NAME III Documentation

Implementation of decay chain modelling within NAME III

Author

P. Bedwell[1]

[1] HPA-RPD, Chilton, Didcot, Oxfordshire, OX11 0RQ

1. Introduction

Whilst NAME III version 5.0 calculates radioactive decay, it does not currently generate decay products. The objective is to include the ingrowth of daughter products from parent radionuclides in the Met Office's NAME III Model.

Note that the development of the decay chain modelling within NAME III occurred in my own space. The development version can be found out: file:/home/h05/appb/Version5_2a. All documentation can be found in: file:/home/h05/appb/Decay_Chain_Mod. All the links below (barring any in the Summary Section at the end of the note) can be found under the above filepaths.

2. Method for resolving the ingrowth of daughter products

In many radiological assessments involving exposure to radionuclides, it is necessary to take into account the effects of radioactive decay i.e. decay products arising from the parent radionuclide. The timescales considered in an assessment, and the half-lives of parent radionuclides and decay products, will impact on the decay and ingrowth of radionuclides in an assessment.

In general the serial transformation by radioactive decay of each member of a radioactive series is described by the Bateman equations (the derivation of which can be found in: Fitzgerald et al, 1967). Assuming that at time zero the activity of the parent nuclide is A^0 and the activity of all daughters is zero, the activity at time *t* of a chain member i is,

$$A_i(t) = A^0 \left(\prod_{j=1}^{i-1} f_{j,j+1} \lambda_{j+1} \right) \sum_{j=1}^{i} \frac{e^{-\lambda_j t}}{\prod_{\substack{k=1 \\ k \neq j}}^{i} (\lambda_k - \lambda_j)} \qquad (1)$$

where t is the time or time step (s), λ is the decay constant (s^{-1}), whereby $\lambda = (\ln 2)/T_{1/2}$, where $T_{1/2}$ is the half-life (s). Note that $f_{j,j+1}$ denotes the fraction of the nuclear transformations of chain member j forming member j+1, also known as the branching ratio. Furthermore note that this equation is slightly modified to that detailed in the HPA's MDH (Radionuclide Data Chapter), which is in error (see Appendix A for details).

Thus the activity of the parent radionuclide is described by:

$$A_1(t) = A_1(0) e^{-\lambda_1 t} \qquad (2)$$

And the activity of the first daughter product decaying in from the parent radionuclide is described by:

$$A_2(t) = \frac{A_1(0)\lambda_2}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \qquad (3)$$

Many models (such as ESCLOUD and PC CREAM) developed by HPA only consider the decay from the parent and the first daughter product and therefore only implement equations 2 and 3 and do not implement the general solution to the Bateman equations, as described by equation 1. This is because in the majority of cases it is sufficient to consider a single parent/daughter relationship (Jones, 1980). However one of the radionuclides recommended for inclusion in NAME III by HPA consists of a decay chain of 5 radionuclides (^{226}Ra decays to ^{222}Rn, which subsequently decays to ^{218}Po, both of which do not contribute significantly to dose, however ^{218}Po decays to ^{214}Pb, which subsequently decays to ^{214}Bi, both of which have the potential to contribute significantly to dose. Equation 1 is clearly the more rigorous approach, especially if further long decay chains wish to be added at a later date. As it is

envisaged that equation 1 would be relatively straightforward to implement, this approach is implemented in NAME III.

3. Implementation of method

There are two steps in implementing decay chain modelling in NAME, analogous to the approach used in NAME to implement radioactive decay. Step 1 is to implement decay chain modelling during the process of atmospheric dispersion. Step 2 is to implement decay chain modelling following the deposition of radioactivity onto the ground. Two separate steps are required here because NAME III utilises a set of routines which consider the concentration (mass) on particles during dispersion and in a separate set of routines considers a fraction of the concentration imparted from the particles onto the ground (but the particle is not deposited). **ONLY** the first of these two steps have been implemented to date.

STEP 1 – Decay chain modelling during atmospheric dispersion

The three sections of the NAME III code which require updating as a result of incorporating the decay chain modelling (during atmospheric dispersion) are:

i) The Input.F90 module i.e. which reads in data, notably daughter and branching ratio specific data, from the NAME input file.
ii) The Species.F90. module, which defines the format of the decay chain modelling parameters for use in the decay chain modelling calculations.
iii) The Particle.F90 module, which is used to perform the decay chain modelling calculations (by way of a subroutine, 'RadioactiveDecay').

Work Plan

After identifying the (Bateman) approach to be implemented, the subsequent steps to be undertaken to implement this approach in NAME III include:

a) Action - Take the fortran code describing the calculation of radioactive decay from the parent and produce a standalone program which implements this code. *Done – program can be found at: H:\Decay_Chain_Mod\RadDecay_v1_0.F90 and RadDecay_v1_0.exe.*

b) Action – Expand upon the standalone program containing fortran code describing the calculation of radioactive decay from the parent and produce a standalone program which calculates radioactive decay from the parent and subsequent daughter products in the decay chain. This should include testing. *Done – program can be found at: RadDecay_v1_3.F90 and RadDecay_v1_3.exe.*

Table 1. Comparison of the activities (Bq) estimated by RadDecay_v1_3 (developed at the Met Office, 2008) and the HPA's Progeny 4 spreadsheet, for a range of radionuclides and timesteps.

	RadDecay_v1_3			Progeny 4.xls		
	1 hour	10 days	1 year	1 hour	10 days	1 year
^{222}Rn	9.93E-01	1.63E-01	1.75E-29	9.93E-01	1.63E-01	1.83E-29
^{218}Po	9.92E-01	1.63E-01	1.75E-29	9.93E-01	1.63E-01	1.83E-29
^{214}Pb	7.58E-01	1.64E-01	1.76E-29	7.43E-01	1.64E-01	1.84E-29
^{214}Bi	4.91E-01	1.65E-01	1.77E-29	4.69E-01	1.65E-01	1.85E-29
^{135}I	9.00E-01	1.17E-11	-	9.04E-01	1.18E-11	-
^{135}Xe	5.89E-02	2.54E-08	-	5.68E-02	2.54E-08	-
^{215}At	0.00E+00	-	-	0.00E+00	-	-
^{211}Bi	2.83E-15	-	-	6.15E-15	-	-

The decay chain's above were selected because they scoped a range of decay scenarios, including a decay chain whereby the parent is long lived relative to the daughter (e.g. ^{222}Rn and ^{218}Po), a decay chain whereby the parent and daughter have similar half-lives (^{135}I and ^{135}Xe) and a decay chain whereby the parent is short lived

relative to the daughter (^{215}At and ^{211}Bi). Furthermore the ^{222}Rn was chosen as it consists of a relatively large number of decay products and ^{135}I was chosen because the branching ratio to ^{135}Xe is not equal to 1.
Table 1 generally shows very good agreement between the two modes of calculating the activity in the parent and decay products. Note that at t=0 the activity of the parent is assumed to be 1 Bq. The largest difference is in the estimated activity of ^{211}Bi. This estimation is very sensitive due to the relative short time period over which the calculation is made and the very short half life of the parent radionuclide. Upon performing a hand calculation it is found that the estimated activity of ^{211}Bi is in agreement with RadDecay.

c) Action – Update NAME III such that it can calculate the activity from a parent radionuclide and all of it's daughters for any non-branching decay chain, including the consideration of a single path of a branching decay chain, during the process of atmospheric dispersion (not deposition).

NAME III has been coded so that it can calculate the activity from a parent radionuclide and all of it's daughters for any non-branching decay chain, including the consideration of a single path of a branching decay chain. NAME III accounts for the branching ratio but the activity from the secondary branch is currently assumed to be lost. For example if you were modelling a release of Sr-91 to Y-91M, NAME would account for the branching ratio from Sr-91 to Y-91M but ignore the branching ratio to the other daughter of Sr-91, Y-91, and therefore the activity decaying from Sr-91 to Y-91 would be lost from the assessment. To work around this problem, two sources can be defined in NAME e.g. Sr-91 decaying to Y-91M and Sr-91 decaying to Y-91 but both sources of Sr-91 must be described by a different species name. The user could not consider a single source of Sr-91 decaying to BOTH Y-91M AND Y-91.

Stephanie Haywood, Jane Simmonds and Mary Morrey (of HPA-RPD) were all in agreement that there is currently no need to include the ability to account for the decay of a radionuclide to two (or more) daughter products, as there are no significant accidental release radionuclides which have two radiologically significant branches. This was detailed in an email from Stephanie Haywood dated: 7th August 2008, and titled: factor of 4 diff and additional runs.

Note that in many cases whereby the parent predominantly decays to one (of the two) daughter i.e. +99% to one daughter and <1% to the other daughter, it is reasonable to assume decay to only one daughter product, for example, U-231, decays to Pa-231 with a branching ratio of 9.9995E-01 and to Th-227 with a branching ratio of 5.50E-05. Where the branching ratio is of similar size between the two daughters e.g. Sr-91 which decays to Y-91M (with a branching ratio of 0.578) and Y-91 (with a branching ratio of 0.422), consideration of the radiological significance of the daughter products must be accounted for. It may be the case that despite having similar branching ratio's one radionuclide is much more radiologically significant than the other and therefore only one of the branches need be considered in NAME. Therefore it is likely that there would only be a very limited number of cases whereby HPA would wish to consider both daughters branching from a single parent.

Below is a summary of how the NAME code has been modified to account for decay chain modelling. The files can be found in: file:/home/h05/appb/Version5_2a/Code_NameIII. Original files are named …..01.F90. Newly updated files are named ……F90.

GlobalParameters.F90

Only update was to define the MaxDecayChainLength (=15), which is subsequently used in the species module. Thus the maximum number of radionuclides currently permitted in a single decay chain is 15.

MainNAMEIII.F90

Calls the subroutine SetUpSpecieses_DecayChains(Specieses) from the module, Species.F90, which sets up full decay chains in Specieses.

Input.F90

In Subroutine SpeciesInputNames(HFBlockForms) added 'Daughter' and 'Branching Ratio', with defaults set to blank.
In Subroutine Tokens2Species(Tokens, Specieses) added in the argument list HasDaughter and BranchingRatio. Added some checks to ensure that if the user enters a daughter product in the input file, they must also enter a branching ratio; a value for the branching ratio must not be entered if a daughter product is not entered; a daughter product must not be entered for a stable species.

Species.F90

Define SetUpSpecieses_DecayChains(Specieses), which sets up full decay chains in Specieses, and is called from the module, MainNAMEIII.F90.
Defined new arguments: HasDaughter, Daughter, BranchingRatio, DecayChain(MaxDecayChainLength) and DecayChainLength which are all included in the Type, 'Species_', which is an array (with the power to consider different types of arguments) detailing all the parameters required to describe a single species.
Added the arguments: HasDaughter, Daughter and BranchingRatio to the argument list for the Function 'InitSpecies' which initialises the species so that parameter information describing each species can be passed to Particle.F90. Subsequently, some further checks are added to ensure that errors do not creep into runs, including if the name of the daughter entered in the input file has too many characters and if the branching ratio entered does not fall in the range 0 to 1, inclusive.
Added a whole new Subroutine, 'SetUpSpecieses_DecayChains(Specieses)', which sets up full decay chains in 'Specieses' (contains all the species and their associated data). A 'do loop', loops over all species and constructs a decay chain for those species having a daughter product. In doing so, the length of each decay chain is also identified.

Particle.F90 (module provides code to treat particles)

The subroutine, 'RadioactiveDecay', within this module is the part of the code which does the number crunching for the radioactive decay calculation.

Defined new local variables: an array 'New_Mass' (which has elements initially set to zero), an integer 'iDaughter' and also a number of reals, 'D', 'E' and 'A' to break down the radioactive decay into bite-sized calculations, where

D = PRODUCT(decayconstant_k - decayconstant_j) from k = 1 to i but not for k = j.

*E = SUM((EXP^-decayconstant_j*RDt)/D) from j=1 to i.*

*A = PRODUCT(decayconstant_j+1*BranchingRatio_j,j+1) from j = 1 to i - 1.*

*Note that decayconstant = Log(2.0)*InvHalfLife. RDt = timestep.*

The first do loop (present in the code pre v5_2a), loops over all the species in the decay chain. The second do loop (added here), loops over all the products in each decay chain. Note, if there exists 2 parent radionuclides, each with one daughter, this loop is performed 6 time:. firstly looping over parent 1 and finding the activity of parent 1 as a result of the decay over that timestep; then looping over daughter 1 (of parent 1) and finding the activity of daughter 1 as a result of the decay from the parent over that timestep; then

looping over parent 2 and finding the activity of parent 2 as a result of the decay over that timestep; then looping over daughter 2 (of parent 2) and finding the activity of daughter 2 as a result of the decay from the parent over that timestep; then looping over daughter 1 (of parent 1) and finding the activity of daughter 1 as a result of the decay of daughter 1 over that timestep; and finally looping over daughter 2 (of parent 2) and finding the activity of daughter 2 as a result of the decay of daughter 1 over that timestep. j1, j2 and k1 return the position/number of the species in the list of species in the input file required for the calculation in the loop. For example, consider the line of code:

j1 = Specieses%Specieses(iSpecies)%DecayChain(j)

Consider (from the example in the paragraph above) the following species provided in an input file in this exact order:

Parent 1
Parent 2
Daughter 1 (of P1)
Daughter 2 (of P2)

1st loop
Do iSpecies = 1, Specieses%nSpecieses = 4
Do iDaughter = 1, Specieses%Specieses(iSpecies = 1)DecayChainLength = 2
Do j = 1, iDaughter = 1
j1 = Specieses%Specieses(iSpecies = 1)%DecayChain(j = 1)
therefore j1 = 1

2nd loop
Do iSpecies = 1, Specieses%nSpecieses = 4
Do iDaughter = 1, Specieses%Specieses(iSpecies = 1)DecayChainLength = 2
Do j = 1, iDaughter = 2
j1 = Specieses%Specieses(iSpecies = 1)%DecayChain(j = 1)
therefore j1 = 1

3rd loop
Do iSpecies = 1, Specieses%nSpecieses = 4
Do iDaughter = 1, Specieses%Specieses(iSpecies = 1)DecayChainLength = 2
Do j = 1, iDaughter = 2
j1 = Specieses%Specieses(iSpecies = 1)%DecayChain(j = 2)
therefore j1 = 3

4th loop
Do iSpecies = 1, Specieses%nSpecieses = 4
Do iDaughter = 1, Specieses%Specieses(iSpecies = 2)DecayChainLength = 2
Do j = 1, iDaughter = 1
j1 = Specieses%Specieses(iSpecies = 2)%DecayChain(j = 1)
therefore j1 = 2

5th loop
Do iSpecies = 1, Specieses%nSpecieses = 4
Do iDaughter = 1, Specieses%Specieses(iSpecies = 2)DecayChainLength = 2
Do j = 1, iDaughter = 2
j1 = Specieses%Specieses(iSpecies = 2)%DecayChain(j = 1)
therefore j1 = 2

6th loop
Do iSpecies = 1, Specieses%nSpecieses = 4
Do iDaughter = 1, Specieses%Specieses(iSpecies = 2)DecayChainLength = 2
Do j = 1, iDaughter = 2

j1 = Specieses%Specieses(iSpecies = 2)%DecayChain(j = 2)
therefore j1 = 4

etc…

The code calculates D, then E and then A, which are then applied to the formulae:

*New_Mass(i) = New_Mass(i) + Mass(iSpecies) * A * E*

This allows 'mass' in a single timestep from the decay of a parent to daughter product to be summed to the 'mass' in the same timestep resulting from the decay of that daughter, for example.

After looping over all species and calculating 'New_Mass(i)' for each species over a single timestep, all the elements of the array 'New_Mass(i)' are assigned to the array 'Mass' (of the same dimension), ready for the next timestep.

The code implemented in NAME III could be made more efficient by modifying the calculation of the decay constant (ln 2/half life), which is currently performed many times for a single radionuclide throughout the calculation of the radioactive decay. However the decay constant only need be calculated once at the beginning of the code and used thereafter.

d) Action – Consider the radionuclides and radionuclide data to be added to NAME III

Version 5.1 of NAME III already includes the radionuclides recommended by HPA as detailed in Radionuclides_HPA_list.xls.

In an effort to identify the extent of the radionuclides (namely decay products) which may be required for use in NAME III, a spreadsheet has been developed. This spreadsheet includes all the radionuclides included in NAME III v5.1 and their respective decay products. The spreadsheet details all of the branching ratios and 1st daughter products of the radionuclides included in NAME III v5.1 and the radionuclides recommended for addition to NAME III. The data incorporated in this spreadsheet has been copied from the HPA's Progeny4.xls. All data has been added to Species.txt and subsequently tested by HPA (see below for details). The spreadsheet considers instances whereby decay chains branch and then rejoin, the implication of which is that a single radionuclide may be included in multiple decay chains. This spreadsheet can be found at: Species.xls. The updated Species.txt file can be found at: H:\Version5_2a\Resources\Defns\Species.txt..

Additions to Species.txt
Daughter and Branching Ratio values have been added for all radionuclides included in Species.txt applicable to NAME III v5.1. Additional radionuclides and their respective details have been added, such that for all the radionuclides included in Species.txt for NAME III v5.1, their daughters and all subsequent daughters have been incorporated. Note that this does not necessarily mean that all decay chains are complete as there may be radionuclides which are parents of the radionuclides included in Species.txt applicable to NAME III v5.1, which are not included. Note that not all of the radionuclides added to Species.txt will be radiologically significant.

Under the 'Radionuclide list with surface resistances' all entries under the columns the 'Daughter' and 'Branching Ratio' were updated for those radionuclides originally in Species.txt but no new radionuclides have been added to this section (because it is not clear which surface resistance values should be applied to the newly added radionuclides).

Under the ‘‘Radionuclide list using deposition velocities as suggested by HPA-RPD’ section all entries have been updated under the ‘Daughter’ and ‘Branching Ratio’ columns for those radionuclides originally in Species.txt and all new radionuclides have been added with their associated data.

Species.txt has been tested by HPA as a result of these updates. Below is a summary of the test plan and test report.

Test Plan

Nothing needs testing before the line:
Radionuclide list using deposition velocities as suggested by HPA-RPD.

1. From Antimony-127 to TEST-NUCLIDE please check, under the column heading 'Daughter', that the correct radionuclide has been entered with respect to the parent radionuclide detailed under the column heading 'Name'. Note that if the 'Daughter' column is blank, this should imply that the parent radionuclide, under the column heading 'Name', decays to something stable - check. Check that the daughter radionuclide is spelt correctly. Check, under the column heading 'Branching Ratio', that the correct branching ratio has been entered with respect to the parent and daughter product detailed on the same row. Check that for each parent species, under the column heading 'Name', all the daughters have been included for that species (whereby the branching ratio is greater than 1.00E-02).

2. From Actinium-225 to Yttrium-91M please check all the items checked under 1. In addition, check the spelling of the parent radionuclides under the 'Name' column heading and check that the correct half-life has been entered under the 'Half Life' column heading with respect to the parent radionuclide detailed under the column heading 'Name'.

Test Report

Discrepancies:

i) The main one is that in the line for THORIUM-234, the daughter is given as PROTACTINIUM-234, whereas it should be PROTACTINIUM-234M. Identified, JW, 11/09/08. Corrected, PB, 03/10/98.

ii) Other than that, there were just a few half-lives which seemed to be very slightly different from the figures I got from ICRP-38. These are listed below. I should point out that in all the cases below, I had to convert my figures from years (which is how they appear in ICRP-38) to seconds, so it could be that the discrepancy is due to a rounding effect, and that your original figure is in fact correct.

Species:	*Your half-life:*	*My half-life:*
PROTACTINIUM-231	*1.04E+12*	*1.03E+12*
THORIUM-232	*4.45E+17*	*4.43E+17*
URANIUM-233	*5.02E+12*	*5.00E+12*
URANIUM-236	*7.38E+14*	*7.39E+14*

Identified, JW, 11/09/08. No update required, PB, 03/10/98.

iii) Also: there were two lines which I couldn't check (because I wasn't sure what they meant). These were "IODINE-TRACER" and "TEST-NUCLIDE".

Identified, JW, 11/09/08. No update required, PB, 03/10/98. End of Test Report.

Comments on Species.txt

Note that where the 'Daughter' and 'Branching Ratio' columns are left blank, this implies that the decay product is stable. If a radionuclide decays to more than one daughter, then all decay products are included, with the respective branching ratios on separate rows. The only exception is if the branching ratio is less than 1.00E-02, in which case the decay product is not considered (similar to the assumption made in ESCLOUD, Appendix A1). In Species.txt some isotopes of tellurium decay to isotopes of iodine. In such instances the decay products have been input as, for example, IODINE-129 and not IODINE_O-129 nor IODINE_E-129. The user must decide which form of Iodine they wish Tellurium to decay to or they could model both.

Note also that the surface resistances need updating and checking. Note also that IODINE_O and IODINE_E have (incorrectly) the same deposition properties in the 'Radionuclide list with surface resistances' section.

It is recommended that if HPA wishes to model the dispersion of radionuclides associated with specific decay chains, NAME Input file 'templates' are setup which can be drawn upon when required. This would be especially beneficial for the longer decay chains.

e) Action – Testing version of decay chain modelling code implemented in NAME III which calculates the activity from a parent radionuclide and all of it's daughters for any non-branching decay chain.

Note that all of the values of parameters are treated as single-precision values i.e. using real(std) and not as double precision values i.e. real(kind = 2). It is possible that there may be issues with precision, for example if a very long-lived parent decays to a very short-lived daughter product, over relatively long time steps, however no issues have been identified to date.

All testing runs can be found in: file:/home/h05/appb/Decay_Chain_Mod/Runs

Tests i-xi below test that the code is correctly calculating the radionuclide activity as a result of radioactive decay and decay chain modelling.

i) NAMEIIIv1pt3a.txt. Test for a release of 1 Bq of Caesium-137 and ingrowth of its daughter product, Barium-137M. Timesteps = 30 seconds. Output to 1 hour.
The output was in agreement with the Progeny spreadsheet and hand calculations. Because of the long half life of the parent and very short half life of the daughter product, (barring the ingrowth of the daughter) the activity does not vary significantly over the course of 1 hour (hence the consideration of Tc-99M in ii).

ii) NAMEIIIv1pt3b.txt. Test for a release of 1 Bq of Technetium-99M and ingrowth of its daughter product, Technetium-99. Timesteps = 30 seconds. Output to 1 hour.
The output was in agreement with the Progeny spreadsheet and hand calculations.

iii) NAMEIIIv1pt3c.txt. Test for a release of 1 Bq of Technetium-99M and ingrowth of its daughter product, Technetium-99 and 1 Bq of Caesium-137 and ingrowth of its daughter product, Barium-137M. Timesteps = 30 seconds. Output to 1 hour. The radionuclides were specifically not entered into the NAME input file in decay chain order to test if the code could handle this.
The output was in agreement with the Progeny spreadsheet and hand calculations.

iv) NAMEIIIv1pt3d.txt. Test for a release of 1 Bq of Astatine-216 (which has a very short half-life) and ingrowth of its daughter product, Bismuth-212. Timesteps = 30 seconds. Output to 1 hour.

The output was in agreement with the Progeny spreadsheet.

v) NAMEIIIv1pt3e.txt. Test for a release of 1 Bq of Radon-222, decaying to Polonium-218, decaying to Lead-214, decaying to Bismuth-214, decaying to Polonium-214, decaying to Lead-210. Timesteps = 30 seconds. Output to 1 hour. This tested quite a long decay chain
The output was in agreement with the Progeny spreadsheet.

vi) NAMEIIIv1pt3f.txt. Test for a release of 1 Bq of Radon-222, decaying to Polonium-218, decaying to Lead-214, decaying to Bismuth-214, decaying to Polonium-214, decaying to Lead-210. Timesteps = 30 seconds. Output to 1 year. This tested quite a long decay chain to a relatively long output time.
The output was in agreement with the Progeny spreadsheet.

vii) NAMEIIIv1pt3i.txt. Test for a release of 1 Bq of TELLURIUM-127M, decaying to 'TELLURIUM-127a', and a release of 1 Bq of ANTIMONY-127, decaying to 'TELLURIUM-127b'. Timesteps = 30 seconds. Output to 1 hour. This test comprises of a suite of tests (vii-viii) to identify the best approach to model multiple radionuclides which decay to the same radionuclide.
The output was in agreement with the Progeny spreadsheet, but the user must sum the activity of 'TELLURIUM-127a', and 'TELLURIUM-127b'.

viii) NAMEIIIv1pt3j.txt. Test for a release of 1 Bq of TELLURIUM-127M, decaying to TELLURIUM-127, and a release of 1 Bq of ANTIMONY-127, decaying to TELLURIUM-127. Timesteps = 30 seconds. Output to 1 hour. This test comprises of a suite of tests to identify the best approach to model multiple radionuclides which decay to the same radionuclide.
The output was in agreement with the Progeny spreadsheet, and the code performed the summation of the activity of TELLURIUM-127 from the decay of TELLURIUM-127M and the activity of TELLURIUM-127 from the decay of ANTIMONY-127.

ix) NAMEIIIv1pt3k.txt. Test for a release of 1 Bq of Strontium-91, decaying to Yttrium-91m, and Yttrium-91. Timesteps = 30 seconds. Output to 1 hour. This test comprises of a suite of tests (ix-xi) to identify the best approach to model a single radionuclide which decays to multiple radionuclides.
The output for the parent and the first daughter (Yttrium-91m) was in agreement with the Progeny spreadsheet. But the output for the second daughter (Yttrium-91) was zero, as expected – the activity should in reality be non-zero but NAME has not been coded to calculate the activity of two daughters from a single parent. Note that where it refers to, "first daughter" within this paragraph, this implies the 1st daughter product identified in the 'Daughter' column heading and not the first daughter identified in the 'Name' column heading.

x) NAMEIIIv1pt3m.txt. Test for a release of 1 Bq of Strontium-91, decaying to Yttrium-91, and Yttrium-91m. Timesteps = 30 seconds. Output to 1 hour. This test comprises of a suite of tests (ix-xi) to identify the best approach to model a single radionuclide which decays to multiple radionuclides.
The output for the parent and the first daughter (Yttrium-91) was not in agreement with the Progeny spreadsheet but is in agreement with hand calculations (therefore there may be an error in the Progeny spreadsheet – hand calc and NAME run estimated the activity of Y-91 after 1 hour to be 2.0065E-04 Bq following a 1 Bq release of Sr-91. The Progeny spreadsheet does not consider this form of radioactive decay (Sr-91 -> (42%) -> Y-91). Instead the Progeny spreadsheet considers Sr-91 decaying by the following method: Sr-91 -> (58%) -> Y-91M -> (100%) -> Y-91. In NAME III a 1 Bq release of Sr-91 results in 8.96E-05 Bq of Y-91 after 1 hour, as a result of an initial decay to Y-91M. In contrast the Progeny spreadsheet estimates the activity of Y-91 to be 8.35E-05 Bq, which is in reasonable agreement with

NAME. The output for the second daughter (Yttrium-91m) was zero, as expected – the activity should in reality be non-zero but NAME has not been coded to calculate the activity of two daughters from a single parent.

xi) NAMEIIIv1pt3n.txt. Test for a release of 1 Bq of 'Strontium-91a', decaying to Yttrium-91m, and a release of 1 Bq of 'Strontium-91b', decaying to Yttrium-91. Timesteps = 30 seconds. Output to 1 hour. This test comprises of a suite of tests (ix-xi) to identify the best approach to model a single radionuclide which decays to multiple radionuclides.
The output for "both parents" and both daughters were in agreement the Progeny spreadsheet/hand calculations (as above). However you calculate output for Sr-91 twice – only one of these is required. An alternative approach, for a 1 Bq release of Sr-91, would be to set the branching ratios for both daughters to 1 and set the source strength for Sr-91a decaying to Y-91m and Sr-91b decaying to Y-91 to 0.578 and 0.422 Bq, respectively, thus the activity for Sr-91a and Sr-91b can be summed at each time step (as undertaken in NAMEIIIv1pt3p.txt).

To avoid confusion NAME III has been coded such that the same radionuclide cannot be entered twice, as occurred in the three examples above. In a single run the decay of a single radionuclide to multiple decay products can be considered (e.g. Sr-91 -> (42%) -> Y-91 AND Sr-91 -> (58%) -> Y-91M -> (100%) -> Y-91) but two sources need to be defined by way of two different species names e.g. Sr-91a and Sr-91b to estimate the total activity of Y-91 from both forms of decay (e.g. in NAMEIIIv1pt3m2.txt).

Tests xii-xix below test that the code suitably handles errors (specific to decay chain modelling) resulting from erroneous NAME input files. The run used in test i (NAMEIIIv1pt3a.txt) is used as the baseline test run here.

xii) NAMEIIIv1pt3r.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to "a1". Timesteps = 30 seconds. Output to 1 hour.
The name of the daughter radionuclide can be anything and is only restricted by the number of characters used. Thus the user can enter throughout the input file, including under the daughter column heading, the name of a nuclide eg "a1" which isn't actually a radionuclide. Furthermore the user could enter the wrong daughter name e.g. that Cs-137 decays to Sr-90 (for example) and NAME will still run. Note however that this shouldn't happen if the details are copied from Species.txt.

xiii) NAMEIIIv1pt3s.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to "Barium-137m", but with only the branching ratio column populated and the daughter column blank. Timesteps = 30 seconds. Output to 1 hour.
Does not run. FATAL ERROR: Branching ratio must not be given when a daughter product is not given.

xiv) NAMEIIIv1pt3t.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to "Barium-137m", but with only the daughter column populated and the branching ratio column blank. Timesteps = 30 seconds. Output to 1 hour.
Does not run. FATAL ERROR: Branching ratio must be given for a daughter product.

xv) NAMEIIIv1pt3u.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to Caesium-137. Timesteps = 30 seconds. Output to 1 hour.
Does not run. FATAL ERROR: decay chain is too long for species "CAESIUM-137".

Note that the branching ratio can be entered in the form: 0.946 as well as 9.46E-01.

xvi) NAMEIIIv1pt3v.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to Barium-137m. The branching ratio is set to: 1.01E+00. Timesteps = 30 seconds. Output to 1 hour.
Does not run. FATAL ERROR in InitSpecies: Branching Ratio is given as 1.010000 and is outside the permitted range [0.0 -> 1.0].

xvii) NAMEIIIv1pt3w.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to Barium-137m. The branching ratio is set to: -9.46E-01. Timesteps = 30 seconds. Output to 1 hour.
Does not run. FATAL ERROR in InitSpecies: Branching Ratio is given as -0.9460000 and is outside the permitted range [0.0 -> 1.0]

xviii) NAMEIIIv1pt3x.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to Barium-137m. The branching ratio is set to: 'ten'. Timesteps = 30 seconds. Output to 1 hour.
Does not run. FATAL ERROR in InitSpecies: Branching Ratio is given as -1.0000000 and is outside the permitted range [0.0 -> 1.0]

xix) NAMEIIIv1pt3y.txt. Test for a release of 1 Bq of 'Caesium-137', decaying to Barium-137m, but Barium-137m is not included under the column heading, "name". Timesteps = 30 seconds. Output to 1 hour.
Does not run. FATAL ERROR: species "BARIUM-137M" not found

Tests xx- below tests the impact the decay chain modelling has on run time.

xx) NAMEIIIv1pt3z.txt. Test for a release of 2.8e12 Bq/s of 'Caesium-137', over 1 hour and including 5,000,000 particles.
Real time (elapsed time) = 9258.37s (2 hours 34 minutes 18 seconds);
User time (elapsed time – minus other jobs; processing cpu time) = 9242.25s (2 hours 34 minutes 02 seconds);
(System time = 7.92s).

xxi) NAMEIIIv1pt3za.txt. Test for a release of 2.8e12 Bq s^{-1} of 'Caesium-137', decaying to Barium-137m. The release duration = 1 hour and includes a release of 5,000,000 particles.
Real time (elapsed time) = 10185.08s (2 hours 49 minutes 45 seconds);
User time (elapsed time – minus other jobs; processing cpu time) = 10167.95s (2 hours 49 minutes 27 seconds);
(System time = 8.79s).
Note that no deposition rate values were output for either Cs-137 or Ba-137m when running in version 5.2 but they are estimated when running in the development version v5_2a.

xxii) NAMEIIIv1pt3zb.txt. Test for a release of 2.8e12 Bq s^{-1} of the decay chain of 'Plutonium-242', to Polonium-210, including 15 radionuclides in total. The release duration = 1 hour and includes a release of 5,000,000 particles.
Real time (elapsed time) = 142220s (39 hours 30 minutes 20 seconds);
User time (elapsed time – minus other jobs; processing cpu time) = 142046s (39 hours 27 minutes 26 seconds);
(System time = 107s).
The run time is very large in this example. In each output file there are 90 columns of data – 6 per radionuclide – for 10 min average, 1 hr average and 3 hr integration, for both air concentration and dry deposition. Grid points are at every 100m downwind for 20 km and off axis too. There are a total of 19 output files, 1 for each ten minutes post release up to 3 hours post release. Apart from the 1st few files the files are each 35-40 MB in size – storage of such large files is also an issue. However many of the calculations undertaken are redundant in this example as the TIAC to 3 hrs is considered but the half life of the parent, Pu-242, is approx 1.0E+13 seconds or approx 1.0E+09 years – thus there is very little decay over this period to the first

daughter product. In fact the TIAC values of 10 of the radionuclides, from Thorium-230 to Polonium-210, are all zero. The TIAC values of Uranium-238, Protactinium-234m and Uranium-234 were all negative – this is likely due to the imprecision in the calculations inherited from the very large half life of the parent, Pu-242.

In summary, decay chain modelling will increase runtime, and is likely to significantly increase runtime for two or more decays in a single chain (i.e. chains of three or more radionuclides). However there are few decay chains with daughters which are likely to be radiologically significant, especially akin to example xxii which is not a realistic example due to the very large half life of the parent radionuclide, Pu-242, and the relatively short timescales of interest in an emergency scenario. Therefore it is key that before applying decay chain modelling in NAME III, consideration of the radiological significance of the daughter products is made.

f) After some further checks by Dave Thomson a few more updates were made (in the Subroutine, “Radioactivedecay”):
   i) the first “if test” was modified to, “If (Mass(iSpecies) > 0.0)”. If the mass is zero there will be no contribution to activity and this avoids extra computational cost if some particles are only carrying a few species.
   ii) changed "Mass =" to "NewMass =" in the 'ND' if block. Otherwise the calculated value will be overwritten at the end of the routine. At the end of the Subroutine, “Radioactivedecay”, updated Mass if species = 'ND' and changed Mass(:) to Mass(iSpecies) since otherwise all species are updated if any one is radioactive.
   iii) In the Subroutine, “Radioactivedecay” D, E and A were updated to be of double precision (for consistency).
   iv) Dave highlighted that stable isotopes at the end of a decay chain wont be produced because their Masses are not updated. However, Pete advised that this was not an issue, at least from a Radiation Protection perspective, because stable isotopes will not contribute to dose and therefore are not of interest to the HPA. Dave advised that if this were to be resolved, there doesn't seem any easy fix except to treat all species as radioactive or possibly to attach a flag to each species to indicate if its either radioactive or is produced by radioactivity.

STEP 2 – Decay chain modelling during deposition

g) Action – Update NAME such that it can calculate the activity from a parent radionuclide and all of it's daughters for any non-branching decay chain, including the consideration of a single path of a branching decay chain, during the process of deposition (not atmospheric dispersion).

To date this action has not been completed and decay chain modelling has not been included for deposited material in NAME III. *Issues concerning the inclusion of decay chain modelling during deposition and how this may be achieved are considered below.*

It is important to note that for the output parameter deposition (Bq m^{-2}), there exist a number of key ‘removal’ processes in addition to radioactive decay (such as resuspension of activity into the atmosphere and the transfer of activity through the terrestrial environment e.g. via root uptake, surface washoff and infiltration through the soil). Therefore if decay chain modelling were to be added to NAME without the consideration of these additional removal processes, estimations of deposition concentration are likely to become less and less reliable as a function of time after deposition. There is nothing which can or should be modelled differently in NAME III but this is something to be aware of. What is important is that NAME III can integrate over deposition rate for any user defined period of time (not just from the start of the

release). Note that this issue does not impact upon the output parameter, deposition rate (Bq s^{-1} m^{-2}).

Below is a summary of how the NAME III code calculates decay in deposited material in NAME version 5.1:

*Currently a decay factor is calculated for each species in the Species.F90 module and Subroutine CalcDecayFactor (at the end of the module). This bit of code (utilising the Time.F90 module) says that to infinity the decay factor = 0 i.e. everything will have decayed to time = infinity; for a very short timestep the decay factor = 1 i.e. no decay has had chance to take place; otherwise the timestep = RdT and DecayFactor = Exp(- RdT * Species%InvHalfLife * LOG(2.0)). This decay factor is then applied in the calculation of the decay of deposited activity in the Output.F90 module. DecayFactor is of type real and therefore can only store one value at any one time – therefore I assume the DecayFactor is calculated for one species at a time.*

Initially dry deposition is calculated in Particle.F90, Subroutine DryDeposition. This uses the surface resistance or deposition velocity to calculate the deposition resulting from turbulent flow and also calculates the sedimentation (gravitational settling). Plume depletion is included. The subroutine considers a single particle at a time – if the particle incurs gravitational settling the code "Returns" out of the subroutine and does not consider deposition from turbulence. It is apparent that particles incurring gravitational settling, will deposit all of their mass and the particle is effectively killed. Particles which incur deposition from turbulent mixing, lose mass (but not necessarily all of their mass) and continue to disperse. The code calculates deposition if there are particles below the dry deposition height in that timestep, for all species considered and then for all particles with mass greater than 0. In all instances the deposition is passed out of this routine as a deposition rate.

Initially wet deposition is calculated in Particle.F90, Subroutine WetDeposition. Firstly the scavenging (deposition) coefficients are determined. These are a function of orographic enhancement, dynamic/convective washout and particle height (below the cloud base, between the cloud base and the cloud top and above the cloud top). Deposition rates are then calculated. Plume depletion is included. Particles which incur washout, lose mass (but not necessarily all of their mass). The code calculates wet deposition, for all species considered and then for all particles with mass greater than 0. In all instances the deposition is passed out of this routine as a deposition rate.

From Particle.F90 the deposition rates are passed to Case.F90 (the subroutine DryDeposition is called and the subroutine WetDeposition is called). The deposition values are then fed into the subroutines CalcParticleResults and CalcPuffResults. The dry and wet deposition values are then passed to Output.F90. Here they are averaged/integrated (in Subroutine Field2AvInt) (to get deposition values as opposed to deposition rates). The Subroutine CalcDecayFactor is called from Species.F90 and then applies the decay factor. Primarily the DecayFactor is dependent upon the species.

To date, NAME III, when considering a decay chain eg Cs-137 decaying to Ba-137M, considers the deposition of both the parent and all subsequent daughter products (assuming that "Dry Deposition?" And/Or "Wet Deposition?" are set to "Yes" in the "Sets of Dispersion Options:" block) when a request for deposition rate, dry deposition rate or wet deposition rate is made. If the column key (heading), "Decay deposition?" is set to "No" then NAME III can consider radioactive decay and decay chain modelling during dispersion for all parents and daughter products but NAME does not account for the radioactive decay of the deposited material for any radionuclides. If the column key (heading), "Decay deposition?" is set to "Yes" then NAME III can consider the radioactive decay and decay chain modelling during dispersion and the radioactive decay for deposited material for all parents and daughter products but NAME does not account for decay chain modelling of the deposited material for any

radionuclides e.g. for deposited material NAME III accounts for the radioactive decay of deposited Cs-137 and the radioactive decay of deposited Ba-137M but it does not account for the ingrowth of Ba-137M from deposited Cs-137. Note that by default, "Decay deposition?" is set to "Yes".

4. Summary

NAME III can account for decay chain modelling during atmospheric dispersion of radioactive material. However NAME III does not currently account for decay chain modelling of radioactive material deposited on the ground.

Future work

HPA should consider if they require decay chain modelling of radioactive material deposited on the ground. HPA should also consider whether it would be of use to consider other key 'removal' processes in addition to radioactive decay (such as resuspension of activity into the atmosphere and the transfer of activity through the terrestrial environment e.g. via root uptake, surface washoff and infiltration through the soil).

In addition, HPA and/or the Met Office may wish to update and finalise the surface resisitance values for all radionuclides included in Species.txt.

Useful files and their respective filepaths

Detailed below are the filenames and paths of the relevant files for running NAME III with the inclusion of decay chain modelling during atmospheric dispersion of radioactive material.

All modifications to NAME III to incorporate the ability to perform decay chain modelling have been added to the version 5_2a in:
file:/net/home/h03/apdg/NameIII/NameIIIDevVersions/Version5_2a/Code_NameIII
Once Dave Thomson has frozen a new version of NAME III (version5_3), the activity on a single particle should be compared against the HPA's progeny spreadsheet, for a handful of model runs, as detailed above.

A text file, 'Species.txt', detailing the species related parameters required in the Species block of a NAME III input file can be found in:
file:/net/home/h03/apdg/NameIII/NameIIIDevVersions/Version5_2a/Resources/Defns
Note that this file replaces the "old" Species.txt file which has been renamed, "Species01.txt". Species.txt has been tested by HPA (see above for details).

An example NAME III input file, 'Example_Decay_Chain_Modelling.txt', for estimating air concentrations whilst accounting for decay chain modelling can be found in:
file:/net/home/h03/apdg/NameIII/NameIIIDevVersions/Version5_2a/Runs

5. References

Fitzgerald, J J, Brownell, G L, and Mahoney, F J (1967). Mathematical Theory of Radiation Dosimetry. Gordon and Breach Science Publishers.

Jones, J A (1980). ESCLOUD: A Computer Program to Calculate the Air Concentration, Deposition Rate and External Dose Rate from a Continuous Discharge of Radioactive Material to Atmosphere. NRPB-R101.

Appendix A

The equation in the Radionuclide Data Chapter of the HPA's Models and Data Handbook is as detailed below (eqn 1a) but should be as detailed in eqn 1 on page 1. There should be brackets around the product of the branching ratio and the decay constant to indicate the product is from j=1 to i-1 of these two functions and not the product of the whole equation.

Furthermore the decay constant in this product term should be subscript j+1, rather subscript j.

$$A_i(t) = A^0 \prod_{j=1}^{i-1} f_{j,j+1} \lambda_j \sum_{j=1}^{i} \frac{e^{-\lambda_j t}}{\prod_{\substack{k=1 \\ k \neq j}}^{i} (\lambda_k - \lambda_j)} \qquad (1a)$$

Appendix B

NAME III has been tested to assess it's capability of handling a large number of radionuclides in the estimation of average air concentrations over a structured grid. A NAME III run has been set up which includes 76 radionuclides. This run can be found in: MaxSpecies. A number of temporary changes to the NAME III code had to be made to enable NAME III to perform this run. These changes are highlighted below:

i) In GlobalParameters.F90 'MaxColumnKeys' was increased from 72 to 140.
ii) In GlobalParameters.F90 'MaxColumns' was increased from 60 to 100.
iii) In GlobalParameters.F90 'MaxSpecieses' was increased from 36 to 80.

These changes alone did not result in a successful NAME III run, as a result of 'insufficient virtual memory'. The 'Max # Particles' parameter in the 'Sets of Dispersion Options:' block in the NAME III input file was subsequently reduced (from 5000000 to 2000000 particles), resulting in a successful NAME III run.