorded from an airplane in July and August (see Figure 6.4). Thus, already in summer the air leaving the ruins was not warm enough to rise to the height of the patrolling airplane. It can be supposed that, since the second half of May, the aerosol concentrations recorded in aircraft samples collected even at the lowest flying height of 200 m were considerably lower than those in flows leaving the reactor – it was not possible to go down lower because of the 150 m high ventilation pipe (VP-2).

Variations of aerosol concentrations (see Figures 6.3 and 6.4) were accompanied by significant differences in their radionuclide composition. Volatile radionuclides (iodine, ruthenium, and cesium) prevailed at low activity concentrations of aerosols. These include samples collected from a helicopter on 1–3, 5, 7–10, 14, and 18 May. The activity of samples collected on 1–3 May were practically fully conditioned by ^{131}I, that of samples collected on 8 May by ^{103}Ru and ^{106}Ru, and that of samples collected on 14 May by ^{134}Cs and ^{137}Cs. Refractory radionuclides (zirconium, niobium, and cerium) prevailed in samples collected in periods of high activity concentrations of aerosols. Such samples were collected from an airplane on 28–29 April, 4, 15, and 16 May. In some cases, they contained small amounts of ruthenium and cesium radioisotopes.

In five cases, samples were collected from a helicopter and an airplane on the same day. However, on account of the differences in the heights and flight durations, the concentrations and radionuclide composition of aerosols coincided only in samples collected on 16 May. This was indicative of the variability of the discharge composition even during short time periods.

At a number of airplane probings, the gondolas were equipped with SFM-I sorption filtering materials developed at the L.Ya. Karpov Physicochemical Institute [38]. In these cases, not only aerosols but also some gaseous radioactive products, including iodine, tellurium, and ruthenium, were entrapped from the filtered air. It was revealed that ^{95}Zr, ^{95}Nb, ^{134}Cs, ^{137}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, and ^{7}Be (a radionuclide of cosmogenic origin) were practically completely entrapped in the front aerosol layer from FPP-15-1.5 material, while ^{103}Ru, ^{106}Ru, ^{131}I, and ^{132}Te were also found in the subsequent sorption layers containing activated carbon impregnated with AgNO$_3$ [39].

The detection of ruthenium and tellurium in SFM-I sorption layers was indicative of their presence in the atmosphere not only in aerosol form but also in gaseous

form. The portion of ^{103}Ru in gaseous components made up 0.6–13%, that of ^{106}Ru made up 2.7–16%, and that of ^{132}Te made up 1–8%. The identity of the ^{103}Ru and ^{106}Ru distributions between sorption layers of material allowed us to conclude that they were present in the air as part of the same gaseous substances (obviously, RuO_4). Gaseous compounds of ^{132}Te were also well entrapped.

The study of radioiodine distribution over the layers of SFM-I material showed that its gaseous substances were adsorbed with more difficulty than those of ruthenium and tellurium. Obviously, ^{131}I is in the air not only in the form of molecular iodine vapor, but also as a part of organic compounds (methyl iodide, CH_3I). It should be noted that, during the sampling period above the reactor from 8 to 19 May, the portion of gaseous forms of iodine increased from 30% to 90%.

During the flight on 14 May, a three-layer filter intended for determination of the dispersion composition of aerosols was placed in one of gondolas besides the sorption filtering material [40, 41]. It was revealed that refractory radioisotopes (^{95}Zr, ^{95}Nb, ^{140}La, ^{141}Ce, and ^{144}Ce) and their compounds were concentrated on particles with active median aerodynamic diameter (AMAD) of about 0.7 μm at a standard geometric deviation $\sigma = 1.6$–1.8. Volatile radionuclides (^{131}I, ^{103}Ru, ^{106}Ru, and ^{132}Te) having gaseous compounds were associated with considerably smaller particles with AMAD of 0.3–0.4 μm at $\sigma = 2.3$–2.5. The difference in sizes of particles containing radionuclides was most probably conditioned by the physico-chemical processes taking place within the ruins of the reactor building in the remaining nuclear fuel and uranium fission products. Aerosols of detected sizes are involved in long-range atmospheric transport and can be observed at the distance of many thousands of kilometers from the place of generation.

6.2.4
Observations of Radioactive Aerosols in the Territory around Chernobyl

After the beginning of the construction of the ''Shelter,'' the Radiometric Laboratory manned with specialists and equipped with apparatus from the V.G. Khlopin Radium Institute was entrusted with aerosol monitoring in the nearest vicinity of the ChNPP [42]. On 11 June 1986 they began daily aerosol sampling at 13 points onboard a BTR armored troop-carrier patrolling around the perimeter of the ChNPP. The air was pumped through AFA RMP-20 filters. The results of gamma-spectrometry of samples on a semiconducting detector with a Nokia LP-4900 analyzer are presented in Figure 6.5. It can be seen that the activity concentrations of the mixture of gamma-emitting nuclides were high and varied considerably; in five of 19 samples the concentrations exceeded 740 Bq/m^3, that is, the temporarily prescribed permissible norm (maximum permissible concentration–MPC). High concentrations were the consequence of construction and decontamination work near reactor 4 as well as intensive flow of traffic. By the end of June, the composition of radionuclides became stable due to the decay of iodine, lanthanum, barium, tellurium, and other isotopes. In July, a number of premises inside the plant were included in the work program. On 14 September,
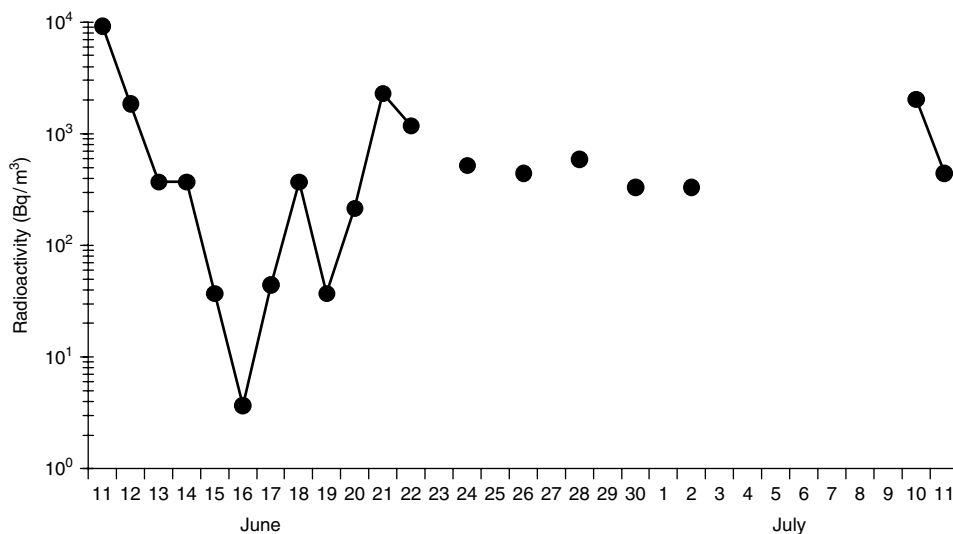
Figure 6.5 Concentration of gamma-active nuclides in aerosols at ground level of the Chernobyl NPP in June–July 1986 (samples were collected along the perimeter of the NPP).

more points, including ones in Chernobyl, were added. Figure 6.6 presents some results of this monitoring (before commissioning the ''Shelter'').

From Figure 6.6 it follows that, near the administrative and communal building (ACB-2) in July, August, and September, the concentrations of aerosol carriers of gamma-emitting nuclides were usually in the range of 100–1000 Bq/m^3. Such high concentrations were conditioned by the fact that the sampling point was only 300 m to the east of the place where intensive activities on erecting the northern cascade wall of the ''Shelter'' were going on. The pit-face made by miners who were erecting a concrete refrigerator plate under reactor 4 was nearer still. Only in October and November, when the construction works at the ''Shelter'' became less intensive and a large area near it was covered with ''clean'' soil and concreted over, did aerosol concentrations decrease to 10–100 Bq/m^3.

The sampling point near ACB-1 was approximately 1.5 km to the east of reactor 4. Though the initial explosion products were thrown away in a westerly direction, aerosol concentrations near ACB-1 and ACB-2 proved to be comparable. Most probably, this was associated with the intensive flow of traffic near ACB-1, where cars were constantly coming from regions with a high degree of pollution of soil, buildings, and machinery. Significant variations of concentrations were also likely to occur under the influence of the meteorological situation (precipitation and wind). For instance, one of the highest concentrations (4440 Bq/m^3) was recorded near ACB-2 on 21 August in a strong west wind with gusts up to 10–15 m/s.

Only in October and November, when radioactive substances that had fallen as a result of the accident were removed from rather large areas near the erected
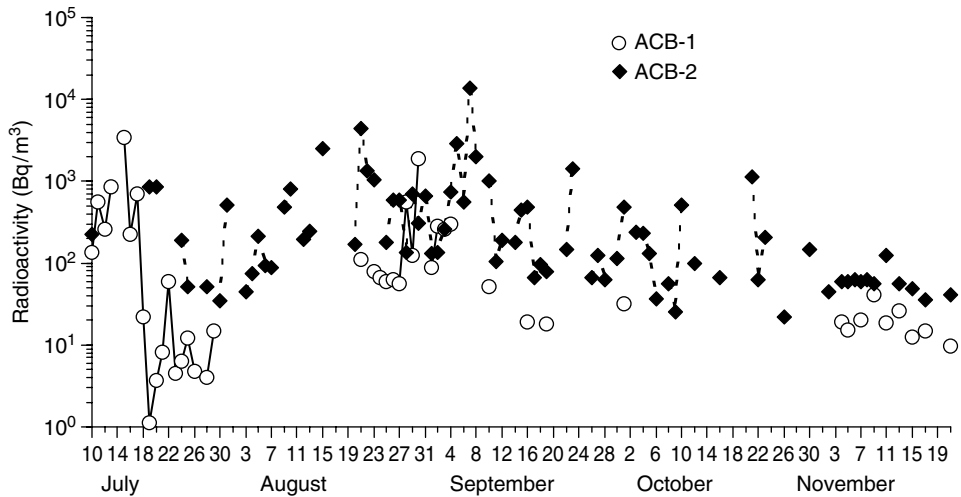
Figure 6.6 Concentration of gamma-active nuclides in aerosols at ground level of the Chernobyl NPP in June–November 1986 (samples were collected near ACB-1 and ACB-2).

"Shelter," and the areas were covered with gravel and sand and concreted over, did aerosol concentration decrease by an order of magnitude as compared with the levels observed in July to September. Rains in the fall also contributed to the reduction of dust rise.

From the end of August to the middle of September, aerosol samples near ACB-2 were also collected by specialists of the L.Ya. Karpov Physicochemical Institute [43]. They were interested in the aerosols that got into the air-raid shelter located in the basement of ACB-2. Figure 6.7 presents the results of such observations.

During the observation period, considerable variations in the concentrations were recorded at the air-raid shelter. For instance, the minimal values recorded on 20 and 25 August made up about 14 Bq/m^3, and the maximum recorded on 21 August amounted to 1110 Bq/m^3. The reason for the spike in concentration was the strong wind mentioned above. Besides the concentrations, the radionuclide composition was also unstable. There was a very high concentration of refractory elements in samples collected on 20–22, 25–26, and 30–31 August. Consequently, aerosols of fuel composition prevailed near ACB-2 on those days. The sample collected on 2 September was different. The portion of volatile substances (radioruthenium and radiocesium) in it made up about 65%. This correlated with [19–21] where it was noted that radiation of individual highly active particles collected in Sweden and Finland (thousands of kilometers away from the ChNPP) was practically completely caused by radioruthenium.

The most probable reason influencing the state of radioactive pollution of the atmosphere near ACB-2 was aerosols transported from the western part of the ChNPP area, where there are places of heavy fallout of accident products, rather
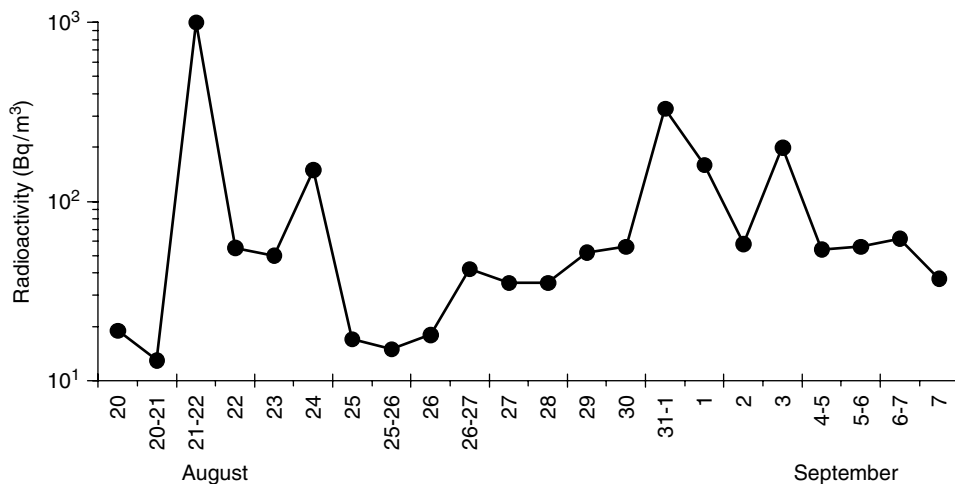
Figure 6.7 Concentration of gamma-active nuclides in aerosols at 1 m above the ground near ACB-2 in 1986.

than aerosols coming directly from the ruins of the reactor building. This confirms the relation of the aerosol concentrations with the wind rose. The highest aerosol concentrations were observed for west winds recorded on 21, 22, 24, and 31 August as well as on 3 September. The stronger the wind (for example, on 21 August its gusts achieved 15 m/s), the higher the concentration. The lowest concentrations of radioactive aerosols were observed in east winds recorded twice (on 20 and 25 August).

Besides the specialists of the V.G. Khlopin Radium Institute and the L.Ya. Karpov Physicochemical Institute, in the fall of 1986 regular aerosol sampling was performed by employees of the F.E. Dzerzhinsky All-Union Heat Engineering Institute. Their four control points were located at the plant periphery at a height of 1 m from the ground; three of these were in the places of the air intakes for input ventilation in three units of the first line of the ChNPP [44]. Figure 6.8 presents the results obtained at surface level. Aerosol concentrations varied from 1 to 220 Bq/m^3 and coincided with the data obtained near ACB-1 (see Figure 6.6) in order of magnitude. On the whole, as noted previously (see Figure 6.6), in the fall of 1986, the concentration of radioactive aerosols in the vicinity of the ChNPP distinctly decreased.

Besides AFA filters, a cascade impactor was used for sampling on 9–26 July. The results revealed the existence of bimodal size distribution of particles around the perimeter of the main building of the ChNPP first line. The portion of finely dispersed fraction (the activity of which was 90% conditioned by ^{103}Ru and ^{106}Ru) made up on average 8% of the total activity of gamma-emitting nuclides. The AMAD of this fraction was about 0.7 μm at $\sigma = 2$–3. The rest of the activity fell into the fraction of coarsely dispersed aerosols with AMAD of 8–12 μm at the same σ [44].

After the commissioning of the "Shelter," the arrangement of the radioactive aerosol control system on its territory took about a year. When, in 1988,

well-boring was started to seek FCMs in reactor 4, a team of specialists from the Complex Expedition at the I.V. Kurchatov Institute of Atomic Energy was entrusted with the control of the aerosol situation inside the ''Shelter'' and its surroundings. Four filtering plants (FPs) were already functioning within a radius of 300 m from the reactor by that time. They provided round-the-clock air pumping through FPP-15-1.5 material at a flow rate of $400-500 \, m^3/h$. The filter exposure lasted five days. The results obtained in the fall of 1988 are presented in Figure 6.9.

As can be seen in Figure 6.9, the highest values amounting to 100 Bq/m^3 were observed in the southern part of the territory. For all three plants the difference between the maximal and minimal concentrations made up 1–2 orders of magnitude. Though the distance between the plants was only 200–300 m, the

Figure 6.8 Concentration of gamma-active nuclides in aerosols collected near the first line of the ChNPP in August–December 1986.

Figure 6.9 Integrated concentration of gamma-active nuclides in aerosols collected in various directions in the vicinity of the ''Shelter'' in 1988.

concentrations at simultaneous samplings practically did not coincide. From this, it follows that the aerosol situation in the places where the plants are located was determined by the effects of local sources rather than by regional air masses, probably like in other points near the ''Shelter.'' Such a situation was still observed in 1989–1990.

Three years after the accident, when radioactive products with half-lives of less than 100 days had practically disappeared, attention was mainly focused on the content of alpha-emitting aerosols, as they began to determine the dose due to ingress of substances by inhalation, and ^{137}Cs, which made a major contribution to external irradiation.

Summarized data on the concentrations of ^{137}Cs and ^{241}Am aerosols in the local zone collected during 15 years in the ''Areal'' laboratory created at the Interdisciplinary Science and Technology Center ''Shelter'' of the National Academy of Sciences of the Ukraine are presented in Figure 6.10. Practically all the samples were obtained at 15-day exposure of FPP-15-1.5 filters. Considerable (up to three orders of magnitude) differences in the minimal and maximal values of both ^{137}Cs and ^{241}Am during comparatively short time periods are observed. The greatest variations are recorded for samples collected using the plant installed in the southern part of the territory. The reason is probably associated with the higher concentrations of uranium, and consequently its fission products, found here after the accident. Nevertheless, in Figure 6.10 it can be seen that, on the whole, ^{137}Cs and ^{241}Am concentrations in the places where the northern, northwestern, and southern plants are installed are very close to each other. One other important conclusion follows from the graphs in Figure 6.10: since December 1992, the concentration of ^{137}Cs aerosols decreased within seven years by approximately five times. This value is much larger than that achieved due to radioactive decay (the half-life of ^{137}Cs is $T_{1/2} = 30.2$ years).

A still sharper decrease in ^{137}Cs concentration in the air during the first years after the accident was reported in [45, 46]. Samplings in Pripyat City, situated at a distance of 5 km to the northwest of the ''Shelter,'' over a period of five years (from July 1987) revealed that the concentration of aerosol carriers of ^{137}Cs had decreased by approximately 20 times. During subsequent years, this process slowed down and even stabilized. The authors of [46] represented the concentrations of ^{137}Cs (μBq/m^3) by the equation

$$C_0(t) = 6725 \, \exp(-t/30) + 346$$

where t is the number of years after commencement of observations. They predicted that the persistence of such regularity will be valid till 2009.

The reason for the sharp (during the first years after the accident) and then delayed decrease in the concentration of aerosols of ^{137}Cs and other radionuclides (in particular, plutonium isotopes) in the air is probably associated with natural and man-made factors: embedding of accident products in the soil, decontamination of the region around the ''Shelter'' and planting of greenery on it, dust control at construction and erection works, and so on. At the same time, a more rapid decrease in the concentration of ^{137}Cs aerosols in the air than that calculated for
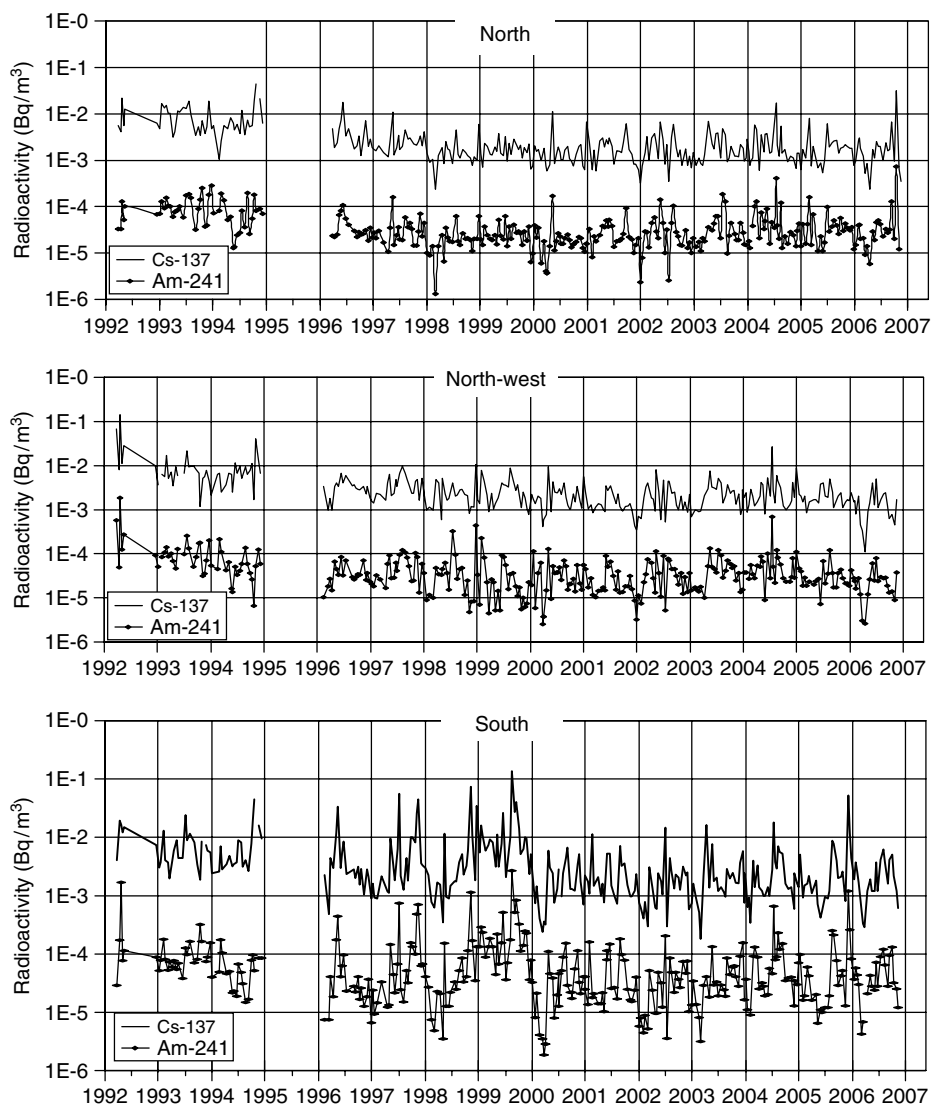
Figure 6.10 Radioactivity of aerosol particles containing
[137]Cs and [241]Am obtained in the vicinity of the "Shelter"
(north, northwest, and south) in 1992–2007.

$T_{1/2} = 30.2$ years indicates that the contribution of discharge from the "Shelter" to the current aerosol situation is insignificant.

Though the nuclear physical origins of ^{137}Cs and [241]Am are different – the first one being the product of uranium fission, and the second one the product of beta-decay of [241]Pu ($T_{1/2} = 15.2$ years) produced in the fuel – the dynamics of their

concentrations in 1992–2007 in the air in the region of the "Shelter" were practically identical (see Figure 6.10). ^{241}Am concentrations in the range of 0.01–0.1 mBq/m^3 prevailed among almost 900 samples. The minimal concentrations were usually not lower than 0.003 mBq/m^3, and the maximal ones only in several cases approximated to or somewhat exceeded 1 mBq/m^3. The highest concentrations were more often recorded near the southern FP-3. The three times that activity concentrations of ^{241}Am amounted to 1 mBq/m^3, it was associated with construction works. For example, in the late spring–early summer of 2000, intensive excavation works were going on at the site of the construction of a spent nuclear fuel repository (SNFR-2) located 2 km to the southeast of the "Shelter." In August–September 2006, a path was laid (scraping and loading of soil, delivery, and leveling of gravel) onto the berm near FP-3. It should be noted that these roadworks caused only an increase in ^{137}Cs and ^{241}Am concentrations in samples from the southern plant. No increased radionuclide concentrations were recorded in samples collected simultaneously at the northern and northwestern plants (see Figure 6.10).

The radionuclide ratio is known to be very important for the identification of radioactive aerosols and the detection of their sources of origin. From Figure 6.10 it can be easily estimated that the ^{137}Cs/^{241}Am ratio was usually in the range of 50–70. These values are typical of nuclear fuel with burnup of 11 MWt day/kg of uranium [47]. Thus, the aerosol situation around the local zone of the "Shelter" is determined by the so-called radioactive fuel particles. It is these particles that fell near reactor 4 after the accident on 26 April 1986. The high ^{137}Cs/^{241}Am ratios (120–190) in December 2000 and January 2001 are probably associated more with inaccuracy of the sample measurements on account of the very small concentrations of radioactive substances observed during the winter season, when the rise of dust from the ground was complicated, than with the ingress of so-called condensation aerosols (see Figure 6.10).

Soon after the commissioning of the "Shelter," the specialists of the "Kombinat" production association started to control the state of the air medium around it at distances above 0.5 km [48]. VP-2 was chosen as the zone center. Within a radius up to 1 km, samples were collected in 11 points, and at distances of 1–3 km in 13 points. Figure 6.11 presents summarized aerosol concentrations of the gamma-emitting nuclides from March to December 1987. It can be seen that the concentrations of radioactive aerosols in both zones were decreasing synchronously. They were usually five times lower at 1–3 km distance than at 0–1 km distance. This is explained by a decrease in both the density of local contamination with radionuclides that had fallen after the accident and the intensity of dust rise due to human activities and strong winds. From the approximating dotted straight lines, it follows that the period of the activity decrease by half was about 22 days. As at the beginning of these observations all radionuclides of accident origin with half-lives less than 30 days no longer existed, the high rate of activity decrease can be explained only by the effect of natural and anthropogenic factors: the decontamination of the locality, roads, constructions, as well as embedding of the fallen substances into soil. Besides, the intensity of construction works including the flow of traffic also decreased.
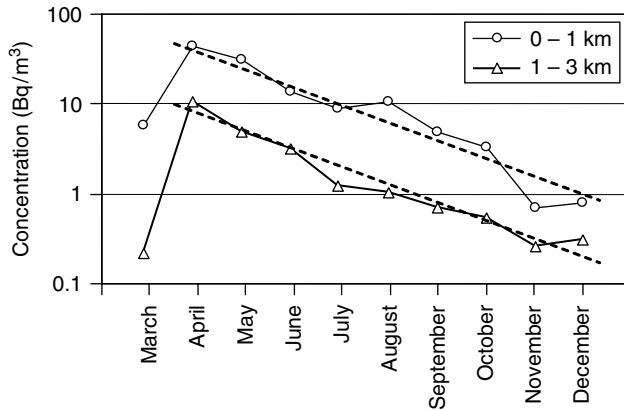
Figure 6.11 Concentration of gamma-active nuclides in aerosols collected at distances of 0–1 and 1–3 km from the fourth block of the ChNPP in March–December 1987.

The Automated Radiation Monitoring System began to function in the 30 km Chernobyl exclusion zone in 1988. It included about 30 stationary points of round-the-clock aerosol sampling using FPP-15-1.5 materials. Five points of this type systematically collecting aerosols are located within the radius of 0.5–3.0 km from the "Shelter" [49, 50]. The specialists of the Exclusion Zone Radioecological Monitoring Center work at these points. Their data are an important supplement to observations carried out in the surrounds of the "Shelter."

At the control point under consideration, the aerosol situation largely depends on the works performed near the FPs and the weather conditions, in particular the *wind speed*. For instance, excavation and construction works in the vicinity of the "Shelter" in the spring of 2000 were the source of radionuclide-contaminated dust, which was recorded practically simultaneously at sampling on 4–18 May in the southern part of the "Shelter" region and on 7–12 May at the points of the outdoor switchgear and Petroleum Storage Depot. At this time, the ^{137}Cs concentration (135 mBq/m^3) in the sample from near the "Shelter" proved to be maximal during the 10-year observation period starting from 1993, and was also maximal at the outdoor switchgear (16 mBq/m^3) and the Petroleum Storage Depot (2.5 mBq/m^3) [51].

In the second half of 2002, radioactive aerosol monitoring with a "Typhoon" aspirator was commenced near the office building standing 2 km to the west of the "Shelter" [46]. The high efficiency of the aspirator (about 4000 m^3/h) allowed representative samples to be obtained within short time periods, for example, a day. Data on ^{137}Cs concentration are given in Figure 6.12. It can be seen that most often the concentrations were within the range of 0.1–0.3 mBq/m^3, that is, an order of magnitude lower than in the vicinity of the "Shelter." During the period under consideration, sharp increases in radioactive aerosol concentration caused by natural factors were observed twice: early in September as a consequence of forest fires, and early in December on account of strong winds. These situations will be described below in more detail. Here it should also be noted that the
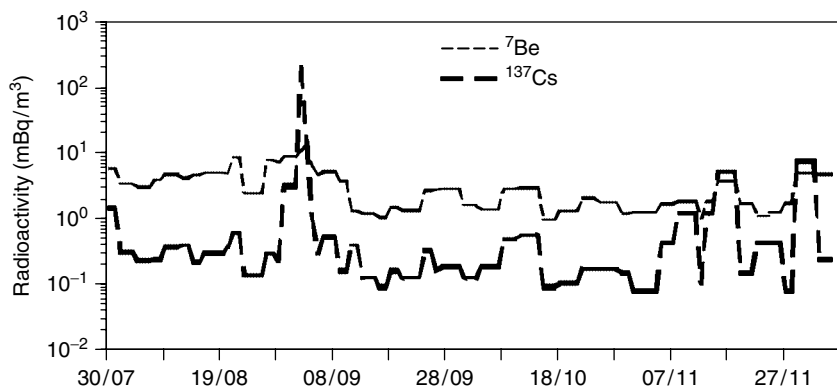
Figure 6.12 Radioactivity of ^{7}Be and ^{137}Cs containing aerosols at ground level 2 km from the ''Shelter'' (near the administrative building) from 30 July to 5 December 2002.

concentrations of cosmogenic ^{7}Be – which does not bear any relation to the products of the Chernobyl accident and is a peculiar marker of air masses coming from the upper troposphere and from the stratosphere – were high in August and in the first half of September and then decreased by 4–5 times.

The influence of high wind speeds is manifest in an increase in the number and variation of sizes of particles, which start to take off from the underlying surfaces (ground, roads, roofs, leaves of trees and bushes, grass, and so on). Naturally, the higher the density of local contamination with the products of the accident and the subsequent ingress of radionuclides of Chernobyl genesis as a result of human activities, the larger will be the concentrations of radioactive aerosols in the air.

The effect of high wind speeds can be traced by the example of the situation in March 2003, which influenced aerosol samples collected by stationary plants in the region of the ''Shelter'' as well as those located in the vicinity of the ChNPP [52]. According to the data of the ''Chernobyl'' weather station, the month's highest wind speeds were observed on 20–22 March: average wind speeds up to 5 m/s and gusts up to 14 m/s. The mean daily temperatures of the air, which had been above freezing since 11 March, went down below zero (to −5 °C), and then went up again to 2–6 °C after 24 March. Snow, which fell on 20–21 March, reduced dust rise from the soil and roads to a certain extent.

High concentrations of ^{137}Cs aerosols were recorded around the ''Shelter'' at exposure of the filters from 17 March to 2 April (Figure 6.13). The concentration increased by an order of magnitude as compared with the average annual value in the southern part of the territory. A similar situation was observed at three aerosol monitoring points located in the vicinity of the ChNPP during the sampling period from 18 to 24 March: ^{137}Cs concentrations at the points of the outdoor switchgear, Petroleum Storage Depot, and bored piles increased by an order of magnitude as compared with the average annual level. It should be noted that, at the Petroleum Storage Depot and bored piles, the increase in aerosol concentration could not be
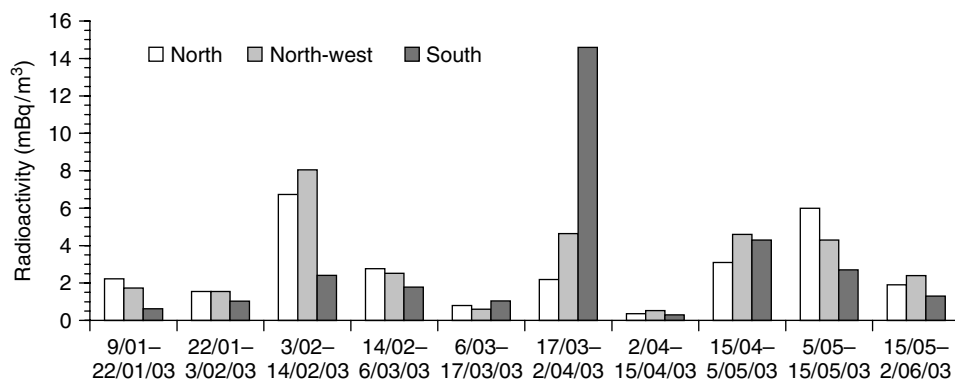
Figure 6.13 Concentration of ^{137}Cs in aerosols at ground level obtained in the vicinity of the ''Shelter'' in January–May 2003.

the consequence of carryover of radioactive substances through the VP-2 pipe and leakages (cracks) of the ''Shelter,'' because a stable northwest wind was observed on 17–22 March. For the Petroleum Storage Depot this is important as the point is located 2 km to the northwest of the ''Shelter.'' The bored piles are located outside the plume of aerosol carryover, 3 km to the east of the ChNPP.

Observations at the ''Chernobyl'' weather station over many years show that dust storms can be observed in the Chernobyl exclusion zone, with gusts stronger than 15–18 m/s. One of them occurred a year after the accident. Gusts of 25 m/s were recorded by the ''Chernobyl'' weather station on 18 April 1987. The probability of occurrence of dust storms in the Kiev region is from 2.1% to 31.7% in different months. The maximal probability falls at July.

A synoptic situation, which also caused a dust storm, emerged in the first 10 days of September 1992 in the northern part of the Ukraine, in the Bryansk region of Russia, and in the southeastern part of Belarus. These regions are among areas highly contaminated with the products of the Chernobyl accident, which fell down from radioactive clouds in April–May 1986. The dust storm was caused by a powerful cyclone, which arose in the Balkans on 4 September and was above the Baltic Sea on the coast of Lithuania on 8 September. Thanks to scheduled and unscheduled aerosol samplings performed on these days at a number of weather stations in the Ukraine and Belarus, as well as at monitoring points in the Chernobyl exclusion zone, it became possible to obtain unique data on the composition of dust raised into the air by strong winds.

A considerable increase in the concentrations of atmospheric dust and radioactive products of the accident was recorded in all the above regions. In the Chernobyl exclusion zone, the duration of exposure of FPP-15-1.5 filters was reduced from 5–7 days to 2–23 hours. It was revealed that the concentrations of the accident products increased during the storm by 1–2 orders of magnitude. Aerosol sampling in Vilnius (500 km to the northwest of the ChNPP) revealed a 100-fold increase in ^{137}Cs concentration. The calculation of the trajectories of air-mass transfer on

6–7 September showed that this ^{137}Cs could have come to the environs of the Lithuanian capital at altitudes of 1.5 km from the Kiev region (Ukraine) and the Gomel region (Belarus).

The evaluation showed that, on 6–7 September 1992, when average wind speeds of 8–12 m/s with gusts up to 25 m/s were observed in the Chernobyl exclusion zone for 30 hours, the amount of the ^{137}Cs radioisotope that could rise into the atmosphere from territories with a density of contamination of this isotope of 3.7×10^6 Bq/m^2 could exceed by four times the admissible monthly release of a normally functioning 6 GWt thermonuclear power plant.

It was mentioned above that fires on areas contaminated with the products of the Chernobyl accident could result in a sharp increase in the concentration of radioactive substances in the air. From 30 August to 4 September 2002 near the administrative building of ''Shelter'' the smell of smoke was in the air, and abnormally high ^{137}Cs concentrations were recorded [53].

As follows from Figure 6.12, the maximal value of about 170 mBq/m^3 fell on 2–3 September. At the same time, increased concentrations of ^{134}Cs, ^{90}Sr, ^{241}Am, and plutonium isotopes were recorded. On those days, the average concentration of ^{137}Cs in the air increased by three orders of magnitude. The ^{137}Cs/^{90}Sr ratio was about 30, which is approximately 20 times higher than that in nuclear fuel at the moment of the accident at reactor 4 [47]. Such an increase in cesium concentration in aerosols had already been observed after forest fires in regions contaminated with the products of the Chernobyl accident [54–56].

Space photographs were used to detect the sources of smoke. The fires were detected using an infrared imager. Not less than 45 forest fires occupying an area of 0.3–1.2 km^2 were recorded over an area of 200×200 km^2. All of them were in the eastern sector relative to the ChNPP near the border between the Ukraine and Belarus, half of them being at the distance of only 20–40 km from the station. The weather favored both the development of the fires and the transport of combustion products toward the ChNPP. According to the data of the ''Chernobyl'' weather station, from 29 August to 5 September 2002 the night temperature was 11–15 °C, and the day temperature went up to 30 °C. Air pressure was in the range of 752–758 mmHg. Wind speed varied from 1 to 2 m/s. Sometimes calm was observed at night, and only in the daytime on 30 August did the wind speed amount to 3 m/s. Maximal gusts up to 8 m/s were recorded. In this period from 12 to 6 p.m. the wind had a southern direction. On all the other days, air masses were coming only from the southern and eastern quarters, which provided the transport of combustion products to the vicinity of the ChNPP.

Thus, the analysis of space photographs and the meteorological situation in late August–early September 2002 showed that the smoke seen in the area around the ''Shelter'' and its administrative building as well as the 100-fold increase in the concentrations of ^{137}Cs and other radionuclides were associated with forest fires in the area between the Dnepr and Pripyat rivers near the northeastern boundary of the Chernobyl exclusion zone, where there are vast territories contaminated with radioactive products of the accident.

The works [57–59] carried out on experimental sites in the Chernobyl exclusion zone by specialists from the Ukrainian Institute of Agricultural Radiology showed that high concentrations of radioactive products of the Chernobyl accident are generated not only in forest fires but also in grassland fires. The formed smoke particles of micrometer sizes containing radionuclides are involved in long-range atmospheric transport.

6.2.5
Dispersity of Aerosol Carriers of Radionuclides

Undoubtedly, the results of the measurement of the disperse composition of radioactive aerosols are interesting from the point of view of radiation safety. Dispersity is the basic parameter determining *aerosol deposition* in respiratory organs. The transport of particles in the atmosphere and working areas as well as the functioning of treatment plants, analysis, and individual protection devices depend on their sizes.

After the accident, the first samples for evaluation of the dispersion composition of radioactive aerosols were obtained by specialists of the USSR Ministry of Defense. In [60] there are data on the distribution of radioactive aerosol at a height of 2 m over the territory of the ChNPP on 12 May 1986. Exact sampling points and techniques are not reported. Only the distribution of particles in 1 cm^3 of air in the range from 0 to 1.6 μm in 0.2 μm steps is presented. Data processing showed that they are well approximated by the log-normal distribution with median aerodynamic diameter of 0.62 μm and $\sigma = 1.5$.

Sampling was performed above the ruins of reactor 4 from an An-24rr airplane at an airspeed of 350–400 km/h, at a height of 300 m above the ground, on two days [36]. In one of the two gondolas installed on the fuselage, there was a pack of three-layer Petryanov filters with an area of 1 m^2 through which air passed at a rate of 0.9–1.2 m/s. The flight over, each of the pack layers was measured on a gamma-spectrometer. On 14 May 1986, it was found that the refractory radionuclides ^{95}Zr, ^{95}Nb, ^{140}La, ^{141}Ce, and ^{144}Ce had AMAD of about 0.7 μm at $\sigma = 1.6$–1.8. These results were practically identical to those recorded on 12 May. As for the volatile radionuclides ^{103}Ru, ^{106}Ru, ^{131}I, and ^{132}Te, they were bound with considerably smaller particles whose AMAD was in the range of 0.3–0.4 μm at $\sigma = 2.3$–2.5. This difference was probably caused by the high temperatures in the ruins of the reactor building and the physico-chemical processes taking place there.

The studies performed on 9–26 July 1986 by the specialists of the F.E. Dzerzhinsky All-Union Heat Engineering Institute in the area of the first line of the ChNPP were mentioned above. They used a cascade impactor to reveal a bimodal size distribution of particles. Approximately 8% of the total gamma-activity of the samples fell in the finely dispersed fraction with AMAD of about 0.7 μm at $\sigma = 2$–3; 90% of the activity was determined by ^{103}Ru and ^{106}Ru. The coarsely dispersed fraction had AMAD of 8–12 μm. Most probably, the generation of large particles occurred as a result of construction works and traffic flow.

On 10 September 1986, before the erection of the roof of the ''Shelter'' was started, a sample was collected on a pack of three-layer Petryanov filters located approximately 20 m from the surface of the ruins of the reactor building [61]. Measurements and calculations showed that the AMADs of the particle carriers of ^{141}Ce, ^{144}Ce, ^{134}Cs, ^{137}Cs, ^{95}Zr, and ^{95}Nb were equal and in the range 0.98–1.14 μm. Obviously, each aerosol particle contained all the isotopes in constant ratio. Only the AMAD of the particle carriers of ^{103}Ru and ^{106}Ru was somewhat smaller (0.74–0.92 μm).

The specificity of the particle carriers of radioruthenium was clearly seen from the results of observations performed in May, July, and September 1986 in different places: they systematically had smaller sizes. This was obviously associated with the presence of the volatile compound RuO_4, which in the gaseous state deposited on small atmospheric particles having sizes of 0.1–0.3 μm.

After the arrangement of radiation observation points in the Chernobyl exclusion zone, packs of three-layer Petryanov filters were regularly used to determine the dispersion composition of radioactive aerosols. Filters with an area of 0.3 m^2 were continuously exposed for 5–7 days at a flow rate of about 1 m/s. Air pumping over, each layer was measured on a gamma-spectrometer and transferred for radiochemical analysis. Not only the isotope products of the Chernobyl accident, but also the DPs radon and thoron, as well as cosmogenic ^{7}Be, were determined. The results of measurements of particle sizes collected at the ChNPP are shown in Figure 6.14 [62]. For almost 20 years, the sizes of the carriers of the radionuclide products of the accident remained stable: their AMAD was in the range of 3–8 μm. No separation of radionuclides (^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{239}Pu) by particle size was observed. At the same time, ^{212}Pb (the DP of thoron) and ^{7}Be were on considerably smaller particles with AMAD
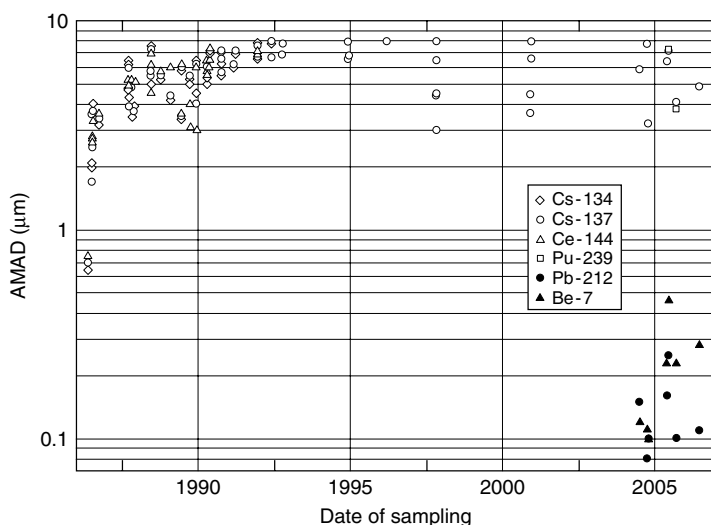

Figure 6.14 Size of radioactive particles (^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{239}Pu) and natural nuclides (^{212}Pb and ^{7}Be) collected at the ChNPP in 1986–2006.

0.1–0.3 μm. This is quite natural, as [212]Pb and [7]Be atoms on emergence deposit on small atmospheric particles, and the radionuclide products of the Chernobyl accident are associated with dispersion particles formed during mechanical processes and the wind rise of dust. It should be added that in 1988–1991, when units 1–3 of the ChNPP were working, ^{131}I was found in some samples. Its carriers were particles with AMAD of 0.3–1 μm [63]. Besides, radioiodine was present in gaseous form.

6.3
Aerosols inside the Vicinity of the "Shelter" Building

6.3.1
Devices and Methods to Control Radioactive Aerosols in the ''Shelter''

The studies and measurements of radioactive aerosols inside the ''Shelter'' were started soon after its commissioning (30 November 1986). They took on special significance in 1988–1991 after drilling activity to seek nuclear fuel remaining in the ruins of the reactor building.

All scheduled samplings were carried out with analytical filters AFA RMP-20 or AFA RSP-20 made of Petryanov fibrous polymeric materials. The use of RSP filters is preferable, as their main filtering layer is made of ultrathin perchlorovinyl fibers with a diameter of about 0.5 μm. This provides for entrapment of aerosols in the front layer and allows measurement of radioactivity without the need for a correction due to the absorption of alpha-particles by fibers. The filters are made in the form of disks with an area of 20 cm^2. The linear flow rate varied from 50 to 150 cm/s. AFA filters have high efficiencies of aerosol entrapment for a broad range of particle sizes and flow rates [3]. The scheduled aerosol sampling in the ''Shelter'' is carried out using portable blowers powered from the electricity network or accumulators.

In premises where personnel stay all the time, aerosol samples are collected once a day, and in the ''Shelter'' once a week. However, when repair, construction or decontamination works as well as special studies are conducted, both the number of points and the sampling periodicity are increased. Statistical data indicate that 1200 samples were obtained in 1992. Later on, their number continually increased. The maximum (20 800 samples) was achieved in 2000–2003. Approximately 10 000 samples were collected in 2006 and 2007. The decrease is associated with the rearrangement of the Radiation Monitoring System at the ''Shelter'' and the introduction of individual aerosol samplers, the filters or impactor heads of which are located in the workers' respiratory areas [64]. On average, such devices collect about 1000 samples annually.

6.3.2
Control of Discharge from the "Shelter"

Control of discharge from the ''Shelter'' is performed in the ''Bypass'' system, where aerosols from the former central hall of reactor 4 come through an air-duct.

They then go either to the VP-2 or to the filtering station. From the ''Bypass,'' an aerosol sampling line is laid (a 35 m long tube with a diameter of 15 mm) to the radiometric plant RKS2-03 No. 9 ''Kalina'' and a cartridge with an AFA filter. Automatic aerosol sampling on a filtering tape made of Petryanov material, the measurement of activity, and transfer of the results to the duty operator control desk are performed in ''Kalina.'' The tape is changed every 6, 12, or 24 hours. The AFA filter may be removed and measured on the instruction of the duty operator at any time. The results obtained form the basis of the calculation of the total discharge from the ''Shelter'' (daily, monthly, and annual).

Air release from reactor 4 through the ''Bypass'' occurs due to the natural draft in the VP-2 pipe. Its top is at a height of 150 m. If the release of the mixture of long-lived beta-emitting nuclides exceeds 26 MBq/day, it can be decided to close the shutter of the ''Bypass'' and to switch the fans for air transport to the filtering station for purification and then to VP-2. However, in the 23 years since the accident, the shutter in the ''Bypass'' has not been closed. This is indicative of the normal aerosol situation in the ''Shelter.''

The problem of the transport of radioactive aerosols from the premises of the ''Shelter'' that houses the remaining nuclear fuel and its fission products has been a subject that has attracted the attention of scientists and specialists of the Radiation Safety Service for many years. The first studies of air flows in the reactor cavity using helium labeling were performed in 1988–1989 by researchers of the V.G. Khlopin Radium Institute. The results showed that air flows often change their direction and speed. In 1990, the work was continued, using not only helium but also molecular tritium, ^{14}C (as a part of methane), and ^{85}Kr. After that, about 30 points were selected, where the temperature, relative humidity, speed, and direction of air flows are measured every week.

The release of radioactive aerosols from the ''Shelter'' occurs not only through the ''Bypass'' to the VP-2 pipe, but also through numerous cracks in the external constructions. Their total area in the late 1980s was estimated at 1200 m^2. Later on, after repair and stabilization work, most cracks were stopped up. By the end of the 1990s, the area of holes had been stabilized at a level of 120 m^2. Air gets into the ''Shelter'' and leaves it through cracks. Much depends on the air temperature and pressure, the wind direction and velocity in the environment, the season, and a number of other factors. Observations showed that air flows in cracks are unstable. The measuring equipment should be placed in all the main openings for exact measurements of the carryover of radioactive aerosols. However, this is not feasible. Besides, the complex radiation situation impedes their maintenance.

6.3.3
Well-Boring in Search of Remaining Nuclear Fuel

Examination of the central part of the ''Shelter'' with well-bores began in 1988. The main task was to seek the remaining nuclear fuel. About 40 horizontal and inclined wells, usually with a diameter of 172 mm, were examined during three years. The length of some of them amounted to 20 m.

Though boring was accompanied by washing, the pollution level of the air medium in the working and auxiliary premises was rather high. Besides dust escaping at drifting of concrete walls and floors, sometimes disintegrated material was spilled onto the floor because of inaccurate extraction of the core and the boring tool. About 1000 people were carrying out boring at the most intensive stage. Dust control using localizing and accumulating solutions was performed to reduce aerosol pollution of the air.

The results of measuring the samples collected in August 1988 in rooms 207/5 and 427/2 serve as an example of the sharp increase in the concentration of radioactive aerosols at boring. When the borers were switched off, the concentrations of alpha-emitting aerosols in these rooms were 0.037 and 0.081 Bq/m^3, respectively. In the process of boring, the pollution level increased to 0.55 and 1.92 Bq/m^3, that is, by 15–20 times. At the same time, in the turbine island of the third unit, which is not connected with the "Shelter," the concentration was 0.0022 Bq/m^3.

Systematic aerosol sampling in the above rooms during the work conducted in 1988–1991 (Figure 6.15) provided important information. When the borers were switched off, the concentrations of the mixture of alpha-emitting aerosols were usually at the level of 0.03 Bq/m^3. On resuming the boring process, the amount of radioactive substances in the air sharply increased by one or two orders of magnitude. In some cases, the concentrations increased by three or even four orders of magnitude. In room 427/2, a concentration peak of 150 Bq/m^3 was recorded on 22 November 1988. Something like that occurred three years later, on 10 October 1991 in room 207/5, when the concentration increased rapidly to 90 Bq/m^3, and on the next day decreased by three orders of magnitude.

Summarized results of average annual concentrations of aerosols of alpha-emitting nuclides in 1989–1991 in seven main premises where boring was
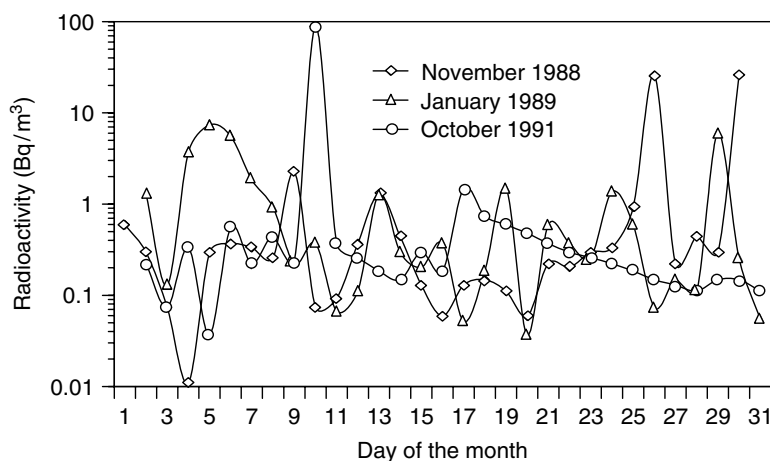

Figure 6.15 Concentration of aerosols containing alpha-emitting nuclides during well-boring activities.

Table 6.1 Average annual concentration of long-lived alpha-emitting aerosols in various areas of the ''Shelter'' (Bq/m^3) in 1989–1991.

Room number	1989	1990	1991 (first half)
207/4	0.53	0.35	0.093
207/5	0.32	0.36	0.11
208/10	0.32	–	0.15
318	0.74	0.21	0.1
427/2	0.92	0.15	0.093
515	0.20	0.26	0.067
605	0.21	–	–

performed are presented in Table 6.1. As noted in [65], only the days when active boring was going on were taken into account in the calculations. Figure 6.15 shows that the bursts of aerosol concentrations were not very long. However, they influenced average daily and even average annual values.

In May 1989, on completion of boring, it was necessary to remove a metal platform in room 208/10. By this time, a large amount of dust had accumulated on the floor, the walls, and the equipment. That is why aerosols rising in the air during metal cutting were supplemented with dust that appeared as a result of the movement of people and use of instruments. The dynamics of the concentrations of alpha-emitting aerosols are shown in Figure 6.16, from which it can be seen that the start of work was accompanied by a sharp increase in the concentration of radioactive aerosols in the air. On the first and second days of the work, the concentrations increased by two orders of magnitude from the background level of about 0.1 Bq/m^3. However, already 1 h after the completion of the work, air pollution considerably decreased, and within 8 h it practically reverted to the initial value.

After the central part of the ''Shelter'' including the reactor cavity had been examined using the wells, some wells were plugged and diagnostic equipment was placed in the others. On completion of intensive boring in 1992, the concentration of radioactive aerosols in decontaminated premises of the ''Shelter'' went on decreasing and in three years did not exceed 0.08 Bq/m^3 for alpha-emitting and 37 Bq/m^3 for beta-emitting substances.

6.3.4
Clearance of the Turbine Island of the Fourth Power Generating Unit

In the fall of 1988, the clearance of the turbine island of reactor 4 was started; in the process of construction of the ''Shelter'' it was separated from the turbine island of the third unit with a metal wall. Among operations with intense dust formation, we can distinguish removal of the roof accompanied by falling of building material fragments and clearance of the island by a ladle robot. The aerosol concentration
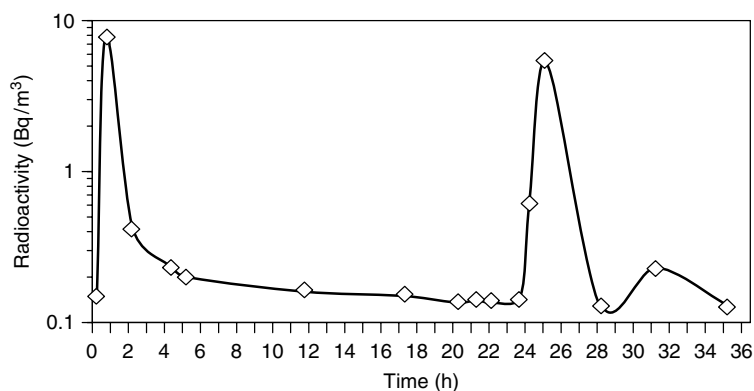
Figure 6.16 Concentration of aerosols containing
alpha-emitting nuclides during metal-cutting activities
in room 208/10 in the ''Shelter'' in May 1989.

in the air sometimes changed sharply within a short time. From 30 April to 25 May, operative control of aerosol concentration with simultaneous measurement of radioactivity of individual fractions was performed by specialists from the A.A. Bochvar All-Union Institute of Inorganic Materials [66]. A system consisting of a laser aerosol spectrometer (for optical determination of the size distribution of aerosols) and an impactor (to measure both the dispersity and radionuclide composition of individual fractions) was created. The results of observations showed that after carrying out the work the total aerosol concentration was increased from 3.6×10^4 to 20×10^4 particles per liter. Up to 70 particles per liter with the diameter of more than 7 μm appeared in the air, while there had been practically no such particles before the work started. On the completion of the work, the concentration of large aerosols (diameter of 1–10 μm) quickly (in about an hour) decreased by 5–10 times. The concentration of submicrometer particles (diameter less than 1 μm) decreased much more slowly – by two times within 2–3 h.

Dust control and treatment of surfaces with decontaminating solutions were performed to improve the aerosol and radiation situation in the turbine island. After that, the reduction of the number of particles larger than 5 μm and an increase in the concentration of fine aerosols up to 2×10^5 particles per liter were observed.

6.3.5
Strengthening of the Seats of Beams on the Roof of the ''Shelter''

In the fall of 1999, the bases under the beams B1/B2 were strengthened at the western wall of reactor 4 to prevent the collapse of the roof of the ''Shelter.'' These beams consist of double-T iron constructions B1 and B2; they are 3.2 m high and about 40 m long. On the beams, there is a roll of 27 tubes with 2 m diameter serving as the floor above the ruins of the central hall [67].

The specialists of the ''Shelter'' were controlling radiation, including the aerosol situation, on carrying out stabilizing work. Aerosols samples were collected during

the work on AFA RSP-20 filters with ''Typhoon'' portable blowers at two monitoring points (MP1 and MP2). Measurements of alpha- and beta-emitting nuclides were performed with a radiometer; the concentration of ^{137}Cs was calculated based on the results of gamma-spectrometry of samples.

In October to December 1999, preparatory operations were performed: cabling, lighting equipment assembly, transport and installation of welding equipment, and fastening of lead sheets to reduce irradiation of personnel. In addition, metal cutting and welding, clearing and slotting of concrete constructions with a perforator, and underpouring were carried out. Naturally, all the works were accompanied by dust rise and aerosol generation.

In the month and a half from 27 October to 13 December 1999, the unique data presented in Figure 6.17 were obtained regarding control of the radiation situation. The highest concentrations of ^{137}Cs (above 1000 Bq/m^3) were recorded during welding. For samples obtained near the bases of beams B1/B2, the aerosol composition depended on at least three factors: dust generation during working
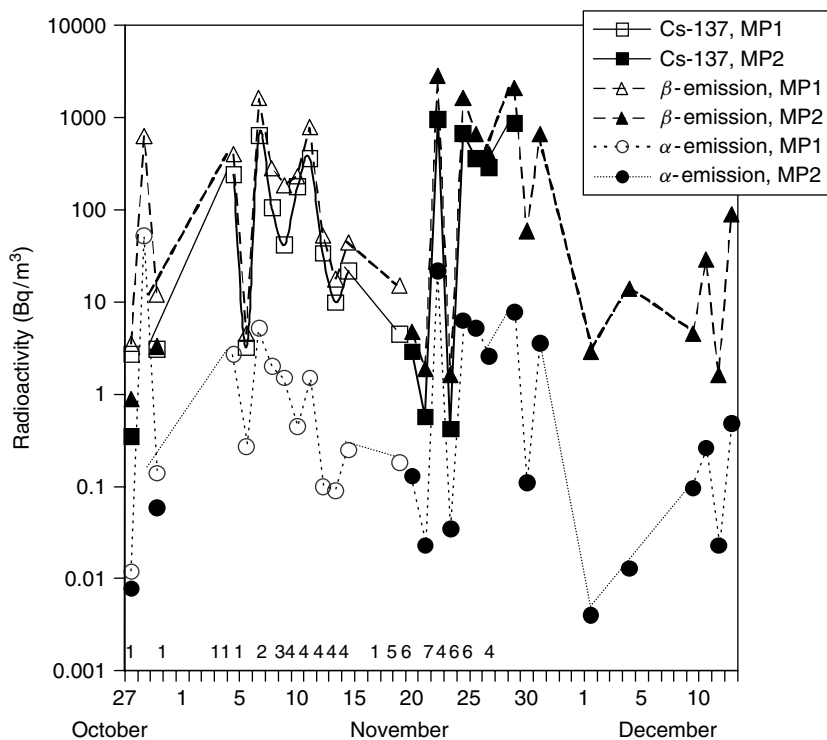

Figure 6.17 Concentration of radioactive aerosols carrying alpha- and beta-emitting nuclides, ^{137}Cs at two locations (MP1 and MP2) of the ''Shelter'' in 1999. Types of work are shown along the *x*-axis: 1, preparation; 2, mounting of welding equipment; 3, covering by lead sheets; 4, welding; 5, metal cutting; 6, hammering; and 7, mounting lights.

operation, air intake from the central hall of the ''Shelter,'' and air inflow from the free air. Naturally, under these conditions the ratios of radionuclides, in particular, ^{137}Cs and alpha-emitting nuclides, significantly changed, which can be seen in Figure 6.17. The aerosol situation during the work directly or indirectly also influenced other premises of the ''Shelter.''

6.3.6
Aerosols Generated during Fires in the ''Shelter''

Seven fires occurred on completion of the acute phase of the Chernobyl accident at the former reactor 4. The most dangerous ones were at the end of May 1986 in rooms 402/3 and 403/3, and in January 1993 in room 805/3 [68, 69]. Ignition in room 805/3 (2880 m^3 – air-duct of exhaust ventilation) caused complete burning of 1 m^3 of sleepers and 20 m of cables (the mass of burned material was 3500 kg). The average rate of flame spread over the cable surface was 0.5 m/min. The temperature in the fire zone exceeded 800 °C. Toxic and poisonous combustion products were released into the ambient air of the ''Shelter.''

The release of radioactive smoke from the ''Shelter'' occurred both through cracks in external constructions and through the pipe VP-2. The results of gamma-spectrometry on the collecting filter working in the system for control of air passing through ''Bypass'' showed that there was a sharp increase of radioactive aerosol concentration on 14 January (Figure 6.18). While on the preceding and subsequent days of January the daily release of the mixture of gamma-emitting nuclides varied in the range of 0.37−3.7 MBq/day, on the day of the fire it increased up to 33 MBq/day. As the air flow in the ''Bypass'' made up 25×10^3 m^3/h, the average daily concentration was 55 Bq/m^3. To calculate the average concentration
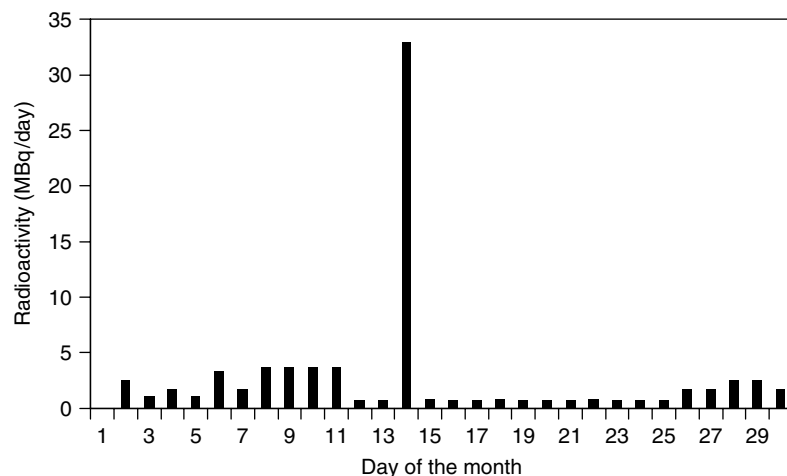

Figure 6.18 Daily emission of gamma-emitting aerosols for the ''Shelter'' in January 1993 (the fire event occurred on 14 January).

for the actual time of the fire, which lasted for about 6 h, this value should be increased by four times, that is, to 220 Bq/m^3. This is two orders of magnitude larger than the concentration of aerosols released on ordinary days.

The danger of fire from the point of view of the formation and carryover of radioactive products is also associated with the appearance of powerful convection currents. The motions of air masses that exceed the routine flows in their intensity and direction can emerge inside the ''Shelter.'' In addition, high temperatures can cause crumbling of different materials and the formation of *erosive aerosols*.

6.3.7
Dust Control System

The reduction of dust rise has always been a primary task when carrying out technological and research works in and around the ''Shelter.'' A complex series of activities involving the application of special polymeric materials on the building constructions and equipment was started after the commissioning of the ''Shelter'' to fix radioactive substances onto the surfaces. At first, manual sprayers were used for this purpose. The efficiency of the localizing action of the coverings was evaluated by taking smears from representative samples before and after film application. The determined activity decreased by 2–3 orders of magnitude.

A stationary dust control system intended for the application of coverings directly on the surface of the ruins in the former central hall of reactor 4 was created at the end of 1989. The materials were sprayed using 14 injectors inserted into the airspace above the ruins of the reactor building through roof hatches of the ''Shelter.'' However, spraying through the roof hatches only allowed less than a half of the roof space to be treated [70]. To increase the sprayed area, 35 additional injectors were installed in 2003. The aerosol concentration in the space of the former central hall increased more than once on testing the new system. This was caused by the fact that, before getting into the injectors, the solution forced air out of the collectors. Reaching the dry surface of the ruins, the compressed gas flows raised dust. The first drops hitting the surface and breaking into pieces also caused an inertial rise of dust particles. In 2004–2005, an 80–100 μm thick polymeric film was created on the surface of the ruins of the reactor building [70]. This considerably reduced the dust rise at the time of spraying solutions through injectors [71]. The system was put into commission in 2006. In accordance with the regulations, the film is replaced once a year.

6.3.8
Control of the Release of Radioactive Aerosols through the "Bypass" System

After the commissioning of the ''Shelter'' in November 1986, the specialists of the ChNPP Radiation Safety Service began to carry out scheduled observations of the release of radioactive aerosols from it using the ''Bypass.'' Aerosols were collected through 35 m pipes with the diameter of 15 mm onto LFS-2 filter tape located in the ''Kalina'' radiometer, and on AFA RMP-20 or AFA RSP-20 filters. The

Figure 6.19 Mounting of the aerosol sampling pump in the ''Bypass.''

imperfections in this system are discussed in [72]. It was shown that, depending on the dispersity of aerosols getting into the ''Bypass,'' the concentrations can be reduced down to 100%, especially for particles larger than 3 μm.

To get rid of uncertainties associated with aerosol deposition in the tubes, it was decided to perform aerosol sampling with a blower located immediately in the ''Bypass'' air flow (Figure 6.19). The specialists of the Institute for Safety Problems of NPP started such control in 2002. The blower with a pack of three-layer Petryanov filters sucked $10-12\,m^3$ of air within 2 h at a rate of about 0.8 m/s. On completion of the session, each filter layer was measured on radiometers, and the radionuclide composition, concentration, and dispersity of aerosols were determined. To provide isokineticity of aerosol sampling, a conical nozzle directed toward the flow was attached before the filter pack.

In 2002–2008, about 300 aerosol samples were collected in the ''Bypass.'' Figure 6.20 presents data on the activity concentration of the mixture of beta-emitting nuclide products of the accident $(\sum \beta)$ from September 2003 to December 2004. As follows from Figure 6.20, the $\sum \beta$ values were mainly in the
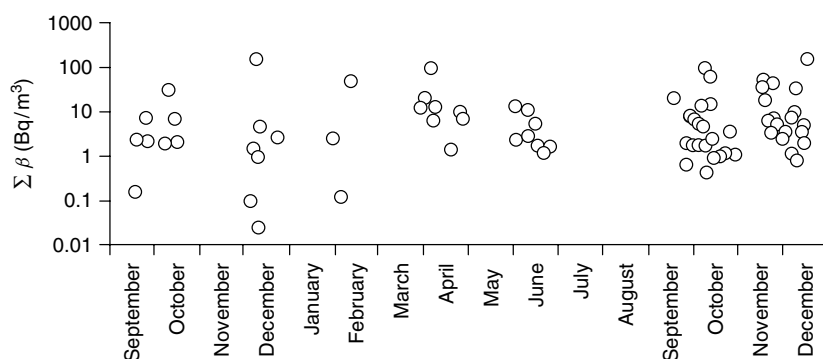

Figure 6.20 Concentration of radioactive aerosol containing a mixture of beta-emitting nuclides measured from September 2003 to December 2004 in the ''Bypass'' of the ''Shelter.''

range of $1-10$ Bq/m^3. About 30% of the activity was due to ^{137}Cs. The highest concentrations were conditioned by strong winds. For instance, on 8 December 2003 when $\sum \beta = 165$ Bq/m^3, the gusts amounted to $12-13$ m/s. In four months, on 5 April 2004, $\sum \beta = 110$ Bq/m^3 was recorded. This was preceded by three days ($1-3$ April) with gusts of $10-11$ m/s and one day (4 April) with the gusts of 8 m/s.

A considerable increase in the concentration of radioactive aerosol occurred after intensive technological activities within the premises of the ''Shelter,'' for example, as noted above, at spraying of polymeric solutions through injectors located under the light roof of the ''Shelter.'' For instance, on 11 February 2004, immediately on completion of the injectors' work, the average $\sum \beta$ within 2 h of sampling came to 164 Bq/m^3. Similar data were obtained during the subsequent years both at switching on injectors and for other works. For example, in September 2005 and in February 2006 on carrying out stabilizing measures in the southern part of the ''Shelter,'' including the welding and slotting works, not only were $\sum \beta$ values amounting to $500-1000$ Bq/m^3 observed but also aerosols with unusual staining (black and red).

Figure 6.21 presents $\sum \beta$ values for 2008 when the technological activities conducted in the premises of the ''Shelter'' were insignificant, most of the stabilizing works having been completed in $2006-2007$. The comparison of the data in Figures 6.20 and 6.21 shows that the $\sum \beta$ values again were mainly within the range of $1-10$ Bq/m^3: $\sum \beta$ exceeded 10 Bq/m^3 in only nine out of 72 samples collected in 2008. A strong wind was observed in the environment in all these cases either during sampling or a few hours before it. For instance, the largest value of $\sum \beta = 100$ Bq/m^3 was recorded on 8 April when, according to the data of the ''Chernobyl'' weather station, the gusts amounted to 14 m/s. Half a year later (22 September), after gusts of 10 m/s, $\sum \beta$ made up 34 Bq/m^3.

The effect of the wind on the aerosol situation inside the ''Shelter'' is one of the properties of this construction. It is conditioned by the presence of cracks and operational openings in its external construction. The effect of meteorological conditions on aerosol behavior in the ''Shelter'' is discussed in more detail in [73].
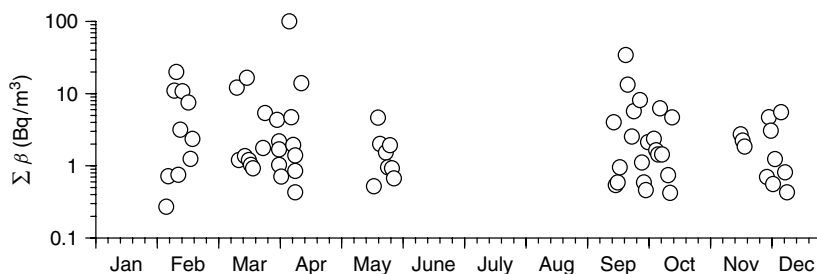

Figure 6.21 Concentration of radioactive aerosol containing a mixture of beta-emitting nuclides measured in 2008 in the ''Bypass'' of the ''Shelter.'' The results were obtained for the time intervals when no technological procedures were being performed.
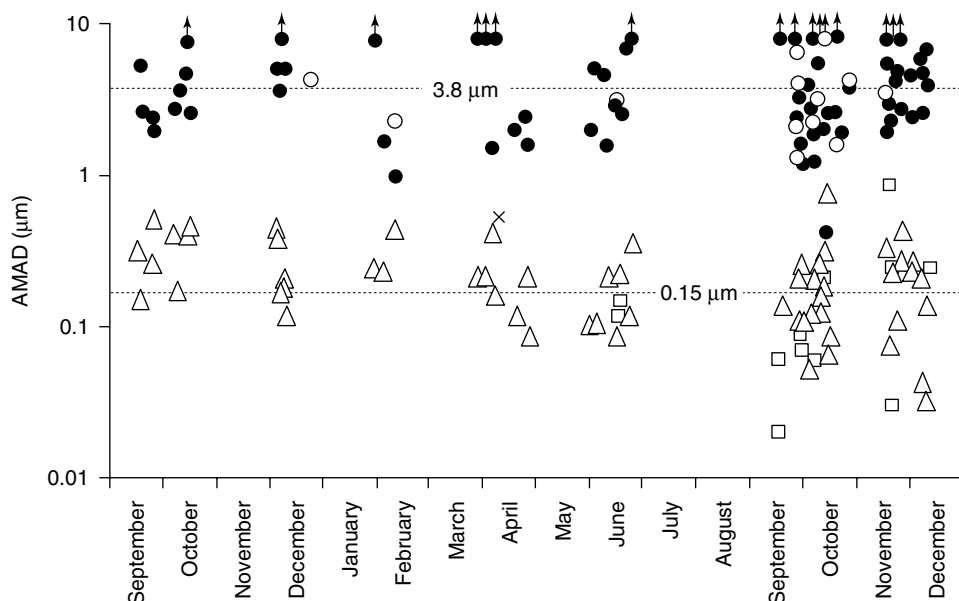
Figure 6.22 Size of radioactive aerosols measured in 2003–2004 in the ''Bypass'' of the ''Shelter.'' *Key*: beta-emitting aerosols aerosolized naturally by the wind (full circles) and during dust suppression activities (open circles); and DPs of radon and thoron containing aerosols aerosolized naturally by the wind (open triangles) and during dust suppression activities (open squares); AMAD larger than 8 µm (up-arrows).

In particular, it is noted that, in low winds and fog, the concentrations of aerosol products of the Chernobyl accident are usually low.

Processing of the results of measurements of the ''Bypass'' samples showed that aerosol carriers of radionuclide products of the Chernobyl accident usually had AMAD within the range of 1–10 µm at σ values from 1.1 to 3.5. Data for 2003–2004 are presented in Figure 6.22. For 80 selected samples, the average value was AMAD = 3.8 µm, and 67% of the samples had AMAD from 1.7 to 8.5 µm. Similar data were obtained in 2005–2008. From this it follows that carriers of radionuclides of Chernobyl genesis usually have a dispersion origin. No fractionation of radionuclides (^{90}Sr, ^{137}Cs, and ^{241}Am) by particles of different sizes was observed.

6.3.9
Radon, Thoron and their Daughter Products in the ''Shelter''

The DPs of radon (^{222}Rn) and thoron (^{220}Rn) take a special place among the radioactive aerosols that are present in the ''Shelter.'' They influence the radiation situation in the ''Shelter'' and the detection of radioactive aerosols of Chernobyl genesis. The two gases emanate from concrete constructions of both the former reactor 4 and new elements of the ''Shelter'' (the cascade and separating walls,

materials thrown onto the reactor from helicopters, and so on) containing the natural radionuclides ^{226}Ra and ^{232}Th. Radon and thoron also come from the soil (mainly sand) in which the foundations and lower levels of the ChNPP are embedded. A certain amount of thoron can appear from the irradiated fuel remaining in the ruins of reactor 4 as a result of the decay of ^{232}U formed during two years of reactor functioning before the accident. A chain of three successive alpha-decays of ^{232}U results in the appearance of thoron-generating ^{216}Po and ^{212}Pb. As the half-life of ^{232}U is 72 years, thoron will be generated from it for a long time.

If air containing radon and thoron is inhaled, their DPs present the greatest danger. First, some of them emit alpha-particles. Second, being on submicrometer aerosols, they penetrate into the lower sections of lungs, the bronchi and alveoli.

For radioactive aerosol monitoring in the ''Shelter,'' the DPs of radon and thoron are an interfering factor, because they complicate the radiometry of the samples. In 1987, this was noted by the specialists of the All-Union Instrument-Making Institute on the creation of the system for radiation control and diagnostics at the ''Shelter'' [74]. According to their data, the activity concentration of aerosols of DPs of natural radioactive gases in controlled premises on average made up 150 Bq/m^3. That is why, in determining the concentration of aerosol products of the Chernobyl accident, filters with collected samples should be kept for about 6 h so that the content of radon DPs has decreased by approximately 1000 times and for about 4 days for the same decrease in the amount of thoron DPs. However, there is also a positive thing: radon and thoron DPs present an original label of submicrometer aerosols. They can be used, for example, to evaluate the efficiency of work FPs and respirators.

For the first time, long-term observations of radon and thoron DPs were conducted in the ''Shelter'' in December 2000 [75]. Systematic samplings from the ventilation flow coming to the ''Bypass'' were started in 2002. Radon and thoron DPs were entrapped simultaneously with aerosol products of the Chernobyl accident in a pack of three-layer Petryanov filters. The activity concentrations of ^{212}Pb in 2002–2008 were within the range of 0.5–9 Bq/m^3 (see Figure 6.23). This is 50–100 times higher than estimates obtained in simultaneous samplings performed according to the same technique in March–May 2007 in the vicinity of the ''Shelter'' [76]. Thus, the thoron source was inside the ''Shelter.'' Consequently, ^{212}Pb was also generated inside the ''Shelter'' and did not come from outside with
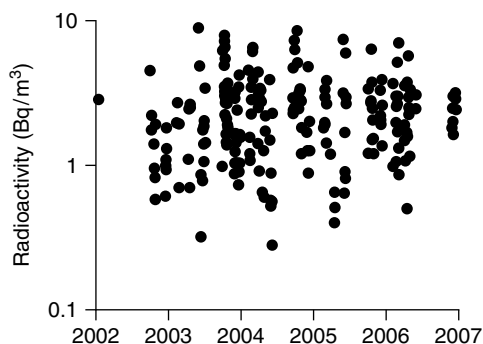

Figure 6.23 Concentration of radioactive aerosol containing ^{212}Pb in the ''Bypass'' of the ''Shelter.''

external air. This is confirmed by the data obtained in February–March 2008 in rooms 207/4 and 318/2 of the ''Shelter'' by the employees of the Department of Radiation Technologies, Materials Science and Environmental Research of the Institute for Safety Problems of NPP. They revealed that ^{212}Pb concentrations made up about 2 Bq/m^3 [77].

Aerosol particles with AMAD of 0.05–0.4 μm (see Figure 6.22) are carriers of radon and thoron DPs both in the ''Bypass'' and in the ''Shelter'' [62, 76]. In accordance with Figure 6.22, the average value is AMAD = 0.15 μm, which is typical of carriers of radon and thoron DPs in other regions both outdoors and indoors [78]. This is associated with the fact that carriers of radon and thoron DPs usually have a condensation origin.

The studies in some parts of the ''Shelter'' showed that radon and thoron DPs, like the parent substances themselves, can accumulate when there is no longer an air draft through the ''Bypass.'' This usually occurs in the second half of spring when in the day time the air temperature in the vicinity of the ChNPP amounts to 30 °C, and non-heated premises inside the ''Shelter'' still remain cold after winter. Such situations were recorded in room 207/5 during the last 10 days of May 2003 and on 23–24 May 2007. In the first case, ^{212}Pb concentration amounted to 15 Bq/m^3, and in the second to 12–13 Bq/m^3 [76].

The detection of radon concentration exceeding 100 Bq/m^3 and thoron concentration higher than 6 Bq/m^3 in premises of the ''Shelter'' is a negative factor, which has not been taken into account previously in monitoring the radiation situation. Owing to inhalation of aerosol carriers of the DPs of these noble gases, the average annual radiation dose for the personnel of the ''Shelter'' can amount to tens of percent of the maximal equivalent dose of 20 mSv/year.

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