Material Flow Modelling for Environmental Exposure Assessment – A Critical Review of Four Approaches Using the Comparative Implementation of an Idealized Example

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Abstract

Newly developed materials such as engineered nanomaterials are produced in increasing amounts and applied in a growing number of products. Once released to the environment, they can pose a hazard to ecosystems and human health. To assess potential risks, the exposure of the material to humans and the environment has to be determined. For many materials such as engineered nanomaterials, a quantitative measurement of environmental concentrations is not feasible. Material flow models can be used to determine these concentrations indirectly by predicting material flows in the environment. Several modelling approaches can be applied to represent existing knowledge about the flows of materials into and between environmental media or compartments and to consider the uncertainty and variability of the input parameters. In this study we evaluate four existing approaches with regard to their capabilities for indirect exposure assessment, focusing on their ability to treat uncertainty. We first explain how we preselected the four most promising modelling approaches: material flow analysis, system dynamics, material flow networks, and probabilistic material flow analysis. We then define a set of evaluation criteria based on the requirements of environmental exposure assessment and develop a simplified example system that is designed to test these criteria. Based on the comparative modelling and implementation of the example system, we discuss the capabilities and limitations of the approaches and indicate what is missing for a reliable environmental exposure prediction using material flow modelling.

1. Introduction

A main issue in the risk assessment of new substances such as engineered nanomaterials (ENM) is to determine the exposure of humans and ecosystems to the substances. An understanding of the environmental fate of a chemical product leads to knowledge about actual environmental concentrations, exposures and potential risks (MacLeod et al. 2010). For many pollutants, a direct measurement of environmental concentrations is not feasible and so the environmental fate cannot be determined directly. Material flow modeling holds the opportunity for an indirect assessment (Klaine et al. 2012). Instead of a direct assessment of environmental concentrations, material transfers between environmental compartments are regarded. This enables an estimation of material accumulations in the respective compartment and so the prediction of environmental concentrations based on standard sizes for environmental compartments (ECHA 2010).

Even though material flow modeling provides means for environmental exposure assessments, the informative value is usually limited by incomplete knowledge about some system parameters. This uncertainty results from variances of the actual flows and the partial or total lack of knowledge about their true behavior. (Paté-Cornell 1996; Refsgaard et al. 2007) To obtain reliable results, it is essential to consider

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these uncertainties, represent them explicitly in the flow model and process them through the model, using an adequate simulation method.

Another important issue is that temporal delays should be adequately represented in the model. The transfer of a pollutant from the point where it is released to the environment to the area where it finally accumulates is usually not immediate. Often, a material is bound for a long time in a compartment before it is further transferred through the system and finally accumulates. To investigate such system behavior appropriately, a modeling approach must be able to represent a time dynamic behavior and delayed material release from local stocks.

A good example for the use of material flow modeling for an environmental exposure assessment is found at Mueller/Nowack (2008). These authors used a methodology related to material flow analysis (MFA) to assess environmental concentrations of several ENM. Gottschalk et al. (2010) extended the classical MFA approach to probabilistic material flow analysis (PFMA) by the introduction of Bayesian statistics to represent and process uncertain knowledge about system parameters. Besides MFA and PMFA, System Dynamics (SD) and Material Flow Networks (MFN) appear to be suitable for a comprehensive exposure assessment using material flow modeling.

In this study we provide a detailed look on several most promising material flow modeling approaches. In particular, we will highlight the capabilities and limitations of each modeling approach to represent uncertain knowledge about system parameters and time dependent release behavior from local stocks.

For our investigation we first preselected the most promising modeling approaches. Then, we developed an idealized example system that comprises the most crucial aspects of exposure assessment modeling. Finally, we implemented the example system using each of the modeling approaches and based on that evaluated the specific capabilities and limitations of the approaches.

1.1 Selection of material flow modeling approaches

The selection of the approaches to study was based on two principles: The expected capability of an approach to represent a system of material flows to predict environmental concentrations and second, the coverage of a large variety underlying modeling and simulation mechanisms. That way, four approaches were chosen for further examination.

System Dynamics was developed to represent dynamic systems as sets of material stocks and flows, interconnected by information flows. The approach allows a quasi-continuous simulation by numerical integration with Euler- or Runge-Kutta-methods (Morrison 1991). The original approach was developed by Forrester (1961). There are several software tools available to support System Dynamics modeling and simulation such as DYNAMO compilers and graphical modeling tools such as Stella. We used Vensim[3] for the exemplary model.

Material Flow Networks are based on Petri Nets. They are mainly used to account material flows in operational processes, in particular Life Cycle Assessment. Based on the production of goods the dependent substance and energy flows are determined. The approach was developed by Möller (2000) and extended by Wohlgemuth (2005) and is supported by the graphical modeling tool Umberto[4]. It was used for the implementation of the example.

Material Flow Analysis (MFA) is an approach to model material flows as period-oriented transfer of a material between system entities (Brunner/Rechberger 2004). The main aspects of MFA are included in

[3] Vensim PLE Plus, Computer Program, V.6.1c; http://vensim.com/, accessed: March 22 2013.

[4] Umberto, Computer program; http://www.umberto.de/en/, accessed: March 21, 2013.

the software tool STAN[5], with which we performed the exemplary implementation. The approach was developed by Baccini/Brunner (1991).

Probabilistic Material Flow Analysis (PMFA) is a modeling approach that was specifically designed for environmental exposure modeling (Gottschalk et al. 2010). It extends the classical MFA approach by Bayesian statistics. It describes a stable state in a system of dependent material flows under substantial uncertainties. There is no software program directly implementing the approach. Instead the language R[6] for statistical computing and graphics and some packages that extend it provide the modeling methods.

1.2 Developing evaluation criteria

The criteria to evaluate the different modelling approaches are chosen to reflect crucial requirements of material flow modelling to predict environmental concentrations. Of particular interest are the way how uncertain knowledge is represented and processed and how complex and time dependent release from stock is modelled.

The first evaluation criterion regards the capability to represent and process incomplete knowledge. Usually, the existing knowledge about the actual behavior of a specific pollutant released to the environment is not complete. There is uncertainty about its release, about the flow rates between the compartments of the system, and about the accumulation and degradation rates of the material. The Bayesian concept of probability enables the full representation of uncertain knowledge as different assumptions with different degrees of credibility. (Cullen/Frey 1999) Thus, an adequate system representation in the model and meaningful simulation results should display uncertain information as Bayesian probability distribution. Depending on the knowledge about the process that needs to be examined and the way the information was gathered, usually they are available as theoretical or empirical distribution functions. We evaluated the modeling approaches including both variants.

The second evaluation criterion regards stocks and their ability to represent a dynamic system behavior over time. Usually, the emergence of environmental contamination from a released pollutant is not an immediate process. For instance, a pollutant can be bound in a landfill for several years before it actually reaches ground water. The approach should be able to deal with such a delayed release. The modeler should be able to consider a rate of the total amount stored, a time delay, or external triggers as conceivable factors for a release from a local stock.

Additional attention is paid on the modeling and simulation process, in particular how they are performed with each approach and what support and guidance the respective software tools provide.

2. Definition of the example system

The aim of this study is to evaluate the modelling approaches with regard to the previously defined criteria. Therefore, an idealized example system was developed for comparative implementation. The properties of the system to be modelled are derived from the evaluation criteria. Thus, the fulfilment of a criterion can be assessed by the adequacy with which the corresponding aspect of the system could be implemented.

Beside the requirement to comprise the characteristic aspects of flows of pollutants in the environment, the system should be kept simple in size to avoid unnecessary modelling effort and a concealing of the actual modelling and simulation principles of the approach. The basic system consists of several compart-

[5] STAN, subSTance flow ANalysis, computer program; V. 2.5.1072; http://www.stan2web.net/, accessed: March 21, 2013.
[6] R, Programming language, http://www.r-project.org/, accessed: April 7, 2013.

ments, a source releasing material, relative flows between the compartments, material accumulation, and a stock with a time delayed release of material (Figure 1).

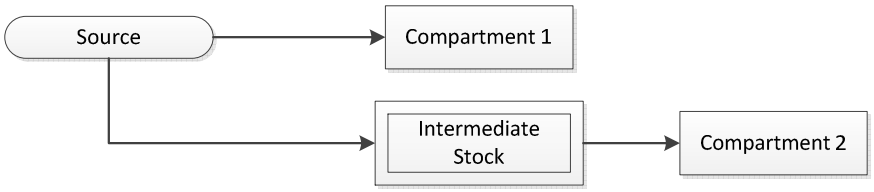

Figure 1
Basic Structure of the Example System

For the idealized example we chose the following specific parameterisation:

- **Material Release:** It was assumed that the knowledge about the actual material release is incomplete. We made the assumption of an empirical distribution function. A periodic material release from source of 500 t/y is assumed with the likelihood of 0.2, 100 t/y with a likelihood of 0.5 and 1500 t/y with 0.3. The periodic release is constant and should not be varied between the periods.
- **Flows:** The knowledge about the allocation of the material released from source leads to a normal distribution. Each period 0.6 of the total material released are assumed to be transferred to the "Intermediate stock" with a standard deviation of 0.2. This value is assumed to have a variant behaviour over time. The remaining part of the material is transferred to "Compartment 1".
- **Delayed release from stock:** All material transferred to the "Intermediate Stock" remains there for 2 years. Afterwards the material is transferred to "Compartment 2" with a rate of 0.5 per year.

The investigation period of the system is 10 years.

3. Implementing the four models of the example system

This section describes the implementation of the models using each of the four modelling approaches. Special attention is paid to the modelling and simulation process and the general procedure, how and to what extent the specific aspects could be implemented and general particularities observed.

3.1 System Dynamics using Vensim

The System Dynamics approach assumed continuous (flow) processes. The underlying mathematical model represents these processes as a set of differential equations. To simulate the model the equation system has to be solved for the time instants of interest. In general differential equations are not analytically solvable, which is the reason why numerical methods are applied. For our implementation we chose Runge-Kutta 4 with a basic calculation time step of 0.125 of the basic time unit 1 year.

In the first step of the implementation of the example system, we had to define the static model structure. All model components had to be placed and connected on a canvas by drag and drop. First the three compartments of the example system are placed as `Levels` (stocks). They can change their values constantly over time. Flows among the stocks or between stocks and the system's environment are displayed as uni- or bidirectional `Rate` pipes. Factors that influence the behavior of material flow amounts and rates were modeled as `Auxiliaries`. They represent the information flow of the system. Auxiliaries do not keep information between the calculation steps. Dependencies in the model are visually defined as arrows between the model variables.

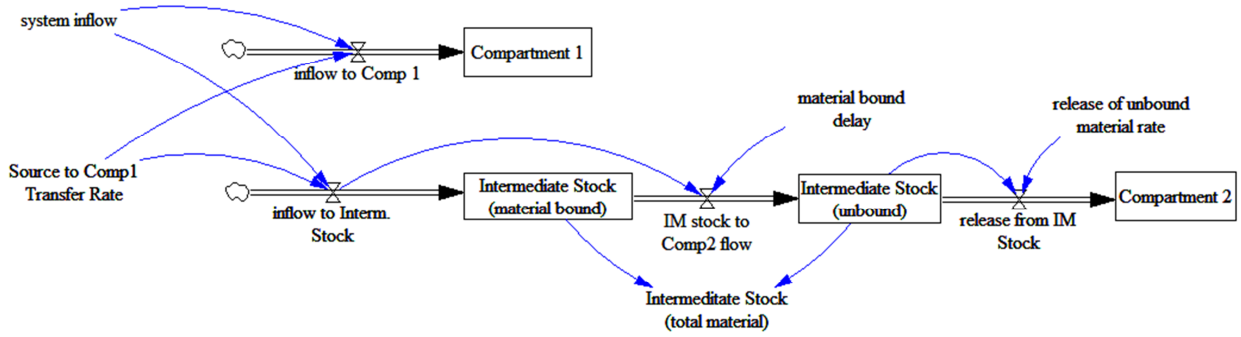

Figure 2
Structure of the Vensim Model

After the model was composed of its static elements the actual behavior of the components was applied to the model components as mathematical functions. The `Levels` were set with 0 as initial value, because our example system starts with empty stocks. The change rates of stocked material in the `Levels` are described as the sum of the inflows rates stock minus the sum of the outflow rates. The values of flow `Rates` and `Auxiliaries` are displayed by a single value that is either constant or a function of values of other variables that is determined in each calculation step. During simulation, the calculation steps are iterated, each calculating the periodic flows and then updating the system state.

Vensim provides an infrastructure to perform Monte-Carlo simulation to examine model behavior under uncertainty. It allows the modeler to assign probability distributions to model parameters to represent the uncertainty about its true value and evaluate the model behavior over a large set of simulation runs. In our example we varied the `system inflow` parameter between the simulation runs. To express the empirical probability distribution we first generated a uniform distribution and had to map it in a second step to the specific distribution using a `Lookup` function. For the transfer coefficient that splits the total system inflow into a flow to `Compartment 1` and `Intermediate Stock` the parameter is varied between all periods. The intermediate stock compartment was implemented by splitting it up into two `Level` variables. The first one represents the stocked material that is bound for the first two periods. After these periods the material is transferred to the second `Level` variable. The flow rate is defined using the inflow to the previous compartment and a `fixed delay` function. The actual material stored in the intermediate

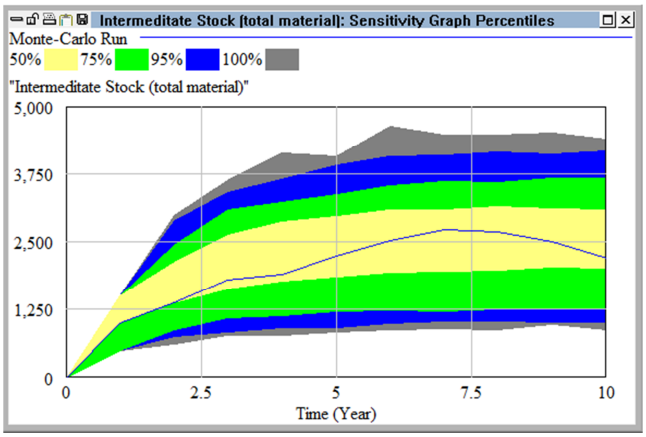

Figure 3
Evaluation of material in Intermediate Stock over time

stock compartment is displayed by an `Auxiliary` that sums up the material from both stocks.

Simulation results can be displayed as tables and diagrams for all model components. Vensim provides functions for data import and export.

3.2 Material Flow Networks using Umberto

In Material Flow Networks (MFN) the static structure of a model is a Petri net (for an introduction to Petri nets see Peterson 1981). It consists of places that hold materials, and transitions that connect two or more places. In MFN, the transitions define material transfers as a transformation rule of inputs to outputs. In particular, they specify all the material types and relative amounts.

To implement the example system we first defined the total simulation time period as 10 single periods of one calendar year. Then we modeled `Source`, `Compartment 1`, `Compartment 2` and the `Intermediate Stock` as places. In Umberto, places are suitable to account the amounts of stored material and their changes over the whole simulated period. All flows between the compartments were implemented as transitions. In accordance with the Petri-net notation the `Source` compartment was designed as "source" place and `Compartment 1` and `2` as sinks. Flows from one place were aggregated to one transition that holds the actual algorithm of the material transfer. In the case of Transition 1 (Figure 3) it comprises the two transfer coefficients of the flows to the two subsequent compartments.

Umberto provides a basic support for Monte-Carlo simulation with the most commonly used mathematical distribution functions. So the split flow from the `source` to `compartment 1` and the `intermediate stock` could in general be assigned with a normal distribution for the transfer coefficients. However, the implementation was not really straightforward because probability distributions cannot be directly assigned to a variable in a transition but have to be declared globally as "net parameter" and then introduced in a second step into the intended model parameter. The implementation of the periodic material release as empirical distribution was not supported by Umberto. Instead a constant value was used. The application of Monte-Carlo simulation in Umberto is limited to a single period. The variance of model parameters is processed only for the current period. To the following period only the average value of the sample is transferred.

The time delayed release of material from the intermediate stock compartment could not be implemented as an internal model logic. Umberto was not built to model time dynamic behavior. To implement the model nonetheless, the release from `Intermediate Stock` was calculated manually and the flow between `Intermediate stock` and `Compartment 2` parameterized accordingly.

The underlying model of a material flow network is a linear equation system which is determined by the transfer rules of the model. Using Monte-Carlo simulation it becomes a probabilistic model. Umberto enables a representation of results as Sankey diagrams for the flows of one period. Results of Monte-Carlo simulation can also be displayed in the form of bar charts.

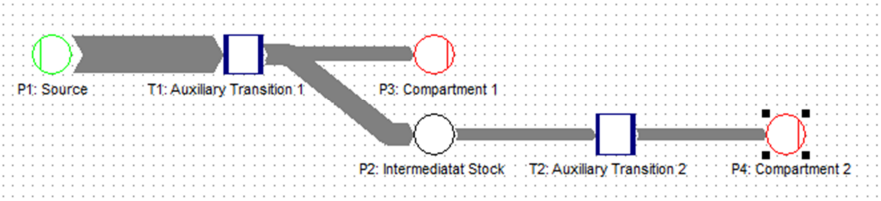

Figure 3
Implementation of the Example System using Umberto

3.3 Material Flow Analysis using STAN

In MFA a system of dependent flows is represented by a set of processes and the flows of substances and goods between them. System changes over more than one time period are represented as changes in stocked material which is accounted for over the entire simulation time.

In the first step of the modelling process we had to determine some global settings. We chose a period length of one year and a total number of 10 periods. A year was also chosen as the temporal reference unit and the finest granularity. In the second step the model structure was defined using a drag and drop interface. The compartments of the example systems were modeled as processes. `Compartment 1` and `2` and the `Immediate Stock` were modeled as stocks. Furthermore, the material flows were defined. There are flows between processes and in- and out-flows over the system border.

Finally, the model was parameterized, by assigning specific values for flows and dependencies. We assigned the inflow to the model as `Flow 1`. The transfer coefficients that split the total inflow to a flow to `Compartment 1` and `Intermediate Stock` are assigned in the `Production Process`. As Umberto, STAN does not provide means to represent the complex and time delayed release behaviour from the `Intermediate Stock`. Therefore the time dependent release from the "`Intermediate Stock`" of the example system could not be defined as part of the model. The amounts were calculated manually and used for parameterizing `Flow 4`.

The underlying mathematical model is a system of linear equations. With the given parameterisation the equation system is determined. STAN can calculate all remaining model variables as dependent values. In this approach, the representation and processing of uncertainty in terms of Bayesian statistics and Monte-Carlo simulation in not feasible. Instead STAN enables the handling of uncertainty as standard deviation of a normal distribution. It supports the concepts of error propagation and equalization calculus. In the example implementation, uncertainty was modeled using standard uncertainty.

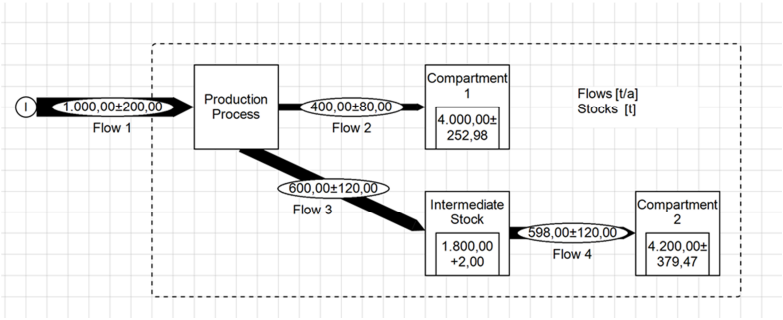

Figure 4
Implementation of the Example System using Stan

3.4 Probabilistic Material Flow Analysis in R

Probabilistic material flow analysis (PMFA) extends the basic approach of material flow analysis. In PMFA incomplete knowledge is expressed as a set of assumptions with assigned Bayesian probabilities. It allows calculating equilibrium for a set of interacting flows and representing existing uncertainties about the actual values of the original system using Monte-Carlo simulation.

The example implementations in system dynamics, material flow networks an MFA were each performed using a specific software tool. Currently, there is no software tool that specifically supports the design and use of PMFA models. The model it is instead implemented in the programming language R.

In the first step of the modeling process all dependencies between system variables were transferred into a mathematical equation system (Figure 5), representing the static structure of the system. In a follow-

ing step, uncertain knowledge is displayed in the model. Bayesian probability distributions are applied considering all knowledge about the system dimensions. In this case we created an empirical probability distribution for the material release from "`Source`" that regards all assumptions made and their likelihood to be true. The transfer coefficients for the flow between "`Source`" and "`Compartment 1`" and "`Source`" to "`Intermediate Stock`" are displayed by a normal distribution as given in the example system. PMFA is not intended to model time dynamic behavior. For this reason the "Intermediate Stock" was not included in the implementation. Instead, the inflowing material is directly transferred to "`Compartment 2`".

$$\delta Comp\ 1 = totalInput * TC_{InComp1}$$

$$\delta Comp\ 2 = totalInput * (1 - TC_{InComp1})$$

Figure 5
Structure of the Example System as Mathematical Equations

The model behavior is produced using Monte-Carlo simulation. Therefore, a large set of simulation runs is performed and evaluated with statistical methods. In each simulation run the model parameters are set with a random value coming from the assigned probability distribution. Then the equation system of the model is solved to determine the values of all variables.

The use of the programming language R instead of a specialized tool demands a larger previous knowledge from the modeler about the modeling approach and the language R. Also, there is no specific modeling guidance by the tool and no predefined visualization. However, as a language for statistic calculation and visualization, R provides a large range of possibilities to visualize simulation results. Figure 6, for instance, displays a plot the simulation results as probability densities, mean values, and quantiles for the stocked material of Compartment 1 and 2.

Furthermore, aspects that are not part of the pure PFMA approach can be represented making use of the general features of the language. This also enables an extension of the existing PMFA approach to system aspects that are not included yet such as the change of the system state over time.

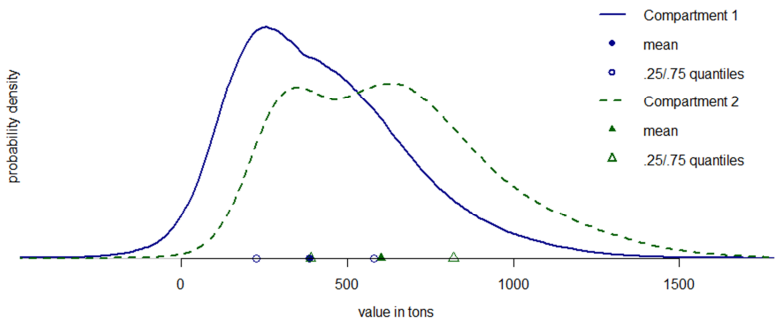

Figure 6
Modeling in PMFA (using R), probability densities of stocked material amounts after one period

4. Discussion

The aim of this study was to evaluate the capabilities of existing material flow modelling approaches for assessing environmental concentrations. Therefore, an idealized example system was developed that includes the most crucial issues of environmental flow processes. This example system was implemented using several material flow modelling approaches. The evaluation of each of the implementations of the example system reveals a ranking of the capabilities of each single approach to perform the entire model-

ling process (see Table 1). While classical MFA and material flow networks were not able to model most of the system's aspects, system dynamics and PMFA showed greater capabilities. Altogether, Vensim could implement the example most adequately, closely followed by PMFA.

Criterion	Material Flow Analysis (STAN)	Material Flow Networks (Umberto)	System Dynamics (Vensim)	Probabilistic Material Flow Analysis (R)
Empirical distribution as model input	○○○	○○○	●●○	●●●
Normal distribution for transfer coefficient	●○○	●○○	●●●	●●●
Time delayed release from stock	●○○	●○○	●●●	●○○

●●● Modelling possible and well supported
●●○ Modelling possible, but not supported; not in the focus of the modelling approach
●○○ Modelling partly possible or only with much effort; not in line with the modelling approach
○○○ Modelling not possible

Table1
Evaluation of the modelling approaches

The approaches differ considerably regarding the way uncertain knowledge is represented and processed. STAN handles uncertainty as standard deviation from a mean value. Thus, uncertainty can be processed by error propagation and equalization calculus. The other approaches represent uncertain knowledge using Bayesian statistics and Monte-Carlo simulation. That way, they enable a more differentiated treatment. Monte-Carlo simulation in Umberto is constrained by some general limitations. It can only be applied for one time period. The sample that represents a system variable is transferred to the next period as the single average value. Furthermore, the probability distributions have to be defined in a rather complicated way as global 'net parameters'. In a second step they have to be assigned to the specific model variable. Empirical probability distributions cannot be represented in Umberto.

In contrast to that, Vensim and PFMA are able to represent and process all aspects of uncertainty of the example model. However, in Vensim some small limitations occur. At the implementation of the empirical probability distribution for the material release an auxiliary modelling step had to be performed. To emulate the probability function, a uniform distribution had to be mapped onto a lookup function.

The representation of a delayed material release from a local stock could be best implemented using Vensim. Since System Dynamics was developed to display dynamic and time dependent system behaviours, this system aspect fits to the scope of the approach. In STAN and Umberto the system behaviour is represented as the change of stocked material. The level of a stock at the end of a time period is transferred to the next period as initial value. The actual model logic is explicitly parameterized for each period, which makes it impossible to deal with a more specific time dependent behaviour. PMFA does not include a method to represent system changes over time. However its implementation using R enables to model a specific system behaviour that exceeds the current specifications of the approach.

As a general remark, none of the four approaches was able to represent and simulate the entire example model satisfactorily. In the modelling and simulation process of a real-world case, this will presumably have even stronger consequences on the reliability and significance of the obtained results, because these systems are usually larger in size and complexity than our example system.

Therefore, a new approach that combines the advantages of the modelling approaches investigated could be of great value. For the development of this approach it seems reasonable to take PMFA as a start-

ing point and extend it by the stock and flow methodology of System Dynamics. Since PMFA is implemented using the language R it provides a large range of probability distributions to represent uncertainty. To base the new approach on PFMA instead of System Dynamics has the advantage to avoid the inherent discretisation error and additional computational effort of continuous simulation in System Dynamics. Furthermore, the model implementation using the R language leads to a straightforward adjustment and extension of the existing modelling approach.

5. Acknowledgements

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