Spontaneously generated, radioactive nanoparticles in the environment

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ABSTRACT

For almost 100 years, and perhaps longer, observers have detected spontaneous dispersal of radioactivity from macroscopic quantities of radioactive materials with the first published observations reported in 1910. In the 1960's and later, radioactivity was observed to migrate through HEPA filters despite their well-established filtration characteristics. In measurements of ground water, radioactivity has been found to disperse from its original location of deposition in soil, despite the size, insolubility, and resistance to chemical reactions of the radioactive particles originally deposited. Similarly, measurements of the uptake of these materials in lung tissue and studies of their solubility in simulated biological fluids showed the solubility to be related to the radioactivity of the materials. In numerous practical examples, the migration and deposition of radioactivity affects work and operational practices. Despite this long and varied history indicating the importance of self-dispersal of radioactive materials, no measurements had ever been reported of the materials which were actually dispersed until recently and the results are quite surprising, suggesting a well-defined process creating discrete nanoparticle fractions. This paper will review the history of the observations of spontaneous dispersal of radioactivity and close with a description of the first measurements of the dispersed nanoparticles and suggestions of the physical processes involved in their formation.

Keywords: nanoparticle, aerosol, radioactivity, production, transport, environment, colloid, groundwater

1. INTRODUCTION

Transport and interaction properties of condensed-phase materials introduced into the environment largely determine their environmental effects. Thus, the graphite of a pencil lead would be of little environmental interest whereas graphitic structures of nanometer diesel exhaust particles are of considerable environmental interest for their human health effects if inhaled, for their effects as cloud condensation nuclei, and for their possible effects on the earth s radiation balance. The importance of all of these environmental effects are critically dependent upon the chemical and physical interactions of the diesel particles, where they are located, and their ability to be transported to locations where their effects are either enhance or suppressed.

In the atmosphere, air pollution control has evolved since the 1960's from regulating only the total mass emissions to total mass for aerosol particles smaller than, perhaps 20 μm, to today s controversial fine particle standard that seeks to control emissions of aerosol particles below 2 μm, thereby moving into the submicrometer, or the nanometer, range of particle sizes. In this size regime, particles generally do not arise from comminution, or breaking-down, processes but rather from direct emissions, such as in the diesel soot example, from various gas-to-particle conversion processes,

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to a limited degree from evaporated water sprays, and to a very limited extent from resuspension of particles by air motion.

Analogous observations may be made for particles in soils and sediments where groundwater is the means by which transport occurs. There, solubility and chemical reactivity are generally taken to determine all transport characteristics for materials too large to be carried by the water. To the extent that this model fails to explain observations, colloid-assisted transport is adduced to explain the observations. There, nanometer-dimensioned particles are thought to facilitate the transport process. As for aerosols, the critical actors again turn out to be nanoparticles.

Under the assumptions that particle size, chemical, and physical reactions (e.g. dissolution or condensation) are entirely adequate to determine material transport, radioactive materials are generally considered to be of no environmental concern beyond their locations of original deposition or appearance, with a few well understood exceptions (naturally occurring ^{222}Rn). The reason for this conclusion is that these radioactive species, and particularly those arising from the heavy elements, are mostly non-reactive and appear as particles much larger than are readily transported by ordinary atmospheric or subsurface processes.

This paper will examine this assumption based on data from a number of fields and will conclude with a few comments on recent work.

2. TRANSPORT OF RADIOACTIVITY: SELECTED OBSERVATIONS

2.1. HEPA filter penetration.

High Efficiency Particulate Air (HEPA) filters are widely used for reducing hazardous, airborne particulate matter in gas process stream emissions to workplaces and the atmosphere to concentration levels too low to be measured. The operative filtration processes involve deposition (and adhesion) of particles to filter surfaces from flowing gas streams primarily (1) by inertial processes for particles larger than 1000 nm and (2) by thermal diffusion for particles smaller than 100 nm. Normally, particles that are geometrically compact and too large to penetrate the filter passages and the very large particles that gravitationally settle are removed by other means prior to reaching the HEPA filtration stage to prevent airflow restriction and the necessity of frequent filter replacement. The particle size interval of 100 nm to 1000 nm is where filtration occurs by a combination of inertial and diffusion processes and filter efficiency falls to no less than 99.97% for 300 nm diameter (i.e. aerodynamically equivalent diameter) particles; filtration efficiency is larger for both larger and smaller particles. Extensive measurements and regulatory guidelines underlie these operational parameters.

To provide the best possible protection for personnel working with, or in the vicinity of, radioactive materials, HEPA filtration of workplace air has been standard practice for many years. However, in the 1960's, monitoring of worker radiation exposure showed incidences wherein unexplained, high exposures were recorded. These exposures caused a re-examination of filtration efficiency and various anecdotal and internal laboratory reports from the 1960's and early 1970's indicated an inconsistent picture of the effectiveness of filtration for the removal of radioactive materials from room air.

A study was conducted to address the questions of retention and penetration characteristics of glass fiber filters[1] challenged by radioactive ^{212}Pb particles. Generation of ^{212}Pb was via the decay chain

$$^{228}\text{Th} \rightarrow \ldots\ldots. \rightarrow {}^{220}\text{Rn}(55\text{ s}) \rightarrow {}^{216}\text{Po}(0.16\text{ s}) \rightarrow {}^{212}\text{Pb}(10.64\text{ hr}),$$

where the numbers in parentheses are the half-lives of the nuclei. First, ^{228}Th on a ferric oxide carrier was placed in a cup that itself was covered by a metal screen and two glass fiber filters, all located in a sealed chamber. In this chamber, a 22.5 V electric field was maintained between the cup and a needle placed near the filters and screen covering the cup. The ^{220}Rn penetrated the filters and screen to enter the space in the vicinity of the needle. Its decay in this zone lead to the formation of charged, gas phase ^{212}Pb atoms that accumulated on the needle tip. This lead-covered needle was removed from the generation chamber and placed in the filter test apparatus which consisted, in order, of a constant source of clean sample air flowing through the radioactive particle source chamber and then through the filter exposure chamber followed by an air pump. The needle was placed in the particle source chamber where decays of the ^{212}Pb dislodged additional ^{212}Pb atoms due to the deposition of decay kinetic energy in the ^{212}Pb deposit on the needle. These newly-aerosolized lead atoms (or clusters) became the challenge aerosol for the filters.

In the first set of measurement, five filters were placed in sequence in the filter exposure chamber and the radioactivity due to the lead captured by each filter was measured. On filter 1, 81% of the emitted radioactivity was measured, on filter 2, 18% was measured, on filter 3, about 0.2% was measured, etc. These results clearly confirmed that despite the small size and high filtration expected of the lead particles, substantial radioactivity was transported through the first, and second filters. In the second set of measurements, the first filter was exposed to the aerosol, counted, and subsequently was backed by four clean filters. Clean air was then drawn through the filter stack to obtain a direct indication of transfer of radioactivity from the first filter to the subsequent filters. The results showed that the fraction of the total activity determined for the first filter became redistributed as follows: first filter, 76%, second filter, 8%, third filter, 8%, etc. with the sum of the activities equaling the expected activity on the first filter. The conclusion is therefore that ^{212}Pb is injected into the air stream from the material deposited on the first filter.

2.2. Groundwater transport of Pu.

A study[2] of actinides in the groundwater of the Nevada Test Site (NTS) was conducted with the objective of determining if these actinides are stably located on soil materials and migrate only to the extent consistent with their solubilities or if evidence indicates otherwise. In those studies, the presence of Pu, and other actinindes, in subsurface water was measured over a kilometer from their source and determined to be about 80% associated with particles under 1000 nm with only a small part (less than 10%) in the ulrafiltrate fraction, corresponding to solution-phase species below 7 nm. This established that pure aqueous phase actinides could not be responsible for the measured migration. In addition, Pu migration was established based upon the measured ^{240}Pu/^{239}Pu ratio at various sites since this ratio is considered a unique marker of each underground test (each at a different location). The conclusion of their study indicated Pu is not immobile in the subsurface, but can be transported over significant distances. Pu transport model that only take into account sorption and solubility may therefore underestimate the extent to which this species is able to migrate in ground water.

The context for this work and its conclusions on Pu migration is the contemporary picture[3,4] of groundwater transport of insoluble chemical species, such as Pu. Before about 1990, transport was considered only in solution with the remainder of a species immobilized. However, if a highly insoluble molecular species strongly associates with a colloid particle, it would shift the apparent solubility of the molecule, assuming no other interactions with immobile soil particles. The colloidal transport itself could arise via two mechanisms: (1) condensation or precipitation of the actinide to comprise the primary material of the colloid and (2) adsorption onto colloids of impurity or other foreign material in aqueous phase.

The authors of the NTS study indicate this picture of colloidal transport is incomplete. In their study, they measured aqueous concentration of Pu of less than 10^{-14}M. However, Pu(V) is the species likely in the NTS water, but its solubility has been measured in the lab as about 10^{-8}M. Even if Pu(IV) is present in NTS water, its calculated solubility limit range of 10^{-12} to 10^{-17} M only brackets the measured range and is therefore too low to lead to precipitation of a solid phase of Pu. Nonetheless, their measurements indicate that less than 1% of the observed Pu is in the dissolved fraction of the groundwater.

In conclusion, there is a fundamental difficulty in rationalizing these data with the contemporary picture. The Pu associated with the colloidal particles must somehow be transferred from an immobile source phase via the intermediate aqueous phase. Some evidence of that transfer must be manifested either as a dissolved phase or in the chemistry of the trace species in the aqueous phase that facilitate that transfer and neither is apparent in the discussions of this subject in the literature.

2.3. Biological observations

In the assessment of the biological effects of inhaled radionuclides, the distribution of these nuclides throughout the body is of critical importance. PuO_2 is known to be insoluble in water and generally not subject to reaction or dissolution in lung fluids. Consequently, if PuO_2 particles that are too large to be transported across membranes are inhaled, then one would not expect the Pu to appear in other tissue in the body, nor even to be assimilated into the lung tissue itself. Nonetheless, extensive observations on both rat and dog tissue have shown different tissue distributions for $^{238}PuO_2$ and $^{239}PuO_2$ with a higher translocated fraction for the 238 isotope than for the 239 isotope[5].

One study[6] reports the assimilation by beagle dogs of Pu from inhaled 1400 nm diameter $^{238}PuO_2$ particles. Measurements were made of alpha tracks (autoradiography) due to the retained radioactivity in the animal lungs at different times from 4 days to 730 days following exposure. Throughout all time intervals, those measurements showed quite clearly that Pu appeared in the lung tissue, with the largest part of the activity (90—99%) identifiable as originating from particles of under 40,000 atoms and small fractions associated with larger intervals of particle size up to 181,000—320,000 atoms (the original 1400 nm particles contained about 10^9 Pu atoms). The authors comment that there even appeared to be a slight bimodality in the activity-size distribution of the particles. While this is not the first study showing the presence of Pu at locations removed from their points of initial deposition, it gives explicit information of the size associations of this radioactivity and is therefore of interest relevant to earlier studies seeking the cause of this assimilation from measurements of the dissolution of Pu.

Based on earlier data, a study[7] of Pu compared the dissolution rates for the 238 and 239 isotopes. The carrier fluid is reported to be simulated lung fluid and was intended to give insight to biological measurements. In the data, dissolved Pu was taken to be matter containing Pu that passed through a 100 nm pore size membrane filter. It showed the ratio of the dissolution lifetimes for ^{238}Pu to ^{239}Pu to be on the order of 200. According to the author, the ratio of dissolution rates is, however, in agreement with the ratio of the specific radioactives(-ities) , and therefore suggests that radiation damage of some type plays the primary role [7], in causing the dissolution.

2.4. Observations from practice.

Throughout work involving radioactive materials, numerous observations and procedures recognize the existence of spontaneously translocated radioactivity.

Radioactive dusts are a universal observation in Pu and U handling [8] regardless of how the materials are treated. These so-called dusts are either radioactive aerosols or deposited particulate materials of largely the same composition as the materials being handled and their removal from the workplace is one utilization of HEPA filtration as discussed in section 2.1, above. As in the other cases, these particles appear to arise simply from the presence of the materials in bulk and their suspension is unlike any contamination arising from non-radioactive materials.

Another common indication of spontaneous translocation of radioactivity is the observation of the upwardly-drifting baseline for scintillation crystals employed for routine measurements of radioactive materials. Accepted practice is for the zero level , or noise floor, to be established for each scintillator immediately prior to each new set of measurements. This observation is consistent with the accumulation on the crystal surface of mobile radioactive particles.

A third, commonly observed phenomenon in the routine handling of radioactive materials is associated with cleaning a floor or other laboratory surface after a spill of radioactive material. Following the clean-up, radioactivity is often measured on surfaces in the vicinity of the spill, but not necessarily at the spill location itself. Once again, this observation in indicative of the spontaneous translocation phenomenon.

3. LABORATORY STUDIES RELATED TO MECHANISM OF FORMATION

While the preceding examples demonstrate that radioactive materials translocate without external stimulus, the physical processes by which this occurs have not been elaborated. The deposition of recoil kinetic energy from decaying nuclei near the surface has long been understood to initiate the removal of material from the parent phase and this understanding has been used in qualitative models[7] [above]of the above process. However, detailed product characterizations and quantitative analyses of energy deposition are required for a thorough understanding of the dispersal process. In the two examples of this section from the literature, the original observation of decay-initiated translocation is described and a well-studied process of desorption induced by the deposition of nuclear kinetic energy is cited.

3.1. First observation of mechanical dispersal of radioactivity by decays.

While the above examples all date from well into the second half of the 20th Century, apparently the first published report of spontaneous translocation of radioactivity appeared as early as 1910[9] and was based upon the observed decay chain (here in modern notation and neglecting the 164 μs half life ^{214}Po daughter of ^{214}Bi)

$$^{222}\mathrm{Rn}(3.82\ \mathrm{d}) \rightarrow {}^{218}\mathrm{Po}(3.05\ \mathrm{m}) \rightarrow {}^{214}\mathrm{Pb}(26.8\ \mathrm{m}) \rightarrow {}^{214}\mathrm{Bi}(19.7\ \mathrm{m}) \rightarrow {}^{210}\mathrm{Pb}(22.3\mathrm{y})$$

as it was understood at that time. In that work, a clean negative electrode was first exposed to ^{222}Rn gas to capture its $^{218}\mathrm{Po}^+$ decay product. On removal from the radon gas chamber, ^{218}Po, ^{214}Pb, ^{214}Bi were found on the electrode plate but decayed to ^{214}Bi and ^{210}Pb after a short time. This plate was then placed a short distance from another plate, B, that received the ^{218}Po, ^{214}Pb, ^{214}Bi atoms due to their recoil from the first plate as they were formed from their parent nuclei. Since the half-life of ^{210}Pb was long, the authors expected to be able to study ^{214}Pb decays from the first disk by observing ^{214}Bi decays, alone, on plate B. However, what they found was that if the disk B was exposed to the active plate P in a vacuum, there was in general radiated to the disk, not only radium C (^{214}Bi), as was to be expected, but also a certain amount of radium B (^{214}Pb). Since sufficient time had elapsed for the disappearance of the ^{218}Po via radioactive decay, its presence would have been negligible, so none of its recoil daughter atoms should have been in evidence. The conclusion is that ^{214}Pb and ^{214}Bi decays are mechanically dislodging the ^{214}Pb. This is, in effect a process in which the kinetic energy originates within the material, not externally, and suggests that it could be universal process affecting radioactive materials, or at least alpha-active materials.

3.2 Plasma desorption

Volatilization of heavy biomolecules from surfaces without decomposition is a challenge for analytical methodology. Several methods have been developed that heat or otherwise impart sufficient energy to the surface to expel the biomolecule. In a paper reporting the plasma desorption method[10], decay products of ^{252}Cf are accelerated for collision with a film on which the biomolecule is deposited. The authors report that each fission fragment produces pulsed localized heating that is ideal for the vaporization of involatile biomolecules without decomposing them.

Here is an example in which the kinetic energy imparted to a condensed phase results in an effective heating of the collision medium, similarly as might be expected if the energetic decay product had originated within, but near the surface, of a condensed phase. In other contexts, this process is known as sputtering, or spallation and its experimental and theoretical study is a well developed area of research[11].

4. SIZE DETERMINATION OF SPONTANEOUSLY GENERATED PLUTONIUM PARTICLES

Throughout the work cited above, measurements or observations of radioactivity translocated in air did not address the characteristics of the translocated materials; they only determined that materials earlier located at one position evidently gave rise to material of the same type located elsewhere. As a result, no experimentally based assessment of environmental interactions, transport mechanisms, or even production processes for the translocated materials could be developed beyond the most rudimentary stages. Recently, the first determination of size characteristics of these materials as aerosol particles has been made[12] and the results suggest not only how they should be modeled as components of the background aerosol but their overall form suggests formation mechanisms that may not be consistent with the operational assumptions applied in describing the process[7].

4.1. Experimental methods and materials.

The source for the translocated radioactivity was a 1.1 g, 6.25 mm diameter disc of material similar to that utilized for the Light-Weight Radioisotope Heater Unit used to warm (by alpha-decay heating) critical components on spacecraft. This material is 80% $^{238}\mathrm{PuO}_2$ with the balance other Pu isotopes and uranium. Order-of-magnitude estimates indicate that even if all the nuclear recoil kinetic energy were deposited in creating surface (i.e. fracturing the material), the largest possible particle would be around 100 nm. For aerosol particles of this size and smaller, measurements using aerosol diffusion battery methods to gain size-related separations among the particles are entirely appropriate. A diffusion battery is an arrangement of diffusion cells that are porous media for which the removal of particles from a flowing gas stream to its interior surfaces is entirely by thermal diffusion and is well characterized[13]. In this experiment, a serial arrangement of cells[14,12] followed by a filter was employed. Therefore, most of the highest diffusivity, and therefore smallest, particles are removed from the air stream in the first stages of the diffusion battery with the deposition of larger particle occurring over a number of stages. After extended exposure of the diffusion battery to a flowing air stream that entrained the particles emitted by the sample, the ^{238}Pu activity deposited on each cell of the diffusion battery and the following filter were counted to provide the raw data from which the activity-size distribution was determined.

4.2. Data analysis and results.

Since the particles separate themselves to become deposited on the diffusion battery surfaces according to their thermal diffusivities, the activity-diffusivity distribution, strictly speaking, is the calculated quantity. However, there are unique relations between the particle diffusivity and diffusivity-equivalent radius and these relations are used to convert the diffusivity to radius for the purposes of interpreting and using the results. In the following discussion, reference to particle size should be understood in this context, i.e. as the size equivalent to a particular thermal diffusivity.

Extraction of size distributions from the data were accomplished by employing the widely-used Expectation-Maximization algorithm[15]. A distribution of discrete particle sizes exceeding the likely range of associated particle sizes was selected. The activities attributed to each of these particle sizes was repeatedly adjusted according to the E-M algorithm until the best-fit to the raw data set was obtained.

Three sets of data were taken and analyzed. All show the very distinct and suggestive characteristic of two widely separated and narrow particle sizes associated with the radioactivity: In data set 1, approximately 55% of the radioactivity is associated with 0.5 nm diameter particles and about 45% is associated with particles between 4 nm and 5 nm; in data set 2, about 70% is associated with particles about 0.5 nm and the remainder with particles between 2 nm and 3 nm; in data set 3, about 60% is associated with particles between 0.7 and 1.0 nm and about 40% with particles about 10 nm.

4.3. Interpretation of results and implications for formation mechanism.

In a solid, radioactive, material, the kinetic energies of the decay products of an internal atom are absorbed by the material with the energy deposition process occurring over a time interval of 10^{-11}s or less and distances on the order of a few micrometers or less. Consequently, two decays are generally well separated in location and time, implying that they may be treated as independent sources of the particles originating from the condensed phase. In the data quoted in the preceding paragraph, if only the smaller of the two size fractions were present, then the interpretation would be relatively clear, suggesting a sublimation or evaporation process from the material s surface. Similarly, if nanometer particles, only, were produced and they were distributed among a broad range of sizes, then a process of energy deposition that fractured the surface, as has been assumed in model calculations heretofore, would be appropriate. In this picture, the polydispersity in particle size corresponds to the random depths and orientations of the decaying nuclei below the surface. However, the production of the very narrow distribution in size, of the nanometer mode indicated above, does not readily arise in this picture without arbitrary assumptions.

Size distributions, such as quoted here, are typical of homogeneous condensation (or nucleation) events[16] (e.g. supersonic nozzle expansions of condensable vapors). In the processes investigated here, the deposition of the Pu decay energy in a small volume bordering the material surface raises the local temperature, perhaps sufficiently to expel variable quantities of near-surface material into the adjacent gas where the rapid cooling of the expelled materials takes them through their nucleation threshold. Because the volume is very small, few nucleation events can occur before the dilution of the gas-phase species terminates further collisions. In this picture, condensable species on the periphery of the expanding vapor all escape capture, as well others in subcritical nuclei. An example, in which a particle size distribution very similar to those described above, has been calculated[17] (for the case of ethanol) and corroborates the viability of this proposed picture.

5. CONCLUSIONS AND ISSUES YET TO BE ADDRESSED

In this paper, observations of the dispersal of radioactivity have been cited in a variety of fields. All appear to arise from a common physical cause, which is decays in the source materials occurring near their surfaces. While this origin has been acknowledged in some of these fields for many years, the first measurements of the dispersed materials have only recently appeared and their activity-size distribution differs considerably from what followed from prior treatments of the phenomenon. The new results suggest an ordered process of decay energy deposition in the parent phase followed by energy dissipation via evaporation, or sublimation of parent phase species into the ambient background gas followed by nucleation. Since decays at interfaces with condensed media are of considerable importance also, the question also arises as to what the form of the materials dispersed into those media may be, taking into consideration the drastically different thermal properties of the liquid, vs the gas. This process is likely of importance for all aspects of the storage, handling, transport, and disposition of transuranic and mixed wastes from weapons, electrical power, medical, and other sources throughout the world.

While work to date is suggestive, considerable additional research is required before a truly quantitative picture capable of predictive modeling is possible: To what extent do ionized species from the solid, vs neutrals, play roles in the gas-phase processes (e.g. ion-induced nucleation)? How do the apparent nucleation phenomenon and the particle properties depend upon both background gas pressure and composition? How do gas-phase species correspond to the sites of their origins in the surface of the parent phase? What roles do the structure and composition of the parent phase play? How sensitive is the entire process to the decay energy? Is the phenomenon restricted to alpha decays or does it also occur in other decays? From the side of developing a theoretically based model of the phenomenon, there are also a number of questions including how the transfer of electronic excitation energy (caused by absorption of kinetic energy from recoil particles) to acoustic (i.e. thermal or vibrational) modes, and finally to evaporation occur, not to mention the many processes occurring in the gas phase? And finally, how do the answers to all of these questions posed for the gas phase surrounding the decaying material translate into treatments of the process in the presence of liquid phases?

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