Understanding the contribution of naturally occurring radionuclides to the measured radioactivity in AWE Environmental Samples

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Outline

- Introduction
- Naturally occurring radioactivity
  - Gross alpha activity
  - ^{212}Pb activity
  - Activity excursions
- Conclusions
- Acknowledgements

Introduction

Background to the study

- The gross alpha and beta activity of surface waters collected by the North Ponds Water Management System is monitored on a daily basis.

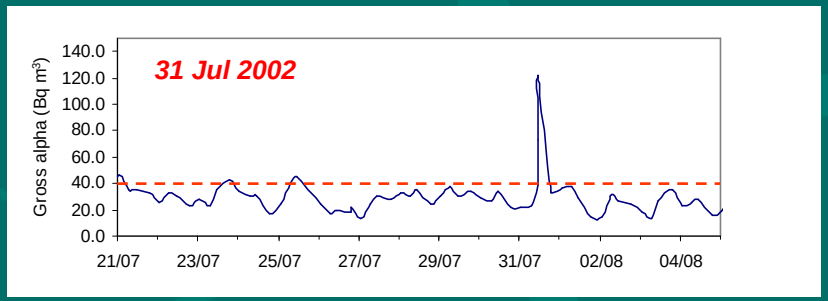
- The purpose of gross alpha monitoring is to screen for isotopes of uranium and plutonium (^{238}U, ^{235}U, ^{234}U, ^{238}Pu, ^{239}Pu and ^{240}Pu).

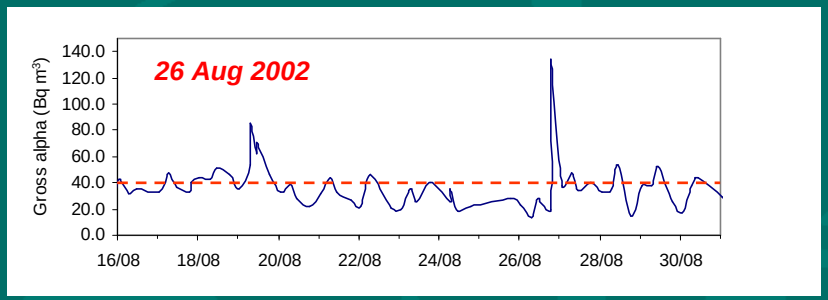
- The activity is typically 30 - 40 Bq m^{-3} and is primarily attributable to naturally occurring radionuclides (NOR) from the ^{238}U and ^{232}Th series.

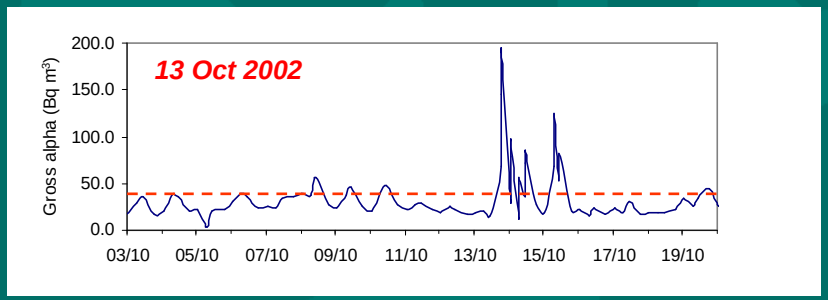
The North Ponds Water Management System.

Introduction

Background to the study







- Waters may not be discharged from site if > 40 Bq m^{-3} and further investigation must be undertaken to exclude U and Pu contamination.
- Typically 25 % > 40 Bq m^{-3} and 0.8 % > 100 Bq m^{-3}.
- Normally this involves a delay of 24 - 48 hours as the activity decays below the exemption threshold.
- This decay indicates a contribution from short-lived NOR.

Introduction

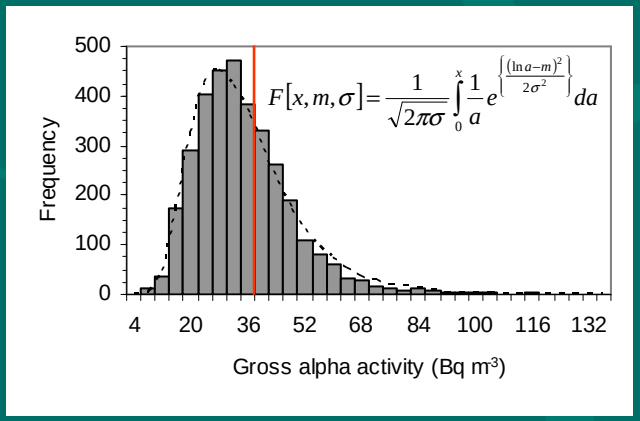
Project aims

- Identify the principal components of the gross alpha activity.
- Identify the origin of the sudden increases in gross alpha activity.
- Evaluate the contribution of short-lived NOR.
- Improve the technical underpinning and credibility of routine monitoring.

The River Kennet near Aldermaston.

Gross alpha activity

Typical measurements



Gross alpha measurements

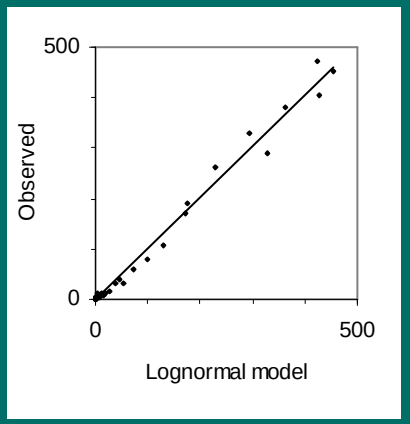
- Samples are prepared using an evaporation technique.
- Counting is by gas flow proportional counting.

- During January 2002 - September 2005:

  25.1 % > 40 Bq m^{-3} and 0.8 % > 100 Bq m^{-3}.

- Predictions made using the lognormal PDF:

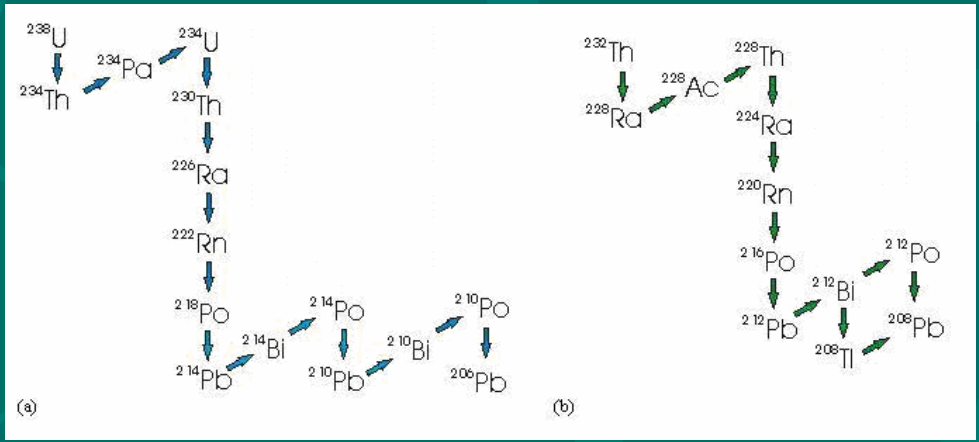
  26.4 % > 40 Bq m^{-3} and 0.3 % 100 Bq m^{-3}.



Correlation between gross alpha and lognormal model

Gross alpha activity

Principal components



The ^{238}U and ^{232}Th series

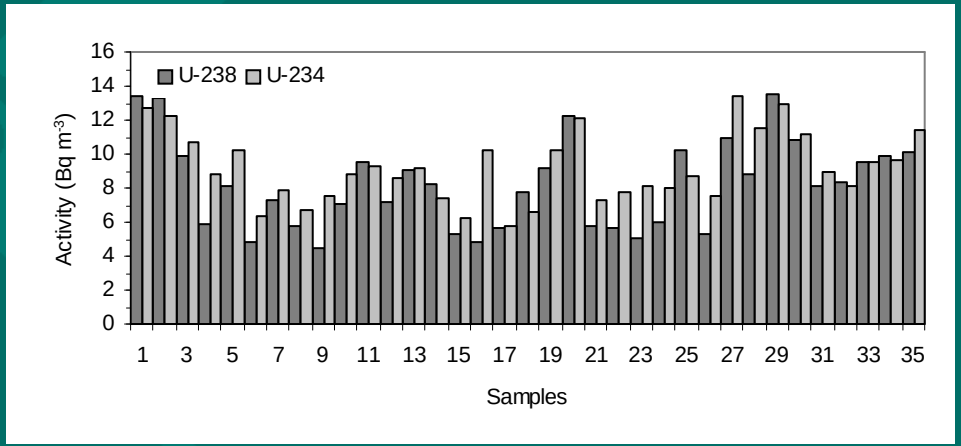
- Primarily NOR from ^{238}U and ^{232}Th series.
- ^{238}U + ^{234}U concentration of 17.4 Bq m^{-3} (26 Bq m^{-3}).
- ^{232}Th concentration of 1.1 Bq m^{-3} (1.8 Bq m^{-3}).

- Considering secular equilibrium to radon isotopes:

  4 x ^{238}U and 3 x ^{232}Th activity

  (4 x 8.2) + (3 x 1.1) = 36.1 Bq m^{-3}

- Plus contribution of short-lived NOR.



Uranium measurements

National Oceanography Centre, Southampton

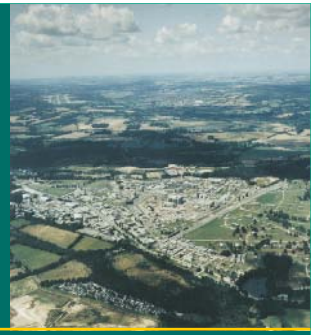
Gross alpha activity

Principal components

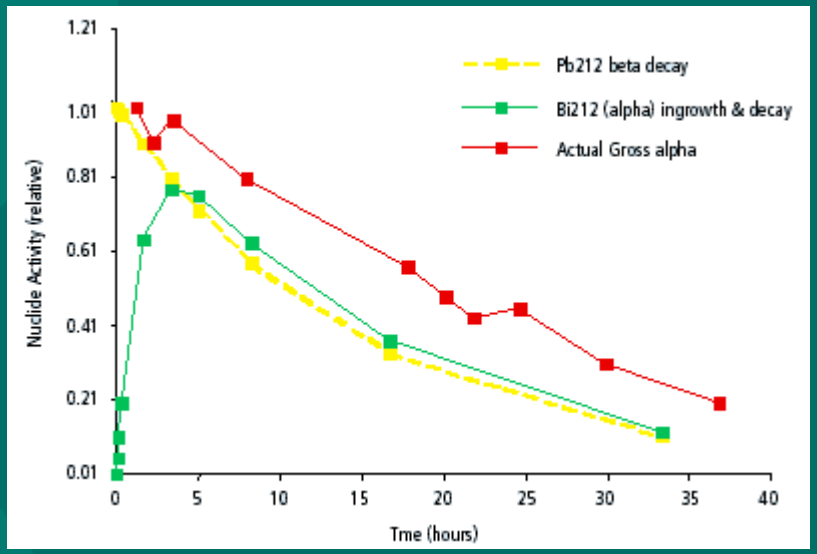
Uranium series (a)			Thorium series (b)		
Nuclide	Decay type	Half-life	Nuclide	Decay type	Half-life
U-238	α	4.5 x 10^{10} yrs	Th-232	α	1.4 x 10^{10} yrs
Th-234	β	24.1 days	Ra-228	β	5.8 yrs
Pa-234	β	1.2 mins	Ac-228	β	6.1 hrs
U-234	α	2.5 x 10^{5} yrs	Th-228	α	1.9 yrs
Th-230	α	7.5 x 10^{4} yrs	Ra-224	α	3.7 days
Ra-226	α	1620 yrs	Rn-220	α	55.6 sec
Rn-222	α	3.8 days	Po-216	α	150 msecs
Po-218	α	3.1 mins	Pb-212	β	10.6 hrs
Pb-214	β	26.8 mins	Bi-212	β	60.6 mins (64%)
Bi-214	β	19.9 mins	Bi-212	α	60.6 mins (36%)
Po-214	α	164 μsecs	Tl-208	β	3.1 mins
Pb-210	β	22.3 yrs	Po-212	α	300 ns secs
Bi-210	β	5.0 days	Pb-208		Stable
Po-210	α	138 days			
Pb-206		Stable			

Gross alpha activity

Short-lived component



- Since 1998 all samples > 40 Bq m^{-3} have decayed below the exemption threshold.
- Therefore the elevated activity must be attributable to short-lived NOR.
- Which short-lived NOR?
  - Samples decay with a half-life of 10 - 20 hours.
  - No alpha-emitting NOR with this half-life.
  - Must be ^{212}Pb in equilibrium with ^{212}Bi (36 %) and ^{212}Po (64 %).



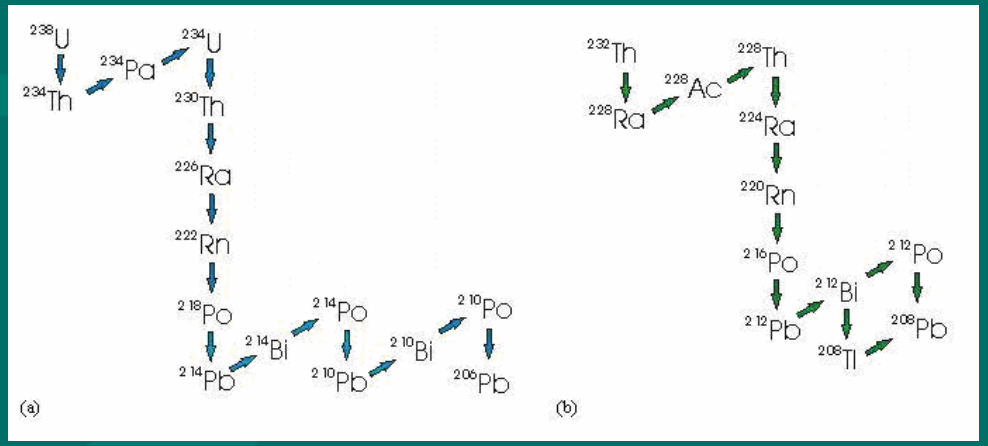
Radioactive decay of gross alpha measurements

National Oceanography Centre, Southampton

^{212}Pb activity

A difficult radionuclide to measure

- Produced by the radioactive decay of ^{220}Rn.
- Half-life of 10.64 hours.
- Detection is difficult due to its short half-life and relatively low activity.



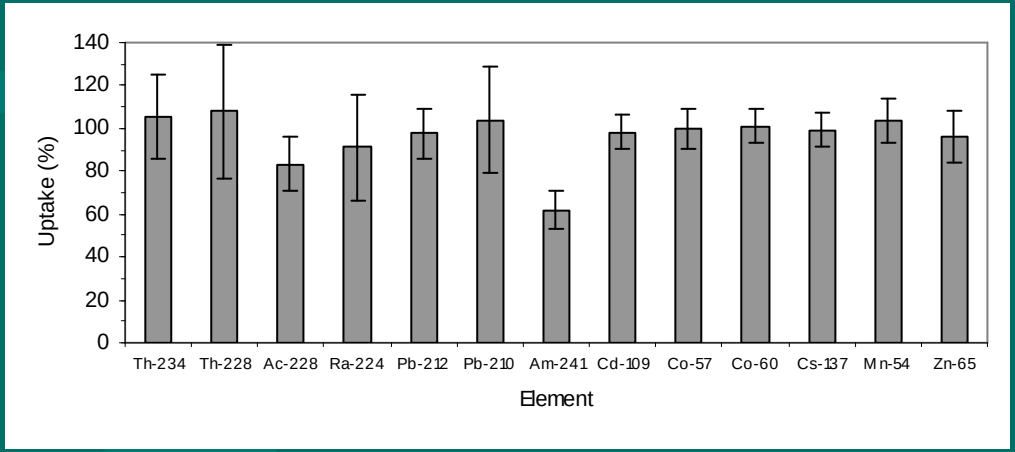
The ^{238}U and ^{232}Th series

- Detection requires:
  - Pre-concentration - Water samples passed through a MnO_2 resin.
  - Separation - MnO_2 resin dissolved and ^{212}Pb separated from interferences using Sr resin.
  - Low-level detection - Direct measurement using LSC with alpha/beta discrimination.
- Measurement within 5 - 7 hours, 92 % recovery, $L_D = 0.006 \pm 0.001$ Bq l^{-1}.

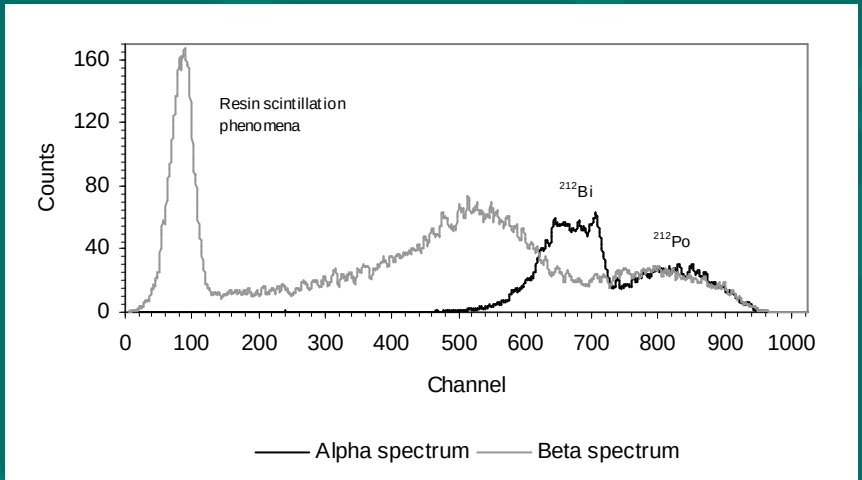
^{212}Pb activity

A difficult radionuclide to measure

- Pre-concentration
  - Uptake > 90 % for a range of natural and anthropogenic radionuclides.
  - Application for Pu and U measurements.



Multi-element uptake onto the MnO_2 resin

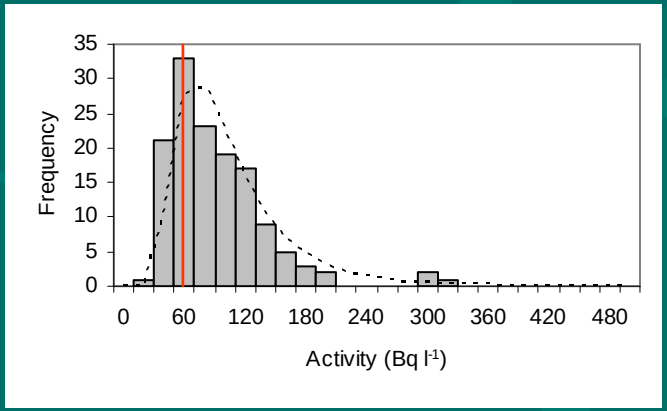


The alpha and beta spectrum of a ^{212}Pb sample

- Low-level detection
  - Measurement of ^{212}Pb is inferred from ^{212}Bi in the alpha spectrum.
  - This allows lower limits of detection.

^{212}Pb activity

A difficult radionuclide to measure



^{212}Pb measurements

- Identification of a significant ^{212}Pb component.
- During March - June 2006:
  - 137 measurements
  - 5 - 530 Bq m^{-3}
  - GM = 71 Bq m^{-3}

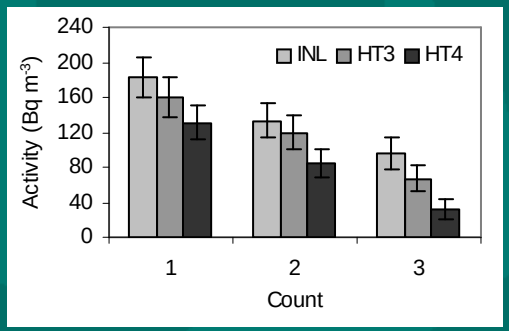
- No direct correlation with gross alpha activity:
  - Sample differences.
  - Decay of gross alpha activity.
  - Calibration issues.
  - No gross alpha > 100 Bq m^{-3}.

Activity excursions

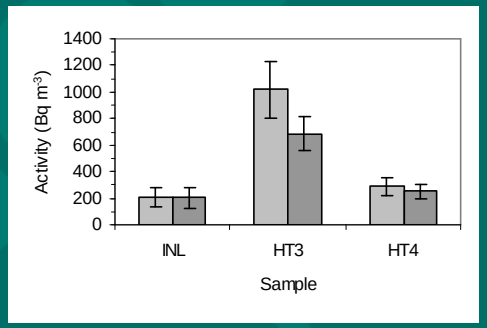
Identifying the ^{212}Pb component

- During March - September 2006:
  - 3 gross alpha measurements > 100 Bq m^{-3}.
    - Significant ^{212}Pb component.
    - No significant increase in U, Pu or Th activity.
    - Alpha/beta ratio of ~ 0.5.

- 5 July 2006



Gross alpha activity



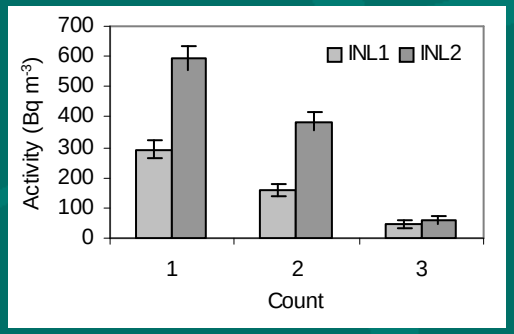
^{212}Pb activity

National Oceanography Centre, Southampton

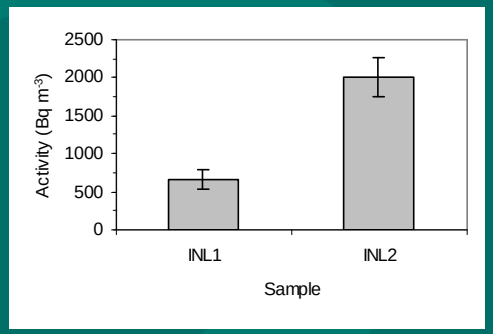
Activity excursions

Identifying the ^{212}Pb component

- 14 September 2006

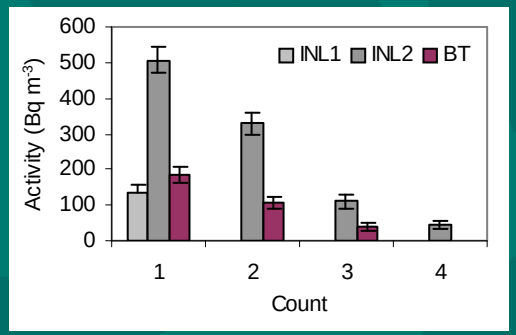


Gross alpha activity

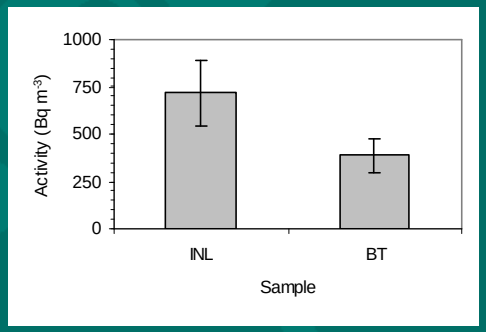


^{212}Pb activity

- 11 October 2006



Gross alpha activity



^{212}Pb activity

- Increased gross alpha activity is attributable to ^{212}Pb.

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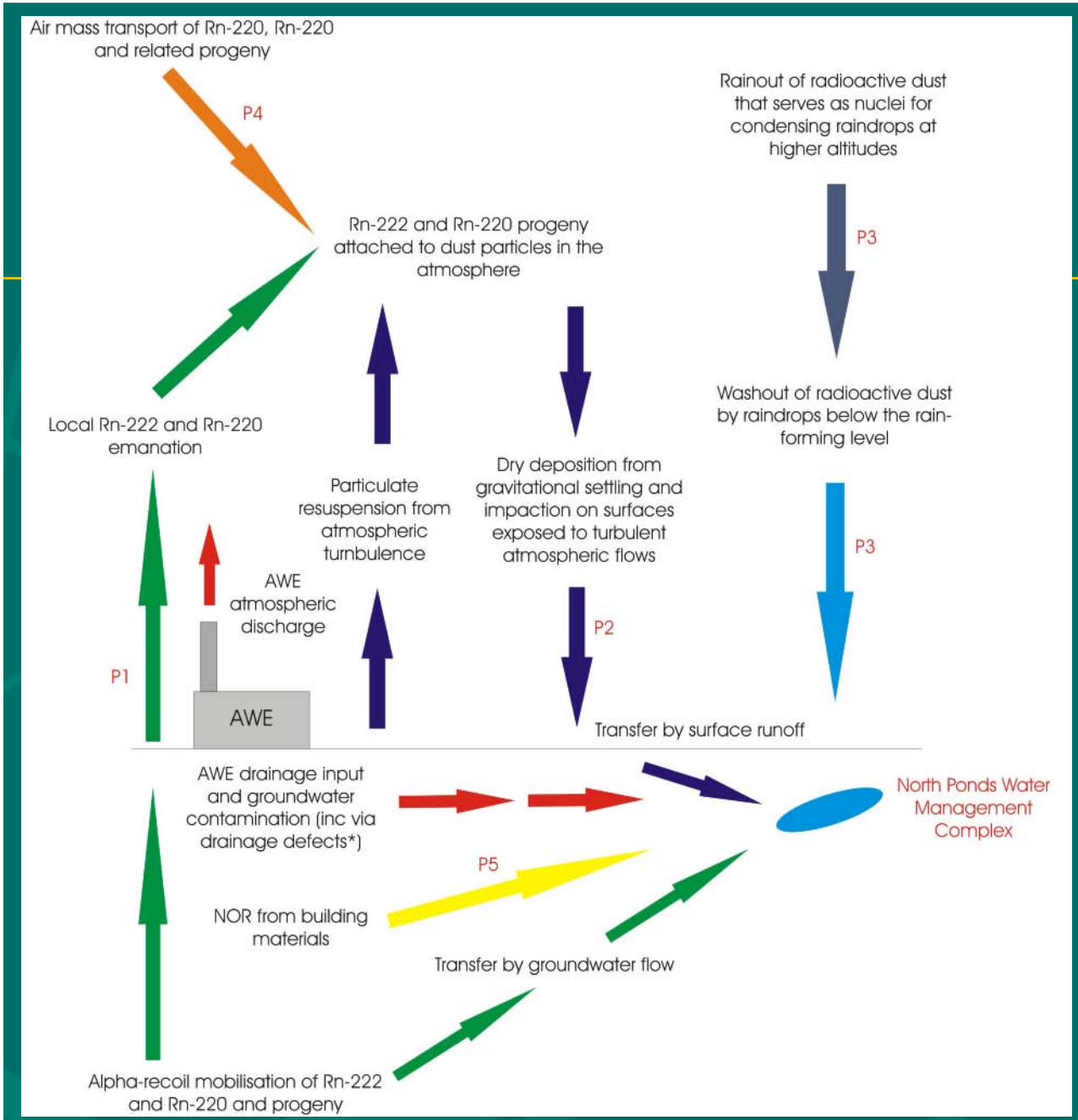


Transport pathways

- What is the origin of the ^{212}Pb activity?
  - Local emanation of ^{220}Rn
  - Air mass transport
  - Particulate resuspension
  - Groundwater flow
  - AWE discharges
- Dry deposition or wet deposition?

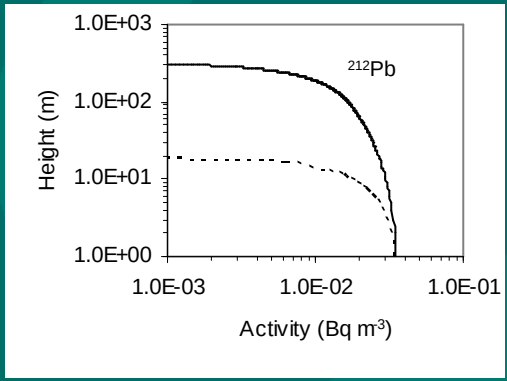
Activity excursions

Origin of the ^{212}Pb activity

- Increased ^{212}Pb activity is observed during periods of heavy rainfall.
  - Does the rainfall:
    - Directly remove ^{212}Pb from the atmosphere (wet deposition)?

    OR
    - Transfer ^{212}Pb deposited on the ground (dry deposition)?

    OR
    - Combination of both wet and dry deposition?



Modelling using the atmospheric diffusion equation

- Rainfall must transport ^{212}Pb already deposited on the ground into the surface waters.
  - Wet deposition unlikely as the majority of ^{212}Pb is below the rain-forming level.
  - Size distribution difficult to remove by wet deposition.
  - No increase in rainwater activity for 46 % of gross alpha measurements > 100 Bq m^{-3}.

Conclusions

- The routine monitoring of surface waters is complicated by the occurrence of short-lived NOR.
  - Dominated by NOR from the ^{238}U and ^{232}Th series.
  - A significant contribution from ^{212}Pb.

- Sudden increases in gross alpha activity may obscure the screening measurements for uranium and plutonium releases.
  - Attributable to natural ^{212}Pb.
  - The ^{212}Pb originates from ^{220}Rn produced by local soil emanation.
  - Dry deposition removes ^{212}Pb from the atmosphere. Deposition is enhanced during fine weather.
  - The deposited ^{212}Pb is washed into the drainage system under conditions of heavy rainfall.

Conclusions

- A technique has been developed to determine the ^{212}Pb activity.
  - Rapid ^{212}Pb determination allows quick identification of the cause of the increased gross alpha activity.
  - This helps exclude any possibility of contamination by U or Pu isotopes.
  - Provides re-assurance when discharging waters from the North Ponds Water Management System.
  - The unique MnO_2 resin has applications for improving U and Pu measurements.
- Published research will be produced.

Acknowledgements

- Dr Richard Greenwood, Dr Ian Croudace and Dr Phil Warwick.
- ASc Analytical Sciences Group.
- Other colleagues at AWE.
- AWE Technical Outreach Programme.