Unprecedented radioactive pollution in Spitsbergen air during the 21st century

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Developments in nuclear science throughout the 20th century have led to the systematic production of novel radioactive contaminants on a global scale. Given the risk of increasing background radiation levels, routine environmental monitoring has become necessary. The research presented provides the first Arctic data since 1999 on time series of ^{238}Pu, $^{239+240}$Pu and ^{241}Am in the lower atmosphere of Hornsund, Spitsbergen, spanning 2007 to 2021. While the general levels of ^{238}Pu and $^{239+240}$Pu were comparable to recent observations from different sites, ^{241}Am was found to be extraordinarily high, with a maximum of 354 nBq/m^3 detected in the 1st quarter of 2019. Subsequent analysis of isotopic ratios revealed a frequent enrichment of ^{238}Pu over $^{239+240}$Pu, inconsistent with previously documented releases. Unexpected single incidents of ^{237}Np were encountered in 2013, 2014 and 2018.
A multivariate analysis incorporating data on ^{7}Be, ^{210}Pb, ^{137}Cs activity concentrations and a wide range of meteorological factors was applied to explain the behaviour of artificial actinides in ground-level air layers. Evaluation of Spearman correlations allowed explicit links between $^{239+240}$Pu seasonal trends and natural processes, such as local resuspension throughout the year and horizontal tropospheric transport of haze layers from remote areas in the 1st quarter. Similar mechanisms controlled a certain proportion of ^{238}Pu, but to a lesser extent. The maximum activity concentrations of 6.61 nBq/m^3 for ^{238}Pu and 15.51 nBq/m^3 for $^{239+240}$Pu recorded in Hornsund during the 3rd quarter of 2015, and simultaneously registered at middle latitudes, could be related to random events such as fly ash particles remobilised by 2015 wildfires. A majority of the significantly elevated levels of ^{241}Am, ^{238}Pu and ^{237}Np were found not to be environmentally induced. The average annual doses associated with the exposure to investigated alpha emitters were negligible, being about a million times smaller than the typical background radiation doses of 2.4 mSv per year. Therefore, the contamination detected did not pose a radiological threat to the Arctic environment.
Nevertheless, the ^{241}Am, ^{238}Pu and ^{237}Np signals were alarming, as their occurrence could indicate man-made emissions, completely unnoticed by the regular monitoring of gamma emitters in Hornsund air. Trajectory simulations performed for the 1st quarter of 2019 showed the most prominent transport pathways from northern Asia and Europe via the island of Novaya Zemlya. Nuclear aerosols were carried at low levels in the troposphere (below 100 m), resulting in weak dilution and intense deposition. Of note, no radioactive discharges of man-made actinides to the atmosphere have been declared in the last decades. However, a ^{241}Am incident was identified exclusively in the urban air of central Poland during a few weeks in 2021, possibly generated by the combustion of isotopic smoke detectors. The research highlighted the importance of including alpha emitters in the routine measurements within radiation situation control programmes.