Data Assimilation and Dismantling Waste: A Methodology Customised for Radioactive Inventory Assessment Purposes - 8174

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ABSTRACT

Data assimilation is a versatile methodology which is widely used nowadays for estimating complex system variables. The estimation of a given quantity by data assimilation involves taking into account both the observational data and the underlying physical principles governing the system under observation.

Let us consider two assumptions, (1) the observational information consists of radioactivity measurements of numerous samples of graphite (or other material) from known points and (2) the physical principles are those of activation under neutron flux during nuclear power plant operation. With these two assumptions, a data-assimilation-like methodology is ready to be applied to radioactive inventory computation for dismantling graphite (or other material).

The paper is written for a planned graphite disposal project in France, for the purposes of its inventory forecasting. It introduces specific concepts for activation laws , presents the beginning of the application to BUGEY 1 plant pile and proposes an original method for overall error quantification.

INTRODUCTION OF THE EXAMPLE

Électricité de France (EDF), a radioactive waste producer, is responsible for managing waste from the dismantling of its permanently shut-down nuclear power plants. This dismantling process has been underway for a few years now and concerns 9 units.

Of these, 6 units are gas-cooled graphite power plants. The main constraint affecting their dismantling is the disposal of the graphite waste and the French act 2006-739 of 06/28/2006 confirmed that the goal is a provide a special repository exclusively for graphite.

EDF is required to assess the radioactive inventory for each of its graphite radionuclides (RN) and communicate the information to French governmental disposal agency (ANDRA), for the purposes of their safety studies in connection with the identification of suitable storage sites.

GENERAL CONCEPTS AND METHODS

By definition, data assimilation in a radioactive inventory assessment context is an estimation problem concerning the impurities which gave rise to the RN. The schemes for solving this problem have different backgrounds. They often relate to either estimation theory or control theory.
Several schemes are theoretically optimal, while others are approximate or suboptimal. Although optimal schemes are preferred because they should provide better estimates, suboptimal methods

are generally the ones in operational use. Most schemes are related in some fashion to least-squares criteria which have had great success.

Estimation Theory

Estimation theory encompasses theories used to estimate the state of a system by combining, usually with a stochastic approach, all available reliable knowledge of the system including measurements and theoretical models. The *a priori* hypotheses and melding or estimation criterion are crucial in the estimation process. At the heart of estimation theory is the scheme derived by Kalman in 1960, the *Kalman Filter*, which is a simplification of Bayesian estimation for the case of linear systems.

Control Theory

All control theory or variational assimilation approaches perform an overall time-space adjustment of the model solution to all observations and thus solve a smoothing problem. The goal is to minimize a cost function penalizing the time-space misfits between the measurement and calculation, with the constraints of the model equations and their parameters. The misfits are interpreted as part of the unknown controls of the system. The model parameters can also be optimized, as in estimation theory.

Estimation theory requires *a priori* stochastic assumptions for the model noise and data errors. Similarly, control theory results depend on the *a priori* control weights and penalties added to the cost function. The activation model is considered as a *strong constraint*. Strong constraints correspond to the choice of infinite weights for the model equations; the only free variables are the explaining impurities (initial conditions and/or model parameters). A rational choice for the weights and form of the cost function is important.

Choice adapted to radioactive characterization

In our case, the cost function is the sum of all data-model misfits at observation locations and we introduce constraints that are specific to the activation, neutron flux cartography and history being provided.

We will use a logarithm fitting and it is explained by the two following facts:

1. the fitting of RN activities should not depends on the date of their measurement,
2. the fitting of RN activities should not depends on the scale of magnitude of activity

Both explanations drive us to exclude the plain fitting of the difference and to use $\frac{C}{M} = \frac{Computation}{Measurement}$ to provide a comparison with no time or scaling effect. It is easy to give the demonstration that the only way to transform a difference into a ratio is the logarithm function. We will see later that the natural logarithm gives us directly the relative error quantification.

Let us consider a given RN with an i suffix which has been measured as M_{ij} in N_i locations with j suffix ($1 \leq j \leq N_i$) and for which the corresponding calculated value at the time of measurement

is C_{ij}. As data-model misfit for RN with an i suffix, we will call "bias", written B_i , the geometric mean of the N_i measurements divided by the geometric mean of the N_i calculations.

$$B_i = \frac{\left(\prod_j M_{ij}\right)^{\frac{1}{N_i}}}{\left(\prod_j C_{ij}\right)^{\frac{1}{N_i}}} \Leftrightarrow \ln B_i = \frac{1}{N_i}\sum_j \ln \frac{M_{ij}}{C_{ij}}$$

Constraint (avoiding any under-estimation): every B_i value must be less than 1.

Cost function: $\sum_i (\ln B_i)^2$

It is the sum of all the $(\ln B_i)^2$ in order to have all the "bias" values B_i as close as possible to 1.

We don't demonstrate here that the optimal solution exists and is single. We only present an intuitive solution considered as satisfactory for operational waste characterization purposes.

Writing for each suffix i, $B_i = 1$, we are faced with a system of simultaneous equations greater in number than the explaining initial elements activated by the neutron flux. Removing a set of equations corresponding to a sub-set of RN, we assume that we can transform this over-abundant system into a system with a single vector solution of initial element concentrations (the number of unknown concentrations being equal to the remaining equation corresponding to the remaining sub-set of RN).

We know that there is a finite number of ways to transform this over-abundant system as we have just described. So, we can choose the solution which gives the highest values of concentrations. For this solution, if we consider the equations (or radionuclides) removed, we will notice that their "bias" is less than 1 because the calculated values came from the highest initial concentration estimates. If we consider the other equations (or radionuclides), remember that we know that their "bias" is equal to 1.

ERROR CALCULATION

In the discussion which follows, the i suffix is omitted, in order to make the equations easier to read. This is permissible because we are only concerned with one RN.

Logarithmic error computation

$\mu_C = \frac{1}{N}\sum_j \ln C_j$ is the mean of the logarithms of the calculated values and $\mu_M = \frac{1}{N}\sum_j \ln M_j$, that for the measured values.

As the RN measurement error is ΔM_j (given for 95% level of confidence), we have, for the measurement error on μ_M : $\frac{1}{N}\sqrt{\sum_j \left[\frac{\Delta M_j}{M_j}\right]^2}$

Let's introduce $\mu = \frac{1}{N}\sum_j \ln\frac{C_j}{M_j}$. By definition, the "bias" B = $\exp(-\mu)$ and its value is fitted to be as close as possible to 1. So, $\mu = \frac{1}{N}\sum_j \ln\frac{C_j}{M_j}$ is as close as possible to 0.

We introduce the usual standard deviation estimator $\sigma^2 = \frac{1}{N-1}\sum_j \left[\ln\frac{C_j}{M_j} - \mu\right]^2$.

We obtain the usual fitting error on μ for 95% level of confidence (with "Student" t-distribution factor depending on the number N of samples) , $\varepsilon = t_N \cdot \sqrt{\frac{1}{N}\cdot\frac{1}{N-1}\sum_j \left[\ln\frac{C_j}{M_j} - \mu\right]^2}$.

As $\mu_C = \mu + \mu_M$, calculation error on μ_C is $\varepsilon_C = t_N \cdot \sqrt{\frac{\sigma^2}{N} + \frac{1}{t_\infty^2}\frac{1}{N^2}\sum_j \left[\frac{\Delta M_j}{M_j}\right]^2}$ with $t_\infty = 1.96$ for 95% normal level of confidence corresponding to measures.

Relative error computation

Taking the same level of confidence, the relative error of the activity is: $\frac{\Delta C}{C} = \exp(\varepsilon_C) - 1$.

If N is large enough, the t-distribution factor remains close to 2 and we have the approximation $\frac{\Delta C}{C} \cong \varepsilon_C \cong 2\sqrt{\frac{\sigma^2}{N} + \frac{1}{4N^2}\sum_j \left[\frac{\Delta M_j}{M_j}\right]^2}$.

The error propagation allows us to compute the impurity estimation error and to propagate it to each computed RN (even not measured RN) which is created starting from that impurity during activation process.

AN EXAMPLE : BUGEY 1 PILE

Overall result of inventory in progress

The fitted value of impurities having been computed, the calculation is ready to be performed. At the present time, calculations are in progress.

Fitted and unfitted impurities from last measurements

The unfitted impurities come from over-estimated values from literature sources because we have no measured RN values which could allow us to identify them.

Fractions of mass

Fitted	BA	3.63E-07	CR	5.80E-07
Fitted	CL	7.89E-08	CU	4.30E-06
Fitted	CO	2.33E-08	DY	5.00E-10
Fitted	CS	3.87E-09	GD	5.00E-09
Fitted	EU	2.80E-09	H	1.00E-06
Fitted	FE	1.96E-06	HF	2.50E-08
Fitted	LI	9.93E-09	HG	1.00E-09
Fitted	MO	9.47E-08	IN	6.70E-09
Fitted	N	2.30E-07	K	1.10E-05
Fitted	NB	1.19E-08	LA	2.10E-08
Fitted	NI	3.05E-06	MG	7.00E-07
Fitted	TH	1.60E-11	MN	2.40E-07
Fitted	U	8.85E-09	NA	1.10E-05
Fitted	ZN	1.18E-06	O	7.64E-03
	AG	1.00E-09	PD	1.00E-09
	AL	2.00E-06	RU	1.00E-07
	AS	1.70E-08	S	1.00E-05
	AU	1.18E-07	SB	8.95E-10
	B	1.00E-06	SC	1.20E-09
	BE	1.00E-07	SI	1.80E-05
	BI	5.00E-08	SM	3.00E-09
	BR	4.00E-08	SN	1.40E-07
	C	Sum = 1	TA	5.80E-09
	CA	5.00E-06	V	3.00E-06
	CD	1.00E-07	W	4.50E-08
	CE	6.00E-07	ZR	1.00E-07

Error computed from measurement and from misfit

When the bias values are very low, the model-based error values will create a high overestimation of the corresponding RN which will be corrected in the overall computation.

If several RN have the same leading impurity, the relative error is chosen as the biggest of the values for the various RNs.

Analysis	Measures >DL
PU239+PU240	14
NB_94F	5
PU241	4
EU155	17
FE_55	8
CO_60F	28
PU238+AM241	14
NI_63	28
EU154F	18
CM242	1
_H__3	28
NB_93M	4
PU238	4
CS137	16
CS134F	27
CM243+CM244	16
NI_59	4
CL_36	28
BA133F	19
_C_14	28

CONCLUSION

Radioactive waste management requires knowledge of the distribution and evolution in space and time of the characteristics of the neutron flux. The functions of space and time, or *state variables*, which characterize the state of the pile under observation, are classically designated as *fields*. The determination of the distribution or evolution of the state variables is a question of *state estimation* or *field estimation* in three dimensions.

A model to approximate the waste behavior consists of a set of coupled prognostic field equations for each state variable of interest. The fundamental properties of the system appear in the equations as parameters. In principle, the parameters of the system can be estimated directly from measurements. In practice, directly measuring the parameters of the system is difficult because of sampling, technical and resource requirements. Data assimilation however provides a powerful methodology for parameter estimation.

Today and in the foreseeable future, data acquisition from the NPP to be dismantled for the making of field estimates by direct measurements, on a substantial and sustained basis, and for state and parameter variables, sampling level and spatial domains of interest, is prohibitively difficult and costly. However, data acquisition for field and parameter estimates by data assimilation is feasible, even though substantial resources must still be applied to obtain enough observations.

REFERENCE

"Characterization of graphite sleeves from Bugey 1 EDF plant for permanent disposal – Measurement and calculation of scaling factors for difficult-to-measure nuclides". Presentation by Bernard Poncet to the congress at the WM’03 Conference in February 2003 at Tucson, Arizona.