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6 Environmental radionuclides as tracers for transport processes in snow

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Abstract

Radionuclides are useful tracers for the determination of transport processes from the atmosphere to the hydrosphere by snowfall. Particularly in high altitudinal regions radionuclides released to the environment are efficiently removed from the atmosphere by snow. Once deposited, they will be concentrated in the snow cover due to evaporation and sublimation as long as ambient temperature stays low and no melt water runoff occurs. Short-term releases to surface waters after snowmelt initiation in spring may lead to high concentrations having an impact on water quality.

To trace radionuclide transport in the aquatic environments related to snow on Mt. Zugspitze, in addition to their concentrations, comprehensive knowledge about the type of precipitation and the size of the aerosols, they are bound at, the snow cover development, the influence of meteorological conditions on snow alteration and melt water runoff are necessary. A brief description of methods for the investigation of the transport of ubiquitously distributed environmental radionuclides from their atmospheric deposition over their behaviour in the snow cover at Zugspitzplatt to the recovery in the surface water at Partnach spring is given. Applications performed in research projects of the authors' work group since 2011 on Mt. Zugspitze are introduced and highlight the advantages of the location for studying water and radionuclide budgets related to snow. The results help to trace the pathways of radioactive particles from the atmosphere to aquatic environments. Consequently, in cases of extensive radionuclide releases to snow covered environments, peak discharges and the scope of action for countermeasures can be predicted to mitigate the impact on water quality and human radiation exposure.

Keywords: radionuclide tracer, snowmelt, wet deposition, radionuclide transport, snow characterisation, scavenging coefficient, meltwater runoff, gamma spectrometry

6.1 Introduction

Ionising radiation originates from decay of unstable nuclei, either in primordial, natural or man-made radioactive material, e.g. in nuclear reactors and accelerators for energy production or radiotherapy. In general, the exposure of human beings to ionising radiation released to the environment in high concentrations has negative effects on human health. Radiation exposure may damage or modify living cells leading to harm to organs or cancer (UNSCEAR 2000). The release of radioactivity after the nuclear accident of Chernobyl and Fukushima showed that trace contaminants can be rapidly transported world-wide within a few days. Once released to the atmosphere, radionuclides will be rapidly attached to aerosol particles. These are most effectively removed from the atmosphere by wet deposition. For snow, the collection efficiency is strongly influenced by the crystal type and the surface structure of the hydrometeors (Tschiersch et al. 2000, Kyrö et al. 2009, Paramonov et al. 2011). The more complex the crystals and the larger the surface area related to their volumes, the more efficient is the washout (Bernauer 2015).

Once deposited with (wet deposition) and onto (wet or dry deposition) snow, radionuclides are continuously concentrated during the winter due to evaporation and sublimation as long as no

meltwater runoff at the snow base occurs (Hürkamp et al. 2017, Marin et al. 2020). In high-altitudinal regions, soils that generally act as filters or sinks for contaminants are missing. Consequently, stored amounts of radionuclides can be directly released into surface and drinking water reservoirs shortly after snowmelt initiation in spring (Fig. 1). They are able to cause severe impacts on the water quality and provide a risk for radiation exposure to humans.

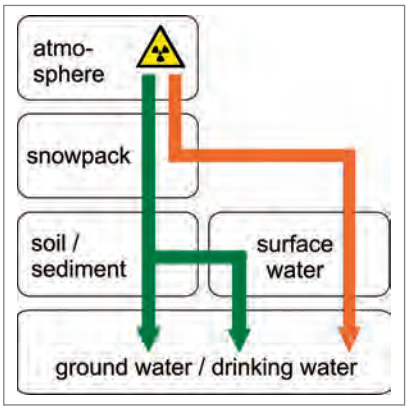

Fig. 1: Pathways of radionuclide transport from the atmosphere to the hydrosphere. Green arrows trace common lowland paths. In high altitudinal or permafrost regions, these are redirected in a way (orange arrow) that surface waters may be directly contaminated due to absence of soils that could act as filters or sinks.

The wet deposition of aerosol-bound radionuclides is dominated by capture processes of falling hydrometeors, called below-cloud scavenging. The major contributors for the collection efficiency are the mechanisms of Brownian diffusion (~ 10 nm), interception and impaction (~1 µm), depending on the aerosol size. Aerosol particles with sizes inbetween are less efficiently scavenged (Sportisse 2007).

The transport through the snow cover is highly dependent on its thickness, snow texture and density and still to a higher degree on meteorological conditions. Radionuclide transport is associated with the percolation of liquid water in the snowpack (Hürkamp et al. 2017). Consequently, as long as ambient temperature stays below the freezing point, no meltwater flow will occur. After snow ripening, radionuclides follow the downward penetration of water front through the snowpack. Inhomogeneities in snow densities, ice lenses or rain-on-snow events could lead to the formation of preferential flow paths, which favour the transport in some areas. Still, meltwater runoff and radionuclide release to soils and surface waters is delayed as long as the snow base layer is not fully water saturated.

Depending on the subsurface composition below the snow cover, radionuclide transport to surface or ground waters is highly variable. It can be rapidly stopped when meltwater is drained by soils or sediments. In many substrates, dissolved or particulate-bound radionuclides are immediately fixed to the grain surfaces. In the case of solid bedrock or frozen ground, meltwater discharge is constrained and turns into surface water. On Mt. Zugspitze, the snowpack covers “Wettersteinkalk” limestones at the so-called Zugspitzplatt. Due to the absence of soil covers, meltwater directly enters a karstic aquifer and is drained subsurface until it flows out again at the Partnach spring. Therefore, Mt. Zugspitze and the adjacent Zugspitzplatt are outstanding locations to study mass transport in snow and surface waters. In chapter 2, the advantages of the study site are discussed. Chapter 3 describes the applied methods for the investigation of the deposition and transport of environmental radionuclides from the atmosphere via snow to melt and surface water. In chapter 4 exemplary applications from research projects of the Institute of Radiation Medicine, Helmholtz Zentrum München since 2011 concerning radionuclide deposition, their behaviour in the snowpack, transport and release to surface water are introduced.

The environmental radionuclides, which are omnipresent in surface air and precipitation on Mt. Zugspitze, albeit in very low concentrations, are of natural and artificial origin. The highest activities in air and fresh precipitation can be measured for Be-7. Be-7 is a cosmogenic isotope with a half-life of 53.2 d. Its production mainly takes place in the upper troposphere and lower stratosphere by interactions of cosmic rays with atmospheric nitrogen and oxygen. Be-7 atoms are attached to atmospheric aerosol particles soon after their formation. With the advantage of the short half-life and relatively easy determination by gamma-spectrometry, Be-7 has been widely used as a tracer in atmospheric science (e. g. Winkler et al. 1998, Ioannidou and Paatero 2014).

The terrestrial radionuclide Pb-210 is a progeny of Rn-222. The main source of atmospheric Pb-210 is the emanation of short lived Rn-222 from rocks and sediments. Pb-210 atoms become quickly attached to aerosol particles after their formation in the planetary boundary layer. Therefore, they are associated with the aerosol population and allow the investigation of tropospheric transport and removal processes. Pb-210 is widely used in atmospheric modeling and sediment dating (e. g. Winkler et al. 1998, Putyrskaya et al. 2015). It has a half-life of 22.2 a.

Cs-137 has a half-life of 30.1 a and is produced anthropogenically by several types of nuclear activities including past testing of nuclear weapons, accidents in nuclear facilities, reprocessing

of spent nuclear fuel and nuclear power plants. Recently measured activities of Cs-137 in central and northern Europe in air and precipitation are mainly ascribed to the fallout of the Chernobyl nuclear accident in April 1986 (Kulan 2006) and related subsequent resuspension processes (Garger et al. 1998). The forest topsoils in the Bavarian Alps and in the surrounding valleys of Mt. Zugspitze are still highly contaminated (Winkelbauer et al. 2012).

6.2 Study Site

The investigations on snow deposition and newly fallen snow were carried out on the terrace of the Environmental Research Station (UFS) Schneefernerhaus (2650 m a.s.l.) (Bernauer 2015, Bernauer et al. 2015, 2016). It is situated at the steep southern slope of Germany's highest mountain Mt. Zugspitze (2962 m a.s.l.). The devices for continuous measurement of aerosol concentration, airborne radionuclide concentration and characterisation of precipitation events were deployed here.

The adjacent Zugspitzplatt is a karstic plateau at 1500–2800 m of altitude (mean 2229 m a.s.l.). The snow measuring field (47°24'22.75"N, 11°59'00.39"E) with instrumentation for meteorological and snow parameters is situated at this plateau; where the the annual snow profiles are taken. It is composed of 600–800 m thick bedded limestones ('Wettersteinkalk', Ladin) with a slightly undulating relief. It is horse-shoe shaped surrounded by several summits and inclined to the east (Miller 1962). The underlying aquiclude consists of marly claystone ('Partnach-Schichten').

At the eastern margin of the 11.4 km^2 catchment, surface water that usually instantaneously infiltrates and drains through the karst aquifer, discharges at the Partnach spring (1440 m a.s.l.). At the Partnach gauge (1365 m a.s.l., 47°24'18.75"N, 11°02'03.07"E) 200 m downstream, discharge is continuously recorded (Fig. 2). Rappl et al. (2010) showed that the basin is almost hydrologically enclosed. No discharge occurs except for the leakage at Partnach spring. This emphasizes the suitability of the catchment for balancing water and radionuclide budgets, since it can be seen as a large natural lysimeter. Discharge velocities for the Zugspitzplatt karst were estimated with 130–170 m s^{-1}, even though during a summer storm event (Rappl et al. 2010).

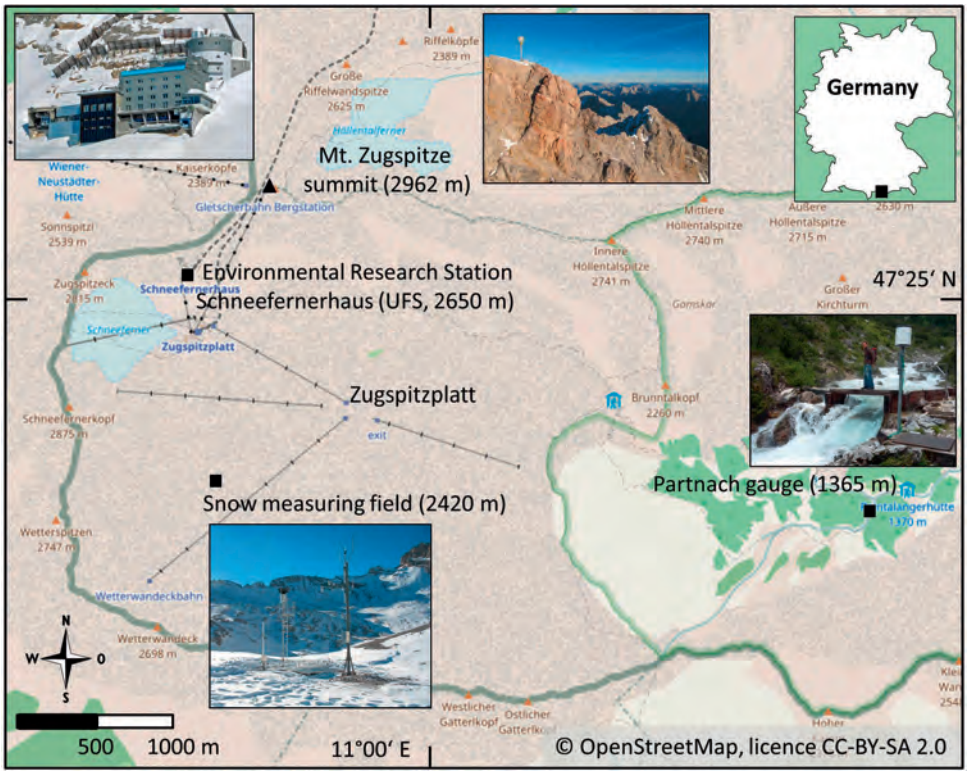

Fig. 2: Study area at Zugspitzplatt with the location of the measuring sites (© OpenStreetMap, Licence CC-BY-SA 2.0, photos: K. Hürkamp).

Wetzel (2004) analysed hydrographs of Partnach spring in 1996 and 1997, and found a diurnal cycle of snowmelt water dominating the runoff during the ablation period and glacier melt influences after the ablation period, both superposed by storm events.

Average annual temperature on Mt. Zugspitze summit (2962 m a.s.l.) is –4.3 °C (1981–2010). The average annual precipitation is 2071 mm, mainly snow at 58 % of all days and 90 % of the precipitation days per year (DWD 2017). Continuous snow coverage exists at 150–300 d a year on Zugspitzplatt, depending on the altitude that increases to the northern and southern margins and to the west (Weber et al. 2016). The region is strongly wind exposed and experiences intensive solar radiation.

6.3 Materials and Methods

Specific methods and procedures were applied for sampling, analysis and parameterisation in the studies of transport processes of environmental radionuclides with snow. They were adapted to the unique high alpine measurement situation.

6.3.1 Snow sampling: fresh snow and annual profiles

Deposited amounts of radionuclides on and with snow can be determined by the analysis of integrated depth snow samples. For this, a continuously accumulating snow cover during the winter months without meltwater runoff at the snow base at the upper Zugspitzplatt offers best possibilities. Either the frequent sampling of fresh snow or the excavation and sampling of snow profiles at the end of the accumulation period in order to determine the total accumulated radionuclide inventory of the entire season are adequate methods.

An approximately 25 × 25 m wide area at the measuring field on central Zugspitzplatt surrounding the meteorological station is fenced each year to protect the natural snow cover against ski tourism. The area is used by the staff of the Bavarian Avalanche Warning Service to dig bimonthly snow profiles in order to generate parameters used for their daily avalanche warning reports. During the winters 2014/2015 and 2015/2016 also monthly fresh snow and seasonal accumulated samples from profiles over the total snow depth for the determination of the environmental radionuclide inventory were taken.

For the monthly fresh snow samples, the freshly fallen snow since the last sampling campaign was taken. The snow surface was therefore marked with a mesh after each sampling day. The samples were taken from an area of a pre-defined size (activity referred to area) over the total depth of the newly accumulated snow cover since the last sampling date.

At the end of the snow accumulation periods 2015 and 2016, integrated depth snow samples of each snow layer were taken horizontally from the profiles. For the gamma-spectrometric determination of the radionuclide activities of Be-7, Pb-210 and Cs-137 at existing environmental concentrations on Mt. Zugspitze, at least 5 L of SWE are necessary to reach the detection limits. Depending on the snow density, canisters with volumes of 20 L and 60 L barrels were filled with snow and used for the transport of the samples to the laboratory at Helmholtz Zentrum München. Preconditioned plastic containers and shovel were used for sampling: They were acidified with HNO_3 (5 mL HNO_3 conc. per L) to avoid radionuclide fixation to the walls of the barrels, canisters or glass beakers during pre-treatment. In addition, the acid prevents the growth of microorganisms, which could lead to radionuclide sorption and inhomogeneous mass distributions in the sample causing higher uncertainties in the measurement.

The seasonal profiles were excavated up to the depth of the solid bedrock and snow characteristics described in detail according to Fierz et al. (2009). Snow layers were distinguished due to differences in snow texture, grain size and roundness, hardness and water content of the layers (Fig. 3). Snow densities of each layer were determined *in situ* by use of a metal tube of known volume and a spring scale to measure the sample weight. Snow water equivalents (SWE) were calculated by multiplication of the density with the snow height. SWE are necessary to determine the activities of radionuclides deposited in the area of the total catchment of the Zugspitzplatt (11.4 km^2, Wetzel 2004). To trace the contents of radioactivity in the snow profiles

Fig. 3: Snow profile description and sampling at the end of the snow accumulation periods 2015 and 2016. In the profile of June 2016 (right figure) a prominent Saharan dust layer is visible, which allows dating the snow layer to an event that occurred on April, 5th/6th and which could be traced in the snow alpine-wide (Photos: E.-M. Schiestl, 2015-05-13 and K. Hürkamp, 2016-06-01).

over time, SWE are additionally needed to relocate snow layers of the same age. Due to evaporation, melt and compaction processes, snow height cannot be used to correlate coeval snow layers over the total season.

6.3.2 Surface water sampling at Partnach spring

About 200 m downstream the Partnach spring (1440 m a.s.l.) an automatic gauge and water sampling station is installed (Fig. 4). Precipitation, fluvial discharge, water temperature and electric conductivity are continuously recorded (Wetzel et al. this issue).

Samples of the Partnach surface water were taken irregularly between May and October 2015 and 2016 as grab samples of 20 L volume or as bulk samples from an automatic sampler which is operated in cooperation with the work group of Wetzel et al. (this issue). The automatic sampler is equipped with 24 × 1 L bottles and is programmable for the sampling period and amount of water pumped per sample. During the sampling campaigns in 2015/2016 the programmes were changed related to the expected time interval between the sampling days, ranging from three-hourly to daily sampling intervals for aliquots of 500 mL surface water each. At the sampling days, always all 24 bottles were merged to one bulk sample and transported in canisters to the laboratory.

Fig. 4: Partnach gauge 200 m downstream the Partnach spring. The station is equipped with sensors for the determination of surface water discharge parameters (left) and an automatic water sampler (right). In addition grab surface water samples were taken (middle) during the summers 2015 and 2016 (Photos: K. Hürkamp, 2016-07-12).

6.3.3 Gamma-spectrometry for radionuclide quantification

The snow and water samples are transported in barrels or canisters to the laboratory at Helmholtz Zentrum München, where they are weighted, and if necessary melted at room temperature. To concentrate the rather low environmental radionuclide concentrations in the snow and surface water of Mt. Zugspitze, and to reduce the sample volumes to measurable amounts, the liquid samples are evaporated by use of a rotational evaporator. Residual samples, that contain the total radionuclide inventory of non-volatile elements, are subsequently evaporated to about 100–200 mL on a heating plate at maximum temperatures of 80 °C and transferred to 250 ml polyethylene containers. The containers have defined geometries which are calibrated for the subsequent gamma-spectrometry measurements. Therefore, a defined filling level must be achieved to reproduce the same geometry as in the calibrated standard. To reach this level, samples will be filled up with deionised water.

Samples are measured by gamma spectrometry with high purity germanium detectors (HPGe, Gilmore 2008). Gamma radiation is an electromagnetic radiation consisting of gamma quants (= photons). Gamma quants do not carry an electric charge. Therefore, the interaction of gamma radiation with matter differs from that of charged particles with matter. The energy absorption is possible by the photo effect, the Compton effect, and pair production. All these effects cause the production of secondary electrons that are detected (as charged particles) in appropriate measurement devices. The operation of germanium detectors follows the interaction of gamma rays with a semi-conductor. Entered photons interact with the germanium crystal and excite electrons to the conduction band causing formation of electron-hole pairs. The number of electron-hole pairs is directly proportional to the energy absorbed in the material. The electric signal is amplified, sorted by height and energy and displayed in a typical gamma-spectrum (Fig. 5). The charges are collected to electrodes. Thermal electrons mask all weak signals. Therefore, germanium detectors have to be cooled down by liquid nitrogen or electronic cooling systems (Fig. 6).

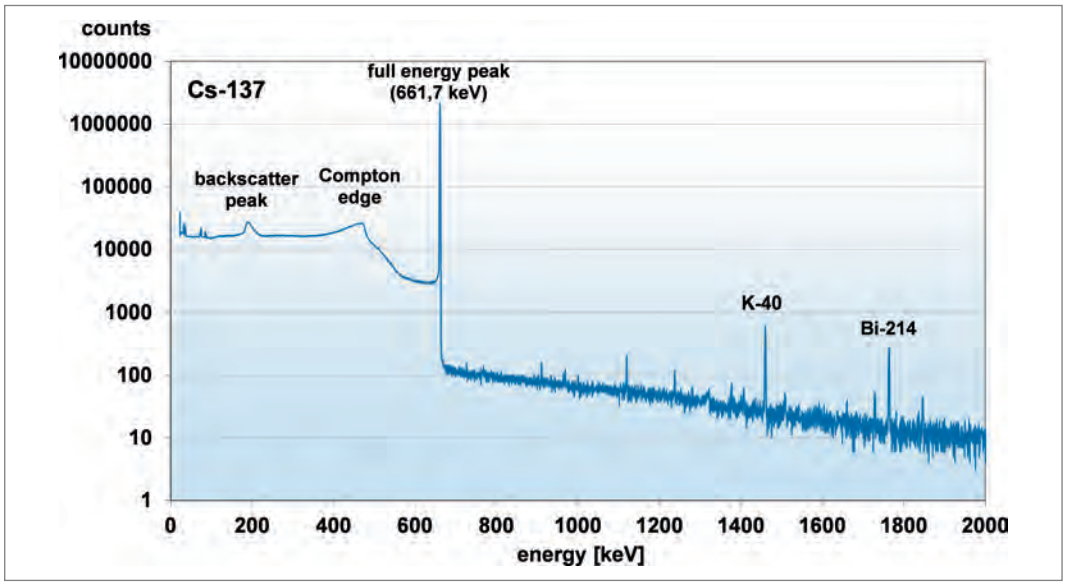

Fig. 5: Pulse-height gamma-spectrum for the discrete gamma energy of Cs-137, measured on a HPGe coaxial detector (rel. efficiency 40 %, resolution 1.9 keV (FWHM) at 1332 keV) in the radioanalytical laboratory at Helmholtz Zentrum München. At the full photon energy of 661.7 keV, the photopeak appears. The gamma quant is completely absorbed by a shell electron of the detector material. The photon vanishes and a free electron that carries the same energy is released. If the photon does not lose its entire energy, only an elastic collision with a shell electron of the detector material occurs (Compton effect) and the photon is scattered carrying still a reduced energy. The maximum energy of the Compton effect corresponds to the Compton edge in the pulse height spectrum. At lower energies, the spectrum continues with a plateau, the so-called Compton continuum. Photons may also pass the detector without any interaction and re-enter the detector with reduced energy after being backscattered in the surrounding material. The primordial K-40 and other radionuclides originating from the natural terrestrial decay chains (e.g., Bi-214, Pb-214) are ubiquitously distributed in the air and contribute to the background radiation.

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Activities for the environmental radionuclides Be-7, Pb-210 and Cs-137 are calculated integrating the areas of the full energy peaks in the gamma spectra at energies of 477.6 keV for Be-7, at 46.5 keV for Pb-210 and at 661.7 keV for Cs-137. The net area of the peak is directly related to the intensity, but it is also necessary to correct for the efficiency of the detector (efficiency calibration is needed for each geometry and detector), the half-life of the nuclide and the respected background. The background mainly consists of natural radiation (e. g. cosmic and terrestrial radiation). The component is minimised by using 10 cm lead shielding enclosing the detector at all sides.

Still some samples are analysed by a low-level background gamma detector located at the Environmental Research Station Schneefernerhaus (Fig. 7). It is equipped with an active veto shield (Burnett and Davies 2012) placed on top of the lead shielding. It consists of plastic scintillators which detect and eliminate any signal of the strong (due to high altitude) incoming cosmic radiation and therefore additionally reduces the background by a factor of 1.9–2.2. A background correction due to additional contribution of terrestrial radiation from the radon decay chain has to be applied for the determination of Pb-210 in the snow and surface water samples. For this, a measurement in an empty lead shielding chamber is carried out bimonthly. The resulting peak areas of the radionuclides of interest are subtracted from the photopeak areas for each subsequent sample measurement.

Mean detection limits for the gamma-spectrometric activity determination depend on the measurement times and were about 30 mBq L^{-1} for Be-7, 50–100 mBq L^{-1} for Pb-210 and 2 mBq L^{-1} for Cs-137 in snow and surface water at maximum acceptable measurement times of 14 d. Measurement uncertainties of ≤ 5 % for Be-7, ~20 % for Pb-210 in snow and ~20 % for Cs-137 are achieved in surface water, even though they are much higher for Be-7 and Pb-210 in surface water and for Cs-137 in snow. Additional overall analytical errors sum up to 3 %. To drop the uncertainties and detection limits, higher initial amounts of SWE or longer measurement times would be necessary. Quality assurance measurements concerning energy calibration, accuracy and reproducibility are monthly carried out with certified liquid multi-nuclide standard materials or calibration point sources of selected radionuclides (Cs-137, Co-60, Am-241) with known activities. The liquid standard, especially used for efficiency and energy calibration, contains eleven different radionuclides in the energy range of 40 keV to 2000 keV to cover the whole range of interest.

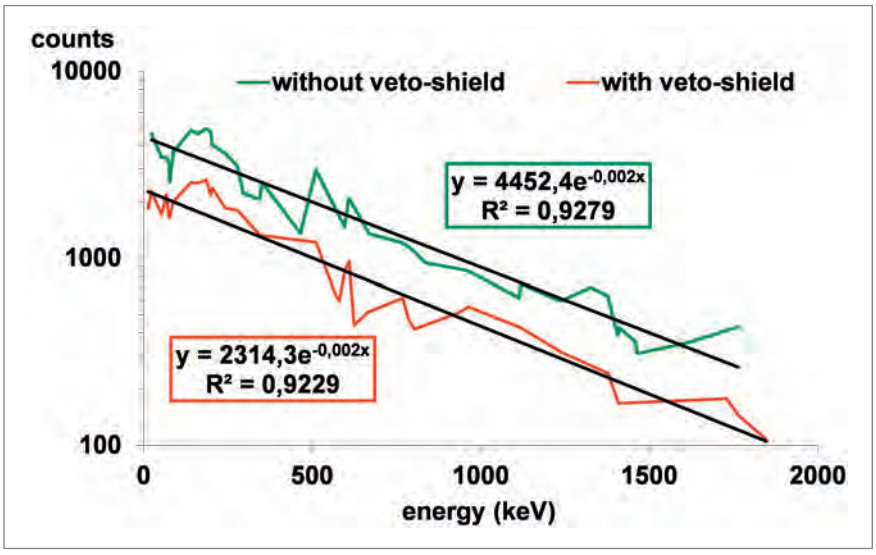

Fig. 6: HPGe broad energy gamma spectrometer system (left) at the Environmental Research Station Schneefernerhaus (Photo: K. Hürkamp, 2011-10-19). Detector cooling is performed with an electronically powered cryostat. An active veto shield (plastic scintillators) on top of the lead shielding provides additional reduction of the background due to elimination of altitude-induced enhanced cosmic radiation by a factor of 1.9 to 2.2. The right figure shows the measured background spectra with and without active veto-shield.

6.3.4 Snow Characterisation

6.3.4.1 Characterisation of hydrometeors

For the determination of washout efficiencies for aerosol-bound radionuclides with different precipitation events, the type of precipitation can be characterised by use of a 2D video disdrometer (2DVD, Fig. 7a). It consists of two line scan cameras that are aligned in a 90° angle to each other and a height difference of 6 mm (Fig. 7b). The cameras scan every hydrometeor falling through the sensitive area. From the shaded images that are matched for both cameras for each snowflake or raindrop (Bernauer 2015, Bernauer et al. 2015), crystal sizes and shapes are derived. The height difference of the cameras gives evidence about fall velocities. From typical distributions in size-fall velocity diagrams (Fig. 7c), falling hydrometeors can be classified into rain, snow and mixed events due to the fact that raindrops fall faster than snow and snowflakes have bigger sizes. Considering the shape of the hydrometeors, additionally three snow crystal classes, single crystals (needles, dendrites, plates), complex crystals (wet flakes and aggregates) and pellets (graupel, densely rimed rounded crystals) are distinguishable (Fig. 7d, Bernauer et al. 2016).

6.3.4.2 Scavenging coefficients

The characteristic measure for the efficiency of wet deposition is the scavenging coefficient. It can be determined from the decreasing aerosol particle concentration during precipitation. The particle concentration $c(d_p, t)$ is a function of its diameter d_p and time t. Under the assumption that precipitation scavenging is a dominant sink for $c(d_p, t)$ a first order decay equation can be set up:

$$\frac{\partial c\,(d_p, t)}{\partial t} = -\Lambda(d_p)\; c(d_p, t)$$

Integration yields the scavenging coefficient $\Lambda\,(d_p)$:

$$\Lambda(d_p) = -\frac{1}{t_1 - t_0} \ln\left|\frac{c(d_p, t_1)}{c(d_p, t_0)}\right|$$

for measurements at t_0 and t_1 (Laakso et al. 2003, Kyrö et al. 2009). Another possibility is to rearrange the equation to

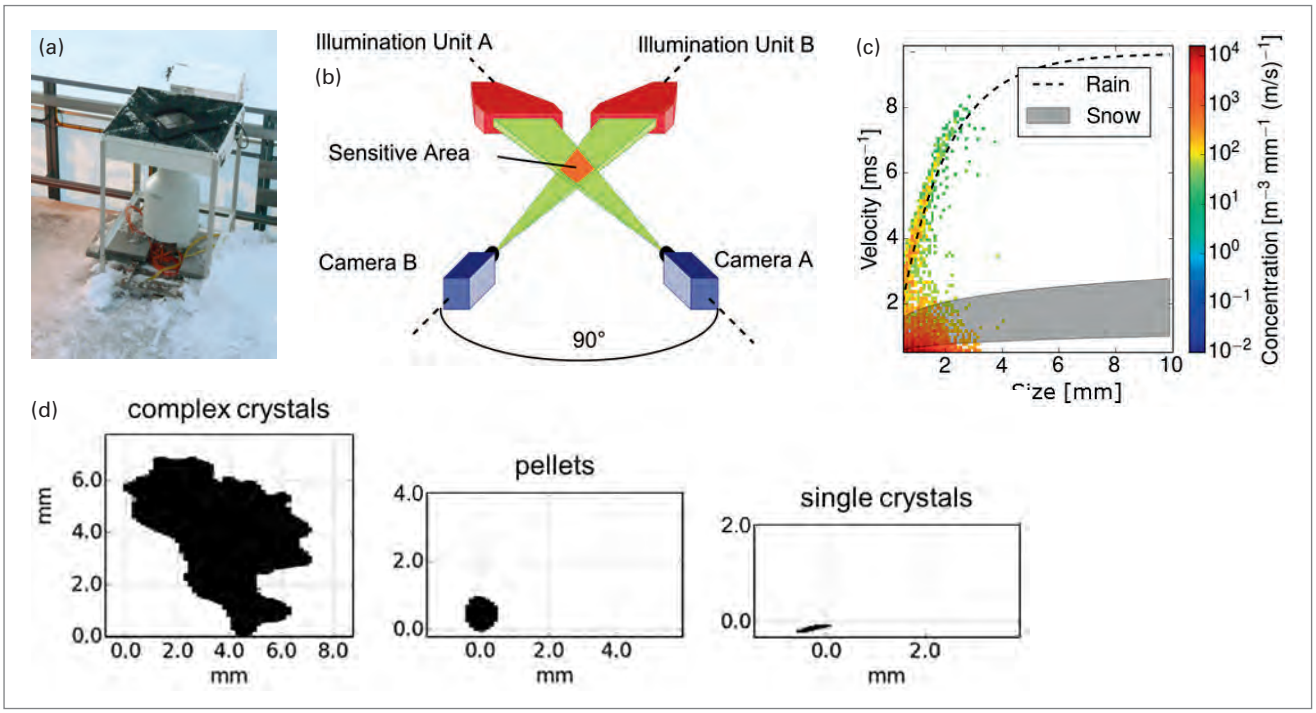

Fig. 7: (a) 2D video disdrometer (2DVD) for the characterisation of precipitation events (Photo: K. Hürkamp, 2013-11-14). (b) Measurement principle. (c) Particle size-fall velocity diagram for the differentiation between rain and snow events. (d) Scanned images of the 2DVD with examples for the three distinguishable snow crystal classes (Fig. c–d modified according to Bernauer 2015).

$$\ln(c(d_p, t)) = -\Lambda(d_p)\, t + \ln(c(d_p, 0)).$$

It shows that $\Lambda(d_p)$ is the slope of a linear fit to the logarithm of the aerosol particle concentration.

For the calculation of scavenging coefficients the aerosol particle concentration in the atmosphere is measured by a Scanning Mobility Particle Sizer (Birmili et al. 2016) before and during a precipitation event. Concentrations are subdivided into 14 size classes from 10–510 nm to cover the whole range of ultrafine aerosol particles that are predominantly absorbed in human lungs and may cause adverse health effects. From three consecutive 10 min-measurements of the size-related aerosol particle concentrations, their decrease is determined and expressed as washout or scavenging coefficient. The scavenging coefficients, calculated for snow events are useful input parameters to improve and regionalise atmospheric radionuclide distribution models in European decision support systems for the emergency management after nuclear or radiological accidents, in which only rain is considered so far.

6.3.4.3 Snow parameters for the observation of the snow cover development at Zugspitzplatt

Snow parameters are continuously recorded since 11/2012 at the central Zugspitzplatt (2420 m a.s.l.), where a meteorological station is additionally equipped with an ultrasonic snow depth sensor, a snow scale and a Snow Pack Analyser (SPA) for the for the determination of snow water equivalents (SWE), snow density, liquid water and ice contents in different heights of the snowpack (10 cm, 30 cm and 50 cm above rock 11/2012-09/ 2017, 10 cm, 20 cm and 30 cm height since 12/2017, Fig. 8). The sensors record and transmit (wireless transmission to a data storage server) data for each parameter every 10 min.

The snow scale measures the SWE for solid precipitation more precisely than precipitation gauges (Błaś et al. 2010, Lundberg et al. 2016). The measurement is carried out on a centre aluminium plate, surrounded by stabilising plates (on an area of 2.4×2.8 m) in order to compensate stress in the snowpack as well as to counteract the problem of ice bridges through the large surface area. Perforation of the plates prevents water accumulation, minimises the difference in temperature between the scale and the ground and promotes uniform melting.

The SPA (Stähli et al. 2004) measures the volumetric contents of ice, water and air and calculates the SWE and snow density when combined with a snow depth sensor. It consists of three 5 m weather and UV-resistant sensor bands that penetrate the snow in 10 cm, 30 cm and 50 cm above ground measuring the complex impedance. Snow consists of ice, air and water. They have different dielectric constants. Each band sends out measuring frequencies and is able to calculate the percentage of liquid water, ice and the remaining value as air from the returned value. The dielectric constant of ice has a strong frequency variation, which enables determination of the volumetric content of both frozen and liquid water. The band installed horizontally at 10 cm above the ground gives valuable information about beginning meltwater runoff.

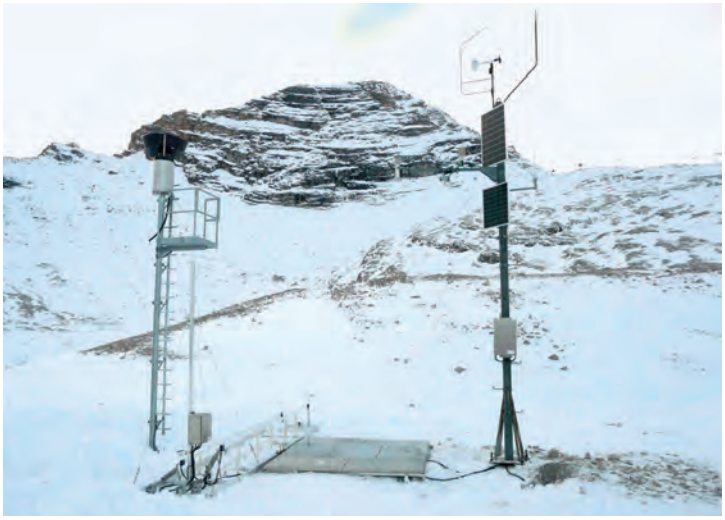

Fig. 8: Meteorological station at central Zugspitzplatt (2420 m a.s.l.), operated by the Bavarian Avalanche Warning Service and the authors on behalf of the Environmental Research Station Schneefernerhaus. It is equipped with sensors for the determination of standard meteorological and snow parameters. The measurement data is recorded and transferred to the Alpine Environmental Data Analysis Center (AlpEnDAC, www.alpendac.eu), where it is publicly available (Photo: F. Bernauer, 2012-11-08).

Percentages of about 3–4 %, up to 7–8 % liquid water content indicate full water saturation of the basal snow layer (Denoth 2003, Heggli 2013). Generally, the increase of liquid water contents in the snowpack coincides with an increase in density and a decrease of snow heights due to snow compaction. When the threshold for irreducible water saturation of the snowpack is reached and amounts of SWE stagnate, runoff will start as indicated by mass loss on the snow scale and decreasing SWE. Therefore, the initiation of runoff and associated mass transport in the snowpack can be predicted by continuous observation of the measurement data.

6.4 Applications

Radioecological transport processes associated with snow and their consequences on human radiation exposure have been investigated in field studies and associated laboratory experiments at Helmholtz Zentrum München since 2011. The environmental radionuclides Be-7, Pb-210 and Cs-137 were used as tracers for transport processes from the atmosphere to the hydrosphere, including deposition, storage in the snowpack, release to melt water, karst aquifer and discharge at the Partnach spring. Monthly total deposition of radionuclides at the Zugspitzplatt and release to surface waters at Partnach spring was balanced for two hydrological years. The snow cover development was observed by use of the installed snow sensors at the central plateau. Residence times in the snowpack and timing of radionuclide release to melt water and recovery at Partnach spring were determined. The results help to trace the pathways of radioactive particles from the atmosphere to aquatic environments. Consequently, in cases of extensive radionuclide releases to snow covered environments, peak discharges and the scope of action for countermeasures can be predicted to mitigate the impact on water quality and human radiation exposure.

6.4.1 Scavenging coefficients for the washout of aerosol-bound radionuclides

Scavenging coefficients for the washout of aerosol-bound radionuclides with different precipitation events at the Environmental Research Station Schneefernerhaus were calculated. From 4800 h of precipitation events between November 2012 and December 2014 580 h met the criteria of meteorological stability (Bernauer 2015). These were classified as rain (7 %), snow (74 %) and mixed (19 %) events as well as into three snow crystal classes considering their sizes, shapes and fall velocities (Fig. 7d, Bernauer et al. 2015, 2016). For every class the mean scavenging coefficients were calculated related to the aerosol particle sizes. Rain is comparably efficient to remove small particles (diffusion driven, Fig. 9a). However, the presence of snow significantly increases scavenging in the intermediate (interception) and large particle range (impaction). Fine single crystals, often in the shape of needles, plates or dendrites, very efficiently remove particles (Fig. 9b). Graupel-like pellets behave similar to rain. Complex crystals like wet snowflakes or aggregates of crystals are a bit less effective compared to single crystals. The larger the surface and the more complex the hydrometeors are, the higher is the washout efficiency for ultrafine aerosol particles.

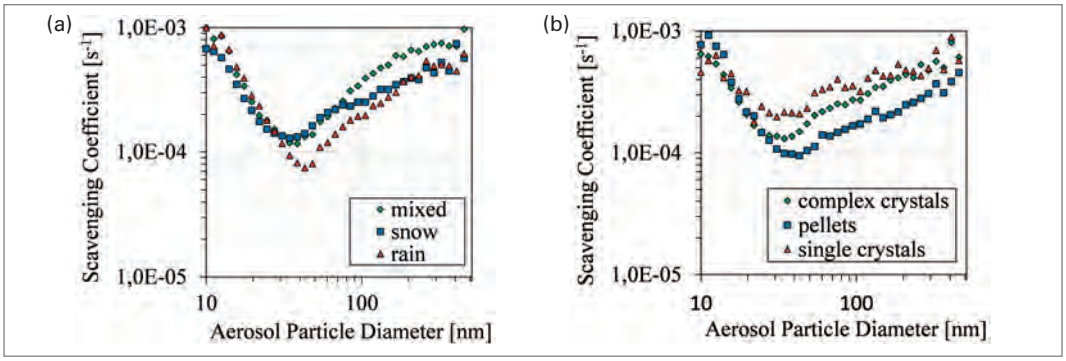

Fig. 9: Calculated scavenging coefficients for the washout of aerosol-bound radionuclides with different precipitation events (a) and differences between three snow crystal classes (b) related to the aerosol particle size. Shown are mean values of 580 h of precipitation at the Environmental Research Station Schneefernerhaus between 2012 and 2014.

6.4.2 Deposition of environmental radionuclides with snow

In addition to the calculated scavenging coefficients, *in situ* total deposition of the environmental radionuclides Be-7, Pb-210 and Cs-137 was determined in monthly fresh snow samples and in seasonal snow profiles at the end of the snow accumulation periods in May in 2015 and 2016 at central Zugspitzplatt.

Monthly radionuclide activity concentrations, determined in fresh snow at Zugspitzplatt, were quite low and range between 530–1770 mBq L^{-1} for Be-7, 10–127 mBq L^{-1} for Pb-210 and 0.2–3.2 mBq L^{-1} for Cs-137 during the snow accumulation periods of October 2014 to May 2015 and November 2015 to May 2016. Radionuclide deposition activity notation is always given as activity referred to the sampled area. Therefore, monthly samples were taken from size-defined areas or otherwise (for samples from deeper seasonal profiles) activity concentrations were multiplied with the snow water equivalents (SWE) for the related snow samples. Resulting deposited mean activities in the fresh snow are calculated with 143 Bq m^{-2} for Be-7, 7 Bq m^{-2} for Pb-210 and 0.16 Bq m^{-2} for Cs-137. The sums for both winters 2014/2015 and 2015/2016 for fresh snow samples, accumulated in the seasonal profiles at the end of the snow accumulation period in May each year and in precipitation samples (mainly rain) from the campus of Helmholtz Zentrum München in Neuherberg (500 m a.s.l.) during the same period for comparison are given in Tab. 1.

Tab. 1: Sums of mean deposited activities of the environmental radionuclides in snow samples at Zugspitzplatt and in fresh precipitation at the campus of Helmholtz Zentrum München in Munich-Neuherberg for comparison.

	Be-7 (Bq m^{-2})	Pb-210 (Bq m^{-2})	Cs-137 (Bq m^{-2})
Sum of mean deposited activities in fresh snow of the winter seasons 2014/2015 and 2015/2016 at Zugspitzplatt	1005	46	0.8
Sum of mean deposited activities in accumulated snow at the end of the snow accumulation periods 2015 and 2016 at Zugspitzplatt	724	88	1.4
Sum of mean deposited activities in precipitation of the same period in Munich-Neuherberg	530	65	0.3

Higher concentrations of Cs-137 on Mt. Zugspitze compared to Munich can be explained by higher resuspension of Chernobyl fallout radiocaesium from the soils in the surrounding forested valleys, where concentrations are still high due to limited post-accidental distribution. In addition, the use of contaminated local wood for domestic heating in the surrounding towns and villages contributes to increased Cs-137 activities in the atmosphere in winters. Pb-210 activities are expected to be higher in the valleys due to its terrestrial origin. Similar to Cs-137, Pb-210 is progressively accumulated in altered snow due to the strong fixation to dust particles. The Be-7 activities of cosmic origin generally increase with altitude. Due to its short half-life of 53 d, the concentrations in the accumulated snow samples at the end of the winter season are much lower.

6.4.3 Snow cover development at Zugspitzplatt

A persistent snow cover with varying thickness of maximum 150–400 cm was existent at Zugspitzplatt since 2012 each year and always lasted from November to at least June/July. The measured snow parameters at the central Zugspitzplatt show that there is almost no mass loss of snow between November and April each year, except for some wind-drift of fresh powder snow during periods of high wind intensity and low temperature. This means, the altitude and recent meteorological conditions at this site induce a continuous accumulation of snow and prevent melt water runoff at the snow base during the winters. Still, partial melting at the snow surface and related snowpack compaction occur. Laboratory experiments with snow columns and applied artificial radionuclide tracers under controlled ambient conditions, adjusted in melt-freeze-cycles to real conditions of a typical spring day with snowmelt on Mt. Zugspitze,

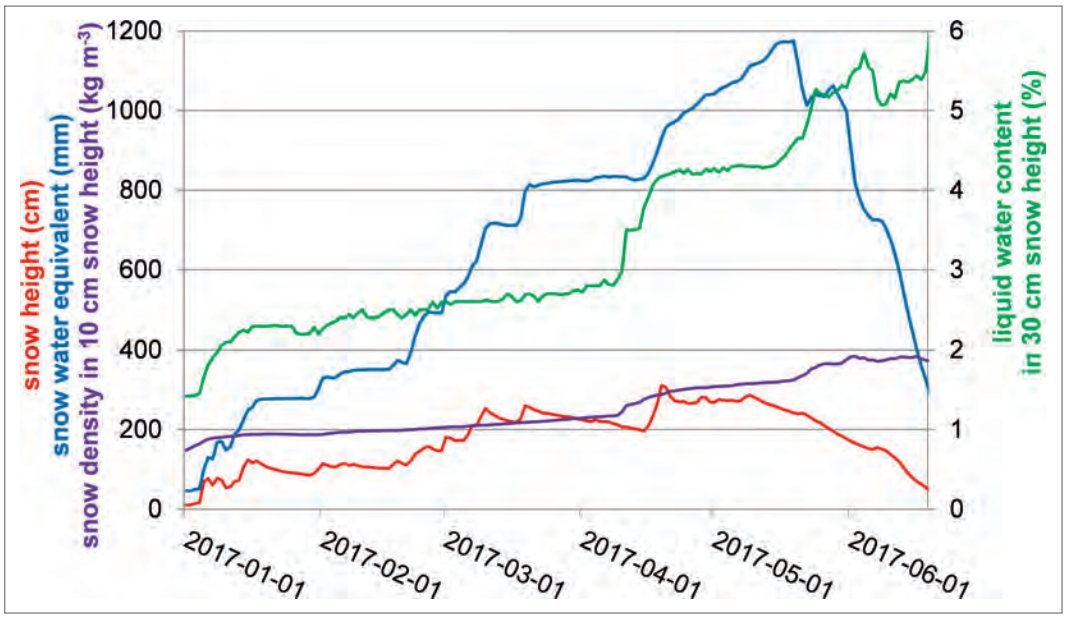

Fig. 10: Snow cover development during the winter season 2016/2017.

showed that radionuclide transport in a snowpack is associated with the percolation of liquid water in a snowpack (Hürkamp et al. 2017). Similar values were found for the release of other ions from snowpack (Cadle et al. 1984, Bales et al. 1989). As long as temperature stays below 0 °C radionuclide transport is almost disabled. It was shown that 80 % of the radionuclides were released within the first 20 % of melt water (> 50 % in first 10 %) in short time periods after snowmelt runoff started. This means, in case of a radioactive contamination on Mt. Zugspitze, the transport of a possible contamination in melt and surface water would be delayed until first melt water runoff in April or May, but will continue in a highly concentrated contamination plume in the melt and surface water.

Using the snow sensors installed at the central Zugspitzplatt, snowmelt runoff initiation and associated mass transport and release from snowpack can retrospectively be determined. By combination of information about air and snow temperature, snow water equivalent (SWE), liquid water contents and snow density, the time of first snowmelt runoff can be predicted. Measurement data of the winter season 2016/2017, plotted in Fig. 10, gives an adequate example.

Despite fluctuations of the snow height due to evaporation, surface snowmelt and related snow compaction, almost no mass loss/melt water runoff was recognised on the snow scale, indicated by a continuously accumulating SWE until 2017-05-19. Nevertheless, density and liquid water contents of the snowpack increased since the beginning of the year due to ablation and progressive snowpack compaction. When the snowpack got isothermal at 0 °C snow temperature (not continuously measured at the station) after several days of air temperatures > 0 °C, a wetting front could be traced by the increase of liquid water contents in different heights of the snowpack and finally at the snow base. A distinct increase was observed after 2017-04-07 and again after 2017-05-15 in 30 cm snow height, indicating full melt water saturation of the basal snow layers and imminent runoff initiation. After the first snowmelt release, liquid water contents generally further increase due to persistent melt water fluxes from upper snow layers.

The period of first melt water runoff in May is relatively constant since 2012. It varied between mid of April and mid of May, independent of the thickness of the snowpack. Assuming a potential radioactive contamination of the snowpack during the winter months of 2016/2017, its first release with snowmelt runoff would have occurred on 2017-05-19. If according to Hürkamp et al. (2017) a release of > 50 % of the radionuclides within the first 10 % of melt water is assumed, it would have taken only 11 h for their release to the surface water and karst aquifer respectively. This time interval strongly depends on the duration and intensity of the first melt-cycle and varied on Mt. Zugspitze between 15.5 d and 11 h since 2012. It also defines the scope of action to mitigate human radiation exposure by means of finding strategies for countermeasures, as

for example snow removal as an option for sealed urban regions. A prediction of a possible temporal and spatial distribution of the contamination plume in surface and ground water is feasible. It gives the chance to at least inform the population to be aware of an expected contamination plume in provided drinking water reservoirs.

6.4.4 Water and radionuclide budget for the catchment of the Partnach spring

Continuous measurements of the water discharge at Partnach spring (Wetzel et al. this issue) supplemented by the analysis of environmental radionuclides in grab samples of the Partnach surface water between May and September 2015 and 2016 were used to calculate a water and radionuclide budget for the catchment of the Zugspitzplatt for two hydrological years. Using the modelled area-weighted means for the snow water equivalents (SWE) according to Weber et al. (2016), the data represents the snow mass of the total catchment to 87.8 % compared to the determined SWE on the snow scale at central Zugspitzplatt. Therefore, a factor of 0.878 was applied to calculate SWE for the entire catchment of the Partnach spring of 11.4 km^2 on the basis of the measured values. The comparison of SWE stored in the accumulated snowpacks in the seasons 2014/2015 and 2015/2016 until May with summed melt water discharges at Partnach spring between May and September each year provided a recovery of > 90 % (Tab. 2). Each year, about 12 million m^3 SWE were accumulated and discharged. This corresponded to a total discharge at Partnach spring until almost drying up by the end of October of 44 % and 39 %, respectively.

Mean activities of the environmental radionuclides deposited in the total area of the catchment summed up to 10 GBq Be-7, 500 MBq Pb-210 and 8 MBq Cs-137 each year. Fallout from the atomic bomb test can be detected in the snow of the catchment as well. The annual inventory is small in comparison: 20 kBq Pu-239, 15 kBq Pu-240 and 95 kBq Am-241 (Gückel et al. 2017).

The activity concentrations of the environmental radionuclides detected in the Partnach surface water samples were quite low and even below detection limits in many samples, especially for Be-7 and Pb-210. Besides the short half-life of Be-7 of 53.2 d, this explains the limited recovery of these radionuclides in the Partnach surface water (Tab. 2). Long residence times of precipitation-borne Be-7 in the snowpack as well as in the karst aquifer led to its almost complete decay when discharged at Partnach spring. Additional supply by fresh precipitation only occurred periodically and carried no weight. However, Cs-137 was well recovered in the Partnach water, and in 2016 even in higher concentrations than determined in the integrated amounts in the fresh snow during the preceding winter season. An accumulation of radiocaesium in long-term stored altered water in the karst aquifer that was replaced by the first fresh melt water fluxes was assumed. In addition, uptake of Cs-contaminated mineral or organic particles on its way to Partnach gauge seemed to have been possible.

The water stable isotopes $\delta\,^{18}O$ and $\delta\,^{2}H$ were used as additional conservative tracers for water discharge and mass transport processes associated with snow. Since their isotopic ratio alters due to fractionation they provide further information about snow alteration and melt processes (Hürkamp et al. 2019). Their distribution in fresh precipitation or deposited snow as input parameters and the isotopic composition of the Partnach surface water as output were applied

Tab. 2: Water and environmental radionuclide budget for the catchment of the Zugspitzplatt (11.4 km^2) for two hydrological years 2014/2015 and 2015/2016.

	2014/2015				**2015/2016**			
	SWE (L)	Be-7 (Bq)	Pb-210 (Bq)	Cs-137 (Bq)	SWE (L)	Be-7 (Bq)	Pb-210 (Bq)	Cs-137 (Bq)
Input (snow)	1.21E+10	1.06E+10	5.42E+08	7.39E+06	1.22E+10	1.23E+10	4.94E+08	9.14E+06
Output (surface water)	1.10E+10	9.48E+07	5.30E+07	5.37E+06	1.13E+10	1.02E+09	< LOD	1.80E+07
Difference	1.05E+09	1.05E+10	4.89E+08	2.03E+06	9.22E+08	1.13E+10	4.94E+08	-8.82E+06
Recovery	91 %	1 %	10 %	73 %	92 %	8 %	0 %	197 %

to model the water transit times in the discharge at Zugspitzplatt (lumped-parameter exponential model according to Maloszewski and Zuber 1982). Residence times of about ten weeks were determined for the direct discharge considering a groundwater contribution of 50 %.

In collaboration with partners from EURAC research Bolzano, Italy within the framework of the Virtual Alpine Observatory (VAO), developments of snow coverage, liquid water contents of the snowpack and expected snowmelt could also be observed in remote sensing SAR Sentinel-1 data (Marin et al. 2020). We found that the multi-temporal SAR measurements allow the identification of the three melting phases that characterize the melting process i. e., moistening, ripening and runoff. In detail, we found that the C-band SAR backscattering decreases as soon as the snow starts containing water, and that the backscattering increases as soon as SWE starts decreasing, which corresponds to the release of meltwater from the snowpack. We show a spatially-distributed application of the identification of the runoff onset from SAR images for the Zugspitzplatt catchment. Results allow to better understand the spatial and temporal evolution of melting dynamics in mountain regions. The presented investigation could have relevant applications for monitoring and predicting the snowmelt progress over large-scaled regions.

6.5 Conclusions

Environmental radionuclides are useful tracers for the comprehension of mass transport pathways and processes concerning snow. In various applications since 2011, radionuclide deposition, snow cover development on Mt. Zugspitze and contained amounts of environmental radionuclides, as well as their transport and release to melt and surface waters, were investigated. It was proven that radionuclides are very efficiently removed from the atmosphere by snow. They are stored and accumulated in snowpack as long as temperatures remain below 0 °C, which is the case on Zugspitzplatt between November and April each year under current meteorological conditions. After initiation of snowmelt, a rapid transport through the snowpack starts until the base layer is fully water saturated. Radionuclides can then be short-term released to surface water. Laboratory experiments showed that > 50 % of the radionuclide inventory of a snowpack will be released within the first 10 % of melt water runoff. In minimum it took 11 h at central Zugspitzplatt in May 2017. Depending on the duration and intensity of the first melt-cycle, variable radionuclide concentrations have to be expected in the surface water and are further distributed into soils, ground – or drinking water reservoirs. It was shown that their spatial and temporal release on Mt. Zugspitze can be predicted in order to improve radionuclide distribution models or to derive strategies and scope of action for countermeasures in case of a potential nuclear or radiological accident. The overall goal is to mitigate impact on water quality and human radiation exposure.

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