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2  
3 **Predicting accidental release of engineered nanomaterials to the**  
4 **environment**

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25  
26 **Abstract:** Challenges in distinguishing natural and engineered nanomaterials (ENM) and the  
27 lack of historical records on ENM accidents have hampered estimating the accidental release  
28 and associated environmental impacts of ENM. Building on knowledge from the nuclear  
29 power industry, we provide an assessment of the likelihood of accidental release rates of  
30 ENM within the next 10 and 30 years. We evaluate risk predictive methodology and compare  
31 the results with empirical evidence, which enables us to propose modelling approaches to  
32 estimate accidental release risk probabilities. Results from two independent modelling  
33 approaches based on either assigning 0.5% of reported accidents to ENM-releasing accidents  
34 (M1) or based on an evaluation of expert opinions (M2), correlate well and predict severe  
35 accidental release of 7% (M1) in the next 10 years and of 10% and 20% for M2 and M1,  
36 respectively, in the next 30 years. We discuss the relevance of these results in a regulatory  
37 context.

38

39

40 **Main:**

41 Particles of all kinds of elemental compositions and sizes between roughly 1 nm and 1  $\mu\text{m}$ ,  
42 generally referred to as colloids, have been present in large quantities throughout terrestrial  
43 history <sup>1</sup>. Within the past two decades concerns have been raised regarding the health and  
44 environmental impacts of engineered nanomaterials (ENMs), which, following the EU's  
45 recommended definition, are manufactured materials where more than 50% of the particles  
46 are between 1 and 100 nm. <sup>2</sup> The overlapping size ranges between natural nanomaterials and  
47 ENMs challenge traditional risk assessment techniques, especially for ENMs having natural  
48 (geogenic) counterparts. For example, experiments with paints for exterior use revealed that  
49 nanoscale titanium dioxide ( $\text{TiO}_2$ ) particles were washed off from facades and reached the  
50 (aquatic) environment. <sup>3</sup> Recent analytical advances in the field of mass spectroscopy based  
51 single particle analysis either used as stand a alone technique <sup>4-7</sup> or in combination with  
52 characteristic total elemental ratios of bulk samples <sup>8,9</sup> allowed estimating the share of  
53 selected ENMs in complex matrices. Although these developments are very promising, they  
54 only allow detecting a limited selection of ENMs, as for example carbon based ENMs cannot  
55 be detected using these methods. Thus, distinguishing between engineered and natural  
56 nanomaterials in the environment is still challenging, <sup>10-12</sup> which hampers connecting the  
57 ENM release from production processes and product uses to exposure concentrations for  
58 either the public or the environment. The traditional strategy of detecting and quantifying  
59 contaminants in the environment to validate release models and inform on potential  
60 environmental relevance and significance of laboratory-based ecotoxicity values has,  
61 therefore, not been fruitful.

62 In response, significant effort has been directed towards estimating the future  
63 quantities of ENMs that might enter the environment to assess the risks associated with the  
64 increasing use of ENM.<sup>13</sup> Assessments of potential ENM releases during production or  
65 product use were based on probabilistic reasoning <sup>14-20</sup> and were expanded to include  
66 unintended ENM releases during ENM product use over the course of their life cycles. Yet,  
67 there are limited opportunities for model evaluation and validation considering the  
68 fragmentary knowledge of production volumes and the remaining analytical challenges  
69 surrounding field studies. <sup>18</sup> There is abundant literature on risk assessments on ENM that can  
70 be sourced from multiple EU funded projects. <sup>21-24</sup> However, none of these projects,  
71 addressed the accidental release of ENM and the associated risks beyond the research and  
72 development stage. Various papers consider the governance of ENM, but an accidental  
73 release of these ENM is outside the scope of such governance, e.g., <sup>25</sup>. This paper, therefore,  
74 stands as a unique attempt to provide a basis for assessing the likelihood the accidental release  
75 of ENM to the environment. The output from this study serves as a basis for reviewing risk  
76 management methodologies to ensure their applicability for the accidental release of ENM.

77 Accidental release will lead to spatially and temporally elevated ENM concentrations, making  
78 it more pertinent for assessing potential risks to the public and the environment as bystanders.  
79 Read, Kass et al., <sup>26</sup> recommend: "*identifying proactive measures to prepare for the*  
80 *occurrence of a negative event in order to ensure that the broader market for all products*  
81 *utilising a new technology are not unduly affected by an isolated incident*". Therefore,  
82 investigating the probability of accidental ENM release is a key component of horizon  
83 scanning exposure and risk assessment efforts.

84 The goal of this study was to make an assessment of the likelihood of ENM released  
85 as a consequence of accidents occurring during ENM-product fabrication, transport and end-  
86 of-life processes. Earlier investigations of such accidents were primarily qualitative  
87 discussions of ENM production equipment failure <sup>27</sup> and an accompanying commentary on  
88 the need for regulation to avoid accidents, <sup>28</sup> or were examinations of workplace exposures to  
89 incidental releases of nanoaerosols.<sup>29,30</sup> In making this first assessment, we provide

90 methodologies that may be readily applied to groups of chemicals for which conventional risk  
91 assessments of accidental release are limited due to lack of knowledge on accident rates,  
92 extents and material hazard.

93

#### 94 **Adjusting modeling approaches from the nuclear power industry.**

95 The developed approach relies on stochastically varying and combining uncertain or  
96 fragmentary (as well as assumptive) past accident frequency data to compute a multitude of  
97 probability generating functions (PGFs) for future accident probabilities. The generated PGFs  
98 serve as a basis for organizing and running the developed stochastic-probabilistic  
99 computations (see the method section below and supplementary S3 for more detailed  
100 information). To assess the frequency of accidental releases of ENMs, we searched for  
101 information and analogies from neighboring fields. In the chemical industry, the estimation of  
102 the accident probability is usually done based on process risk analysis at the plant scale<sup>31,32</sup>  
103 and involves the determination of the likelihood of occurrence of each of the undesired  
104 situations defined in the hazard identification step. This can be done typically by extrapolating  
105 historical failure frequency data. However, these data are missing for ENM sectors and do not  
106 encompass accidents occurring at customer plants, where the ENM is an ingredient, or during  
107 the transportation of ENMs to customer sites.

108 In that context, we decided to evaluate alternative approaches developed in the nuclear  
109 power sector for which an extensive probabilistic risk analysis (PRA) methodology has been  
110 developed.<sup>33-35</sup> (see also supplementary S3 for a review of the method).

111 There are differences between the nuclear power and the ENM sectors (see also  
112 supplementary table S5). The nuclear power industry can focus on a limited spectrum of plant  
113 sites and possible events, has a well-established definition of release of radioactive material  
114 and benefits from the technological similarity among nuclear power plants which share a  
115 rather uniform set of operational practices. The ENM sector lacks these information affecting  
116 the development of a predictive risk analysis method for accidental release of ENMs. We,  
117 therefore, reviewed experiences and challenges regarding model uncertainty and predictability  
118 of (major) accidents in the nuclear power plant sector to identify adjustments required for  
119 bridging the approaches to the ENM sector. The major adjustment is that mechanistic-  
120 technical considerations of accident-initiating and other events (and their probabilities) had to  
121 be discarded. We instead used a theoretical approach to derive probabilities of accidental  
122 release from a more superordinate point of view that projects such release frequencies  
123 (probabilities) from the past into the future without considering what went or may go wrong  
124 from a mechanistic point of view.

125 Selected combinatory and probability calculations for predicting ENM accident  
126 frequency and likelihoods emerged from our comparative analysis and were embedded into a  
127 stochastic Monte Carlo framework designed to cover as many accident frequency/probability  
128 scenarios as possible. Such a spread of model input scenarios also addresses unspecified  
129 differences between past and future ENM market and technology developments, including  
130 safety progress of ENM producing and handling technologies.

131 We collected data from the chemical industry on accidents involving chemical  
132 compounds, as well as the frequency of such accidents. Special attention was given to the  
133 Analysis, Research, and Information on Accidents (ARIA) database<sup>36</sup> of the French Ministry  
134 of the Environment, which is a compilation of (worldwide) accidents that led to  
135 environmental releases of chemicals. We searched this database to identify accidents  
136 involving chemical compounds which may have resulted in the accidental release of ENM.  
137 The actual presence of the substance in the nanoform is not specified explicitly in the

138 database, which introduces an uncertainty when interpreting the data. The proposed modeling  
139 approach based on stochastic analysis is designed to address this uncertainty.

140

### 141 **Modelling approaches using fixed shares and expert opinions**

142 We considered major accidents with high release volumes but low frequencies of occurrence  
143 as well as smaller incidents with low release volumes but higher frequencies of occurrence.  
144 This resulted in two data sets which were used for two different modelling approaches (model  
145 1 (M1) and model 2 (M2)), but both including global data on accident frequencies (see  
146 comments on data limitations in the supplementary S2). Both approaches aimed at assessing  
147 the probability of the accidental release of ENM, whereas the fate of the released ENMs was  
148 not addressed. In M1 we used the ARIA accidents collection to derive generic accident  
149 frequencies for varying release volumes of chemicals in general (see supplementary Figure S1  
150 and Tables S1-S3). In principle, ARIA distinguishes between non-transport accidents,  
151 including all on-site plant events, and accidental release off-site, during transport on roads or  
152 in pipelines (see supplementary Table S1 and S2). We combined these two categories to one  
153 general accident category. We assumed that past and future accidental release rates of ENM  
154 correspond to the market share of ENM currently amounting to 0.5% of the total chemical  
155 market (see supplementary section S1.1).<sup>37</sup> Consequently, on average 0.5% of accidents in the  
156 ARIA database were associated with the release of ENMs. An uncertainty range of 50% was  
157 considered on each side of all annual accident frequencies derived from the empirical data.  
158 We constructed empirical (hypothetical) accident frequency data as input for M1, using a  
159 Monte Carlo approach to address the uncertainty of the future market share of ENM and the  
160 relative frequencies of various release levels. The amount of ENM released during accidents  
161 increases by a log<sub>10</sub> unit from one level to the next. In M2, the ARIA data were evaluated and  
162 out of 1000 chemical accidents, a dozen accidents possibly resulting in the release of ENM  
163 were identified. The reports of those accidents were classified by a panel of 11 experts from  
164 academia, with a background in chemistry, according to the likelihood of ENM release. Four  
165 categories were defined, ranging from very unlikely release of ENMs (category 1) to very  
166 likely release of ENMs (category 4). The 12 accidents were scaled to 192 out of 16,000 events  
167 reported in total. Based on the expert judgements of the probability of ENM release in these  
168 accidents, the frequencies of past accidents associated with such release were estimated and  
169 used as input data for M2 (see supplementary Figure S1 and Tables S1-S4). Such input was  
170 fed into individual, expert-based predictive Monte Carlo simulations. These calculations for  
171 M2 were conducted for the lowest release level (level 1); the M2 results of higher release  
172 levels were derived by extrapolating the level 1 accident probabilities to lower accident  
173 numbers and frequencies of higher level (more severe) accidents. The supplementary S1 gives  
174 a full description of data sets and methods of M1 and M2.

175 Based on the aforementioned dual evaluation of empirical data following risk analyses  
176 concepts from nuclear power and chemical industry, we assessed for the first time the  
177 probability of accidents associated with the release of ENM in the near (10 years) and mid-  
178 term (30 years) future considering different accident severity levels.

179

### 180 **High likelihood of nano accidents within the next 10 years**

181 The results from the two approaches (M1, M2, supplementary Figure S1) predicting the  
182 occurrence of minor to major accidental release of ENM were in excellent agreement.  
183 Predicted probabilities resulting from the models M1 and M2 are shown in Figures 1-4 and  
184 Table 1. These predictions revealed a wide range of accident numbers (0-200 and more) and  
185 associated probability of occurrence and – for pessimistic scenarios – suggested a 100%

186 likelihood for level 1 ENM accidents within the next 10 or 30 years (Table 1). We note that  
187 the ranges of uncertainty of the predicted accident probabilities often comprise a factor larger  
188 than 2, when comparing the extremes of the model output (Figures 1 and 2, Table 1).  
189 Furthermore, the central values, as given in Figures 3 and 4, only reflect probable estimations,  
190 which still may deviate from the real (precisely known) values.

191 For limited ENM release rates of level 1 (see supplementary Table S2 for a description  
192 of the levels), the results from the 10-year simulation from M1, predict 21–30 accidents with  
193 a very high probability (96%, Figure 3). This translates roughly into 2 - 3 ENM accidents per  
194 year. In the 30-year forecast, the predicted range of 61–80 accidents has the highest  
195 probability (80%). This is in line with the 10 year predictions and would also lead to  
196 (roughly) 2 - 3 events annually. The results of M2—although they are based in large parts on  
197 another modelling approach and an entirely different model’s input data —agree well with the  
198 results of M1. For level 1, the 10 year forecast of M2 predicts > 3 ENM accidents with a high  
199 probability (99%), which is in line with the results from M1 suggesting (with 100%  
200 probability) at least 3 minor ENM accidents in 10 years. Most probable results from M2  
201 further suggest with about 80% probability > 20 accidents within the next 10 years and > 60  
202 events within the next 30 years, which agrees with the predictions derived from M1.

203 At level 2 (more severe accidents) and following M1, at least one ENM accident  
204 (calculated in Table 1 as 1-P(0 events)) is predicted with a high probability (64%) within the  
205 next 10 years, and such an event is even more likely (94%) in 30 years. These values are in  
206 excellent agreement with the results from M2, suggesting a very high likelihood of at least  
207 one accident every 10 years (95%) and every 30 years (almost 100%).

208 For level 3, the probabilities derived from M1 of one or more events within 10 years  
209 were around 50%. For a 30-year period, the probability of one or more events increased to  
210 around 85%. In M2 at least one accident of level 3 within the next 10 years seems even more  
211 likely (79%), and in 30 years the probability of one or more accidents reaches almost 99%.

212 At least one level 4 ENM accident is possible (29%) based on the 10 year predictions  
213 of M1. During the next 30 years, at least one event seems rather likely (probability of 62%).  
214 The results of M2 suggest slightly higher probabilities of 45% (10 years) and 91% (30 years)  
215 for at least one level 4 ENM accident and are, thus, in agreement with the results from M1.

216 For severe accidents at levels 5 and 6, with even more ( $10^4$  and  $10^5$  times more)  
217 material released, the average 10-year probabilities resulting from M1 for at least 1 event  
218 range from 4 to 7% (Figure 3). The most conservative Monte Carlo simulations revealed  
219 probabilities of around 11% and 18% for the 10-year forecast for level 5 and 6 (Table 1),  
220 respectively. The corresponding worst-case probabilities in the 30 year predictions are 45%  
221 and 27% for levels 5 and 6, respectively. The results from the M2 computations for severe  
222 ENM accidents are generally lower compared to the corresponding results from M1, with a  
223 probability of around 10% for at least one level 5 event within a 30 year period (Figure 3). To  
224 what extent a level-6 release event can be excluded based on almost zero M2 results is  
225 difficult to evaluate. Regarding the difficulty of interpreting almost zero M2 probability  
226 values for level 6, one may refer to discussions in the nuclear power sector: “The  
227 interpretation of a probability, such as  $10^{-10}$  per year, runs into some philosophical questions.  
228 It is mathematically correct to interpret this probability as meaning a chance for the postulated  
229 accident to occur once in  $10^{10}$  years, a period of time exceeding the age of the earth (about  $4.5$   
230  $\times 10^9$  years)”.<sup>38</sup> However, per our computations, the output range and uncertainty are  
231 considerable due to both the limited number of accidental release records at this maximum  
232 release level (none of the reported accidents involved nanomaterials) and the uncertainty of  
233 the extrapolation of the frequency observed in the analysed sample (1000 accidents) to this  
234 maximum release level. The past accident of Blanzky,<sup>37</sup> however, during which about 5 tons

235 of carbon black were released suggests that large accidental ENM releases may happen in the  
236 future.

237 The good agreement observed in all the results compared above between the outputs  
238 of model M1 (fixed share of ENMs) and model M2 (expert opinions) is reflected in Figure 3  
239 and visualized by projecting the M1 model output into the optimistic-realistic M2 simulation  
240 in Figure 2. The model values of the probabilities for an accidental ENM release for both  
241 prediction periods (10 and 30 years) are in excellent agreement (see the blue curves in 'Expert  
242 3' calculations in Figure 2).

243 Based on the demonstrated validity of the calculation methodology in the nuclear  
244 power industry,<sup>39</sup> a systematic documentation of accidental ENM releases, also including  
245 specific information about the ENM substance and the spilled amount, would decrease the  
246 model uncertainties. Two scenarios developed during a workshop<sup>40</sup> are offered in the  
247 supplementary S4 that can serve as the basis for identifying the type of data that should be  
248 collected during such incidents for improving, amongst others, the predictive modeling and its  
249 benefit for policy regulation purposes. Of utmost importance are improved information on  
250 accident frequencies and on the severity of their consequences (ENM hazardous  
251 characteristics).<sup>40</sup> Our modeling study establishes a baseline and the results of a related  
252 workshop in our supplementary S5 the range of future challenges for accident probability  
253 estimations when also addressing questions of socio-economic regulation and ENM hazard.

254 Our study expands a robust methodology from the nuclear power industry to predict  
255 the likelihood of accidents involving ENMs by stochastically addressing the extreme  
256 uncertainty in the available data. The quality of such data depends on our level of  
257 understanding in relating documented chemical accidents to accidents involving ENM release  
258 and thus to reconstruct a historical database from ENM accidents. Although M1 and M2 were  
259 different and fed with independent datasets their outputs were very similar. The broad  
260 spectrum of the model input carries over to a large spread of the model output which occurred  
261 for both model calculations.

262 Our modelling approach did not address questions relating to the hazard resulting from the  
263 exposure to accidentally released ENM<sup>41-43</sup> as this would further complicate the assessment.  
264 In accordance with others,<sup>44</sup> commercial ENM are presently compared to nuisance dusts and  
265 pigments and are not necessarily as hazardous as, for example, high-reactivity chemicals.<sup>26</sup> It  
266 is interesting to note that there have been very few reports from insurers regarding how to  
267 deal with ENM risks. However, others<sup>42</sup> have explicitly pointed out insurers' lack of  
268 scientific guidance in this regard for underwriting consumer, occupational, general, accident-  
269 based, or other ENM risks. An early report from Lloyd's of London,<sup>45</sup> concluded that "Our  
270 exposure to nanotechnology must therefore be considered and examined very carefully".  
271 Nanotechnology related accidents apparently caused very few financial losses, so there is  
272 little by way of example that can be used to support insurers' responses to nanotechnology  
273 driven accidents.

274

## 275 **Conclusions**

276 Our pioneering predictions provide a basis for further investigation of the risk of accidental  
277 release of ENM, and help to evaluate the societal impacts of accidental release of ENM,  
278 which range from emergency response to site remediation. With this contribution we provide  
279 initial numbers and uncertainties of the likelihood of future accidental ENM release events, so  
280 that risk analysts and other stakeholders no longer have to rely on their gut feelings when  
281 predicting those accidents. We were challenged by enormous uncertainties due to limited  
282 knowledge and fragmentary documentation of those accidents in the past. However, the well-

283 corresponding results from different stochastic approaches that unfold in two largely different  
284 models, make us feel confident to have approached realistic predictions of the accidental  
285 release of ENMs happening in the future. In the study, we assume that the probability of an  
286 accidental release of ENM is independent of the type (and the hazardous potential) of the  
287 ENM. This assumptions may or may not be justified as different safety measures may be  
288 applied during for example the transport of ENMs. However, the fragmentary data available  
289 on past accidents forced us to lump all ENM accidents into one ENM category. We are aware  
290 of the limitations of our model calculations which are in large parts caused by the  
291 reconstructions of the frequency of past accidental release of ENMs. More accurate reporting  
292 of ENM accidents as suggested in our work, would, thus, greatly decrease the uncertainties  
293 associated with our model results. If, in addition, also information about the the  
294 hazardousness of the released material were reported, the accidental release predictions could  
295 be extended and be used by risk analysts for hazard forecast and classification purposes as  
296 well.

297

298



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316

## 317 **Author Contributions Statement**

318 F.G. and R.K. conceived and designed the study. F.K. and B.P. provided insights from the  
319 industry, F.G., B.D., J.M.L., A.V., V.P.P., S.L. and S.H. developed and commented the  
320 modelling approaches, S.V.C., C.O.H., and C.S. provided context for future challenges for  
321 future accident probability estimations. F.G. and R.K. wrote and edited the paper, with  
322 contributions and support from all co-authors.  
323  
324

## 325 **Competing Interests Statement**

326 The authors declare no competing interests.  
327  
328

329 **Methods** Going beyond qualitative discussion of accidental ENM release, we aimed to compute the first  
330 quantitative probabilities of accidental ENM release. The modelling approach presented here is based on  
331 probability-generating functions (PGF), which are extensively used to perform fundamental probability  
332 calculations and have also been applied to estimate accident likelihoods in the nuclear power sector.<sup>39</sup> With  
333 available past accident statistics, as seen in our review for this sector (supplementary S1), those calculations do  
334 as well or better in terms of their predictive precision than predictions that partly or completely follow the  
335 mathematical-mechanistic considerations of accidental release cause, process, and frequency. However, in  
336 contrast to their use with robust empirical data in the nuclear power sector, we have developed a stochastic  
337 approach to process these functions in order to account for the significant uncertainties associated with the  
338 identification of (past) ENM accidents. Those uncertainties develop in the model by varying e.g., the annual  
339 accident rates and associated probabilities of occurrence, as well as combinations thereof.  
340

341 The developed stochastic methodology (for details see supplementary S3) sets alongside other parameters,  
342 values for the coefficients of the probability mass functions  $p_x(x)$  and the exponents of  $z^x$ , computing a plethora  
343 of PGFs (defined by  $G$  in Equation 1).  
344

$$345 \quad G_x(z) = \sum_{x=0} p_x(x) z^x \quad (1)$$

346  
347 These coefficients represent the probabilities that can be associated with different outcomes in a particular time  
348 period reflected in the exponent, in our case the annual number of accidental release events. However, the PGFs  
349 themselves are only a practical means for handling the probability calculations; the variable  $z$  has no further  
350 meaning for the probability considerations. We stochastically vary the data within and outside of those functions

351 by identifying the possible (empirical or historical) numbers of accidents, their transformation into considered  
352 annual values, and their probabilities, thereby exploring a large spread of model inputs and outputs. Such a  
353 spread can be derived from highly uncertain and incomplete knowledge about the number of past accidental  
354 ENM release events.

355  
356 The minimum requirement for developing ENM-based model input values is that we use a particular past time  
357 period during which accidental release events have either happened or can be assumed to have happened. Based  
358 on such initial raw data frequencies, the probability for varying numbers of accidents is computed (e.g., for 10  
359 and 30 years) using higher-order polynomial calculations (Equation 2). These are needed for multi-year  
360 timeframe predictions and are achieved by raising the corresponding function to the needed target power,  $n$ :  
361

$$(G_x(z))^n \quad (2)$$

362  
363 Hence, expanding the polynomial reveals all possible ways of differentiating the number of accident events and  
364 their respective probabilities over varying time horizons. This, however, only works under the condition that the  
365 accident frequencies/probabilities do not substantially change during the used prediction timeframe. Thus, faced  
366 with significant ENM-accident-frequency uncertainty and knowledge gaps concerning the number of past  
367 accidents involving ENM release (or not), we transformed the deterministic use of PGFs into a stochastic one.  
368 Instead of generating one function (Equation 2) for each prediction, we conduct for example,  $m=10,000$   
369 iterations, based on Monte Carlo (MC) routines, allowing us to combine the emphasized broad spectrum of  
370 possible model input data. In doing so, we algebraically manipulated the polynomial coefficients, multiplying  
371 out the PGFs embedded in the MC iteration routines (Figure S1). This process allows one to vary the empirical  
372 accident frequency for each MC iteration and produce the statistical forecast output that is used to compute  
373 density (probability) curves and evaluate ranges and central tendencies (Equation 3).  
374  
375

$$\frac{\sum_{j=1}^m G_j^n}{m} \quad (3)$$

376  
377 The differences in the model input may stem from, for example, variations in the relevant time periods for  
378 deriving empirical accident data and from the different probabilities (0–100%) of the ENM being released from  
379 past accidents. We may have an sound overview of incidents that resulted in the release of chemicals during a  
380 certain time period; however, it is up to us to assess whether ENMs were released during these incidents. The  
381 data input differentiation covers the assumed differences between past, current, and future accident frequencies  
382 that may change over time due to factors such as technological developments.  
383  
384

385 The modelling was programmed in R,<sup>46</sup> and the possible conceptual programming procedures are shown in  
386 supplementary Figure S1 and S2. These procedures, however, vary from case to case. A single, exemplary model  
387 of a MC routine is shown in supplementary Figure S3, for iteratively drawing, for example, a die with a 1-50%  
388 probability of sampling an ENM accident.  
389

390 We also tested our stochastic output (supplementary Table S6) by remodeling results from the nuclear power  
391 sector.<sup>39</sup> In addition, we performed a comparative evaluation of our results based on own Poisson MC  
392 computations. We modelled the reference case study on worldwide accident predictions (International Nuclear  
393 Event Scale  $\geq 4$ ), and the results were similar.  
394

395 The stochastic variation in our calculations may stem, as discussed, from the derivation of the empirical accident  
396 numbers from selected time periods and the expert-based assessment of the fraction (probability) of the ENMs  
397 released during the respective accidents. If we consider these probabilities at the plant scale, technological  
398 advances may be the major factor affecting plant accidents in the future (including advances in technology  
399 driven by regulation). If we consider the regional scale, the major driver controlling accidental release of ENMs  
400 in the future, is the size of the market and the number of production plants. The supplementary S1 and S3  
401 provide a full description of the data and methods used in the modelling studies and the critical methodology  
402 review that drove our own methodology development.  
403

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557

558

559 **Data availability**

560 The datasets generated during and/or analyzed during the current study are available from the  
561 corresponding author / first author on reasonable request.

562

563 **Code availability**

564 R scripts are available from the corresponding author / first author on reasonable request.

565

566 **Supplementary information**

567 Supplement Information on Materials and Methods (S1), Key limitations of the data (S2),  
568 Methodology (S3), Supplementary section on a Workshop on Spills (S4), Bridging NanoEHS  
569 Workshop on Spills (S5), Figures (Figures S1 to S3) and Tables (Tables S1 to S6) (S6),  
570 References (S7)

571

572

573

574 **Figures captions:**

575 Figure 1. Accident probability predictions according to model 1 (M1). Probability (density)  
576 curves for the probabilities (x-axis) of at least one accident resulting in engineered  
577 nanomaterial release of a certain severity (level 1 - 6) in the next 10 (A) and 30 (B) years  
578 (worldwide) based on model M1. Compare these curves with the M1- results in Table 1

579 Figure 2. Accident probability predictions according to model 2 (M2). Probability (density)  
580 curves for the numbers of accidents associated with engineered nanomaterial release in 10  
581 (density curves on the left side of each graph) and 30 years (density curves on the right side of  
582 each graph) worldwide. Each illustration (1-11) represents results based on data input  
583 following the assessment of one expert (1-11) and computed in model M2. In the graph of  
584 expert 3, the results from model 1(M1, blue line) and M2 (green area) are projected in the  
585 same graph for comparison (see also M2 results in Table 1 for comparison).

586 Figure 3. Probabilities (P in %, mean values) for engineered nanomaterial accidental release  
587 in the next 10 (top) and 30 (bottom) years (worldwide) computed based on two models (M1  
588 and M2) and for different amounts of materials release (Level 1 – Level 6, see Table S2 for an  
589 explanation of the different levels). The most realistic scenarios are highlighted in yellow and  
590 underline the good agreement between the outputs of the two models (M1 and M2). There are  
591 no linearity or dependency between levels and between models. Due to rounding effects, the  
592 values do not necessarily add to 100%. Accident predictions lower than 0.005% (in all our  
593 tables) have been set to zero (n.c. = not computed).

594 Figure 4. Probabilities (P, mean values) of ENM accidental release level 1 in 10 and 30 years  
595 worldwide. These results reflect expert opinions based raw data used in the modeling for level  
596 1 accidents (see Table S2 for an explanation of level 1). There are no linearity or other  
597 dependencies between these sub-models (for each expert 1-11) of Model 2 (M2).

598



599 **Tables:**

600 Table 1. Optimistic and pessimistic accident probability predictions. Minimum (Min) and  
 601 maximum (Max) probabilities for accidental release of engineered nanomaterials (worldwide)  
 602 during the next 10 and 30 years calculated based on model M1 and M2 (see main text for the  
 603 description of the two models and supplementary Table S2 for a description of the different  
 604 levels).

<b>One accident (at least) in 10 years</b>	<b>Min (M1)</b>	<b>Max (M1)</b>	<b>Min (M2)</b>	<b>Max (M2)</b>
<b>Level 1</b>	100.0%	100.0%	97.6%	100.0%
<b>Level 2</b>	31.1%	92.5%	47.9%	100.0%
<b>Level 3</b>	22.6%	82.1%	0.9%	100.0%
<b>Level 4</b>	11.5%	56.2%	0.0%	100.0%
<b>Level 5</b>	2.7%	17.5%	0.0%	0.0%
<b>Level 6</b>	1.5%	10.8%	0.0%	0.0%
<b>One accident (at least) in 30 years</b>	<b>Min (M1)</b>	<b>Max (M1)</b>	<b>Min (M2)</b>	<b>Max (M2)</b>
<b>Level 1</b>	100.0%	100.0%	98.8%	100.0%
<b>Level 2</b>	70.2%	99.9%	98.8%	100.0%
<b>Level 3</b>	51.2%	99.2%	86.7%	100.0%
<b>Level 4</b>	31.8%	93.5%	23.4%	100.0%
<b>Level 5</b>	8.3%	44.6%	0.0%	66.9%
<b>Level 6</b>	4.7%	27.1%	0.0%	0.0%