

Deliverable report for

SUN

Sustainable Nanotechnologies

Grant Agreement Number 604305

Deliverable D 7.6
Guidelines for safe handling of waste flows containing
NOAA

Due date of deliverable: 01/10/2016

Actual submission date: 30/09/2015

Lead beneficiary for this deliverable: DTU

Dissemination Level:		
PU	Public	
PP	Restricted to other programme participants (including the Commission Services)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	X

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1. Description of task

This Deliverable 7.7 is based on the work performed under Task 7.4 and 7.5.

The objectives of Task 7.4 comprised:

- Identification of critical waste flows and treatment types, with specific focus on i) mechanical grinding of solid materials, with volatilization of NOAA; ii) recycling-oriented thermal processing of plastic materials, with volatilization of NOAA; iii) application on land of Waste Water Treatment Plant (WWTP) sludge by means of mechanical spreader, with volatilization of NOAA.
- Identification of safe handling procedures and best waste treatment options for the identified waste fractions
- Screening of the of the state-of-the-art tail-end control equipment for minimizing workplace exposure during waste treatment and evaluation of its efficiency.

The objectives of Task 7.5 comprised:

- Providing recommendations on how to minimize the release of NOAA to the environment during waste treatment.
- Address possibilities for minimization of waste from manufacturing process.
- Providing recommendations on improved collection of manufacturing waste.
- Identification of material properties (e.g. combustibility) that should be considered in the design phase to minimize release of NOAA in the waste phase. The material properties will be specific for individual waste materials, depending on the expected waste treatment. For example, if an item is expected to be incinerated, the complete combustion of NOAA is a desired property.
- Discussing the role of product labelling in facilitating waste handling and routing to proper waste treatment will be discussed.

In this deliverable, we address these objectives. Some of the findings draw on work that went on Task 3.4 of WP3.

2. Description of work & main achievements

The present deliverable was prepared by Alessio Boldrin, Laura Roverskov Heggelund, Nanna Hundebøll and Steffen Foss Hansen. In the following, we first provide an introduction on nanowaste and a definition of its domain, while explaining the importance of addressing NOAA in waste. We thus assess the waste material fractions expected to carry NOAA, the most prominent NOAA types, and the waste technologies involved in nanowaste handling. We then provide a brief description of individual waste treatment technologies, followed by a brief discussion on how the operation of individual processes may be affected by the presence of NOAA, and an overview of NOAA release pathways and important factors affecting it. Finally, based on relevant factors identified in the previous sections, we present a series of roadmaps for assessing release and exposure from different waste technologies. These roadmaps provide the basis for the guidelines on safe handling of nanowaste and safe-by-design recommendations that constitutes that final chapter of the report.

1. *Introduction*

1.1. *Waste and nanowaste*

Waste management is a set of activities aiming at prevention, monitoring, treatment, handling, reuse and residual disposition of solid wastes, where ‘waste is a left-over, a redundant product or material of no or marginal value for the owner and which the owner wants to discard’ (Christensen, 2010). Being “waste” is not an inherent characteristic of a material or product, but it is a property depending on the specific situation waste is generated. The value (present or future) of waste is in fact a function of social, geographic and economic factors, such as time, location, states of the waste material, personal income and personal preferences.

With the increasing production and utilization of engineered nanomaterials, growing amounts of waste containing NOAA are expected to be generated in the near future. As long as the presence of NOAA in waste is not recognized being particularly dangerous (thus needing a possible classification of waste as hazardous), waste-containing NOAA will be collected and managed similarly to regular waste and according to its visual appearance (e.g. plastic, glass, paper). Thus presence of NOAA is considered as one of the many physicochemical characteristics of waste, meaning that, unless specific hazardous properties are reported, no specific classification, regulation, and management are needed. With respect to nanowaste, further clarifications are necessary to distinguish between waste and environmental pollution, where the latter considered as an unwanted (in terms of place, time, and location) release of NOAA to the environment. Such release may occur rather often, in particular because of the inner property of NOAA (i.e. the size). Nanowaste can thus be defined as collected or collectable waste materials being, containing or carrying engineered NOAA (Boldrin et al., 2014). The domain for nanowaste definition is outlined in Figure 1 and include:

- NOAA as a single fraction, e.g. by-products from manufacturing of nanoproducts;
- end-of-life (EOL) nanoproducts;
- individual waste materials being contaminated with NOAA, e.g. sludge from wastewater treatment.

In all cases, nanowaste is generated because the NOAA or the nano-enabled products are considered having no value or no utility to the owner, and are thus discarded.

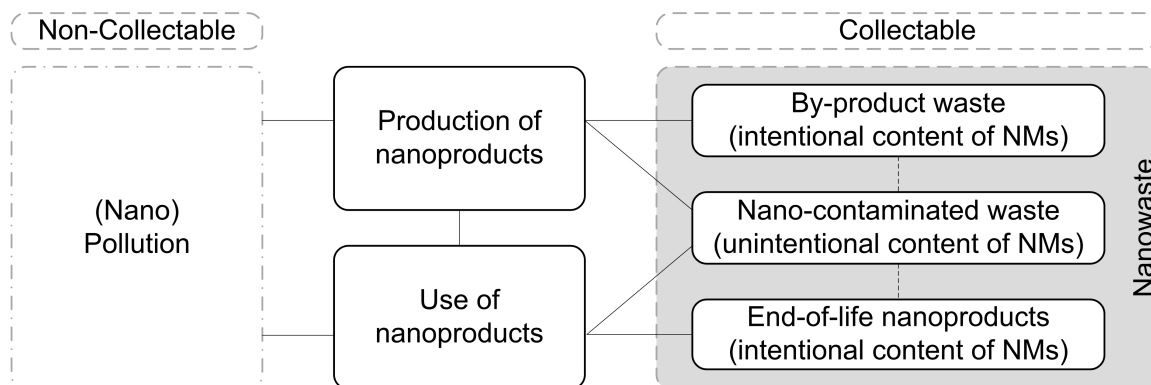


Figure 1 – Generation of solid waste containing NOAA (nanowaste) throughout the life cycle of nanoproducts. Nanowaste is shaded in grey. Nano-contaminated waste can originate from both the production and use phases of nanoproducts, and occasionally from waste treatment. The figure is taken from Boldrin et al. (2014).

1.2. Addressing nanowaste: why and how

The emergence of nanoproducts raises a number of important issues when it comes to the end-of-life of these products (see e.g. Gottschalk, F., Nowack, B., Gawlik, 2010; Roes et al., 2012; Royal Commission on Environmental Pollution, 2008) and waste treatment and waste handling, as very little is known about (Boldrin et al., 2014; Stone et al., 2010):

- the potential transformations of the nanoproducts and NOAA during different waste treatments;
- the interactions between NOAA and other constituents of waste;
- the magnitude of NOAA released to the environment after waste treatment;
- the potential transformations/effects of the modified NOAA in the environment;
- the potential exposure in working environment during waste handling.

With the increasing societal use of NOAA, concerns about the final life stage of products containing NOAA have been surfacing repetitively, but information on the behaviour of NOAA in waste treatment systems is missing. While the EOL phase was excluded in several studies (e.g. Li, 2012) because of lack of data, other studies mention that it cannot be excluded that some of the NOAA-enabled products may be in some cases characterized/classified as hazardous waste (Andersen et al., 2014), thereby suggesting that the waste management should not be excluded a priori from risk and exposure assessment.

Most importantly, the SUN milestone report 4 “Map of release hot spots”, which is part of Task 2.1 “Map hot spots of release of nanomaterials”. The objective of Task 2.1 was firstly to comprise a description of the SUN case studies (CS) with their life cycle stages (i.e., production, use, and end-of-life) and, secondly, to identify potential environmental hot spot releases for every CS in every life cycle stage. In the milestone report, it was found that there was a high or very high release potential of nanomaterials to waste from the use phase of many of the CS. It was furthermore observed that many of the CS EOL phase included either recycling, incineration, landfilling or composting (see figure 2-13 in SUM milestone report 4 “Map of release hot spots”).

Comprehensive assessments are difficult and generalization of results is not yet possible, as more research is needed to unfold several yet unexplored aspects of nanowaste (Andersen et al., 2014). It is nevertheless clear that five overall processes that are relevant to consider with respect to post-consumer nano-enabled products:

recycling, incineration, landfilling, biological treatment (i.e. anaerobic digestion (AD)/composting), and chemical treatment of special waste types (Andersen et al., 2014). The last two are hereby not included, as it is not expected that significant amounts of NOAA are entering biological treatments (Reihlen and Jepsen, 2015), while no data is available with respect to chemical treatment.

2. Waste types, NOAA and waste treatments in Europe

To properly address the abovementioned concerns, basic understanding of NOAA flowing into and throughout waste systems is needed, addressing for instance which types of nanoproducts are expected to enter current waste management systems, as well as which kind of NOAA are the most abundant and in what form they reach waste treatment facilities. This has been recently investigated adopting different modelling approaches, including material flow modelling (e.g. Gottschalk, F., Nowack, B., Gawlik, 2010; Sun et al., 2014; Walser and Gottschalk, 2014), market analysis (Boldrin et al., 2014; Keller et al., 2013; Keller and Lazareva, 2013), life cycle assessment (e.g. Pourzahedi & Eckelman 2015) and modelling by categorization based on consumer product inventories (Asmatulu et al., 2012).

Probabilistic material flow models have been used to predict environmental concentrations of NOAA (Gottschalk, F., Nowack, B., Gawlik, 2010; Sun et al., 2014; Walser and Gottschalk, 2014) and concentrations in recycling processes (Caballero-Guzman et al., 2015), using probabilistic distribution curves as input data instead of the generally uncertain and inadequate datasets available on NOAA fate and behaviour in the environment. Recently, Sun et al. (2014) modelled the concentrations of selected NOAA (nanoTiO₂, nanosilver, nanoZnO, fullerenes and CNTs) in environmental and technical compartments and compared with the non-nano metal based counterparts (TiO₂, Ag, ZnO). Compared to the environmental compartments (air, soil, water), the highest modelled concentrations were estimated for the technical compartments which included e.g. landfilling, waste incineration, sewage treatment and recycling. Among these, the highest concentrations are expected in sewage sludge, followed by concentrations in solid waste and waste incineration ashes (fly and bottom ash). The technical compartments presented concentrations in the mg/kg range, whereas the environmental compartments only showed concentrations in the ng-ug/kg range. These findings support the relevance of further studies on NOAA quantification and characterization in waste treatment scenarios since these constitute a likely sink for the NOAA.

In the following, we present the work that we have done in order to i) develop tools for the analysis of nanoproducts in solid waste flows, ii) assess the relative importance of NOAA and waste types, iii) identify critical aspects with respect to specific NOAA and waste treatments combinations, and iv) establish EU waste scenarios to identify the waste treatment options involved in handling of NOAA.

2.1. Step-by-step procedure

The identification of relevant waste and NOAA types was based on a market approach, as also recently suggested in other studies (e.g. Reihlen and Jepsen, 2015). The work process was divided into four steps:

1. Categorization of the products available in the Nanodatabase into waste material fractions,
2. Identification of the types of NOAA present in the waste material fractions,

3. Identification of region-specific waste management of individual waste material fractions,
4. Combination of steps 2 and 3 to determine the distribution of NOAA routed to specific waste management options.

The four steps are explained in details in the following sections.

2.1.1. Step 1: Categorization of the products available in the Nanodatabase into waste material fractions

The first step in the process towards mapping the abundance, distribution and waste treatment of nanoproducts is to assign individual nanoproducts to waste material fractions. To do this, we used The Nanodatabase (www.nanodb.dk), an online inventory of products known to be available to European consumers either via retailers or via online shops. The database was established by DTU and others in 2012 and currently contains more than 2300 products (19/09/2016) known to be available on the European market and claiming to contain NOAA or be based on nanotechnology. In the Nanodatabase, a series of information about each product is collected based on the information that producers have provided publicly online such as type of NOAA or description of product.

Based on the information available in the Nanodatabase, we identified the main matrix material of individual product and its corresponding waste material fraction, e.g. a product in a plastic container falls into the waste fraction called “plastic packaging”. The categorization is based on an image of the product, along with any other information made available by the manufacturer online. This infers that a waste fraction only exists in the database if a product has been assigned to the given fraction. In some cases, we created specific fractions which were adapted to the products in the database, e.g. “Plastic from used product containers”. Other products made from plastic, e.g. hockey sticks, food containers or baby bottles, are separated from the plastic packaging waste, since the NOAA is usually embedded in the plastic or coated on the surface of these products, and they are considered potentially more suitable for recycling. Hence, they might also be handled differently in the waste management system. Sometimes it was not possible to create a homogenous fraction e.g. when a product contained more than one material which could not readily be separated (e.g. camera lenses, baby carriage and water filtering unit). These products were grouped into a “Multi material waste” fraction, comprising products of many sizes and applications. Products were categorized as unknown if no image is available showing the specific product and the container it is sold in, or it cannot be derived from the product description. An overview of the ten identified waste fractions is provided in Table 1.

2.1.2. Step 2: Identification of the types of NOAA present in the waste material fractions

For each product in The Nanodatabase, the identity of the NOAA claimed to be used in the product has been noted, if reported by the manufacturer. This information was subsequently used to identify NOAA types in the waste material fraction.

Table 1 – Overview of waste material fractions identified in nanodb.dk (taken from (Heggelund et al., 2016)).

Waste material fraction	Description and examples
Batteries	Typically these products are batteries for an electrical item, where the nanocomponent is only present in the battery.
Electronics	Generally products with electric connection, e.g. refrigerator, hairdryer, electric toys etc.
Glass	Cosmetic products, supplements or other products sold in glass containers.
Metal	Generally consists of containers such as cans, or metal sports equipment.
Multi material waste	These products contain more than one material which cannot readily be separated e.g. camera lenses, baby carriage and water filtering unit.
Plastic packaging	Typically waste from used plastic containers for cleaning/cosmetic products, which have a residue of the nanoprodut in them.
Plastic, other	Generally plastic products where the nanocomponent is embedded in/surface coating the plastic.
Textile	Fibrous material typically clothing or bandage with a nanocoating or NOAA embedded in the fibers.
Unknown	If no image is available showing the specific product and the container it is sold in, or it cannot be derived from the product description, this category is used.

2.1.3. Step 3: Identification of region specific waste management of individual waste material fractions

As the involvement of individual waste technologies and subsequent release rates will depend on the local/regional waste management system as well as on its technological level, we collected recent data on waste treatment of selected waste fractions for individual European countries from Eurostat (see

Table 2), and included the following four possible waste treatment options: incineration, recycling, landfilling and AD/composting. In this study, we analysed three case studies: Europe (EU average), United Kingdom (UK) and Denmark (DK), as shown in Figure 2. We chose these regions because they are within the scope of nanodb.dk and represent different waste management systems, i.e. an average for Europe, a system traditionally relying on landfilling (UK) and one making significant use for incineration and energy recovery (DK). The statistical data was used to route individual waste material fractions to their likely waste treatment option. The fractions created for the purpose of this analysis, and which had no appropriate waste treatment statistics available (i.e. “Plastic, other”, “Multi material waste” and “Unknown”), were routed to the waste management system using statistics for handling of “Municipal solid waste (MSW)”. Further, data for “Plastic, packaging” was used to route the fraction “Plastic, from used product containers” into waste treatment options. While in the present we mostly addressed municipal waste, additional waste types of interest could be tyres and construction waste, as reported by Andersen et al. (2014).

Table 2 - Treatment of individual waste material fractions in the three analysed scenarios: Europe (EU), Denmark (DK), and the United Kingdom (UK).

Waste material fraction	Distribution to waste treatment options (%)											
	Incineration			Landfilling			Recycling			AD/compost		
	EU	DK	UK	EU	DK	UK	EU	DK	UK	EU	DK	UK
Batteries	4.8	4.8	4.8	7.7	7.7	7.7	87.6	87.6	87.6			
Electronics	6.0	10.9	4.3	8.8	0.5	9.6	85.1	88.6	86.8			
Glass	11.5	2.2	10.1	15.7	0.1	22.2	72.8	97.7	67.8			
Hazardous	13.9	13.3	12.9	47.8	53.3	38.7	38.3	33.2	48.4			

Metal	11.6	46.0	14.9	15.9	2.2	33.0	72.5	51.8	52.1			
Plastic, packaging	27.4	67.3	23.3	37.3	3.3	51.5	35.3	29.4	25.2			
Average MSW	24.2	52.3	16.8	33.6	2.5	36.9	27.4	32.2	28.4	14.9	13.0	17.8
Textile	17.2	3.3	4.7	26.3	38.9	2.4	56.5	57.7	92.9			

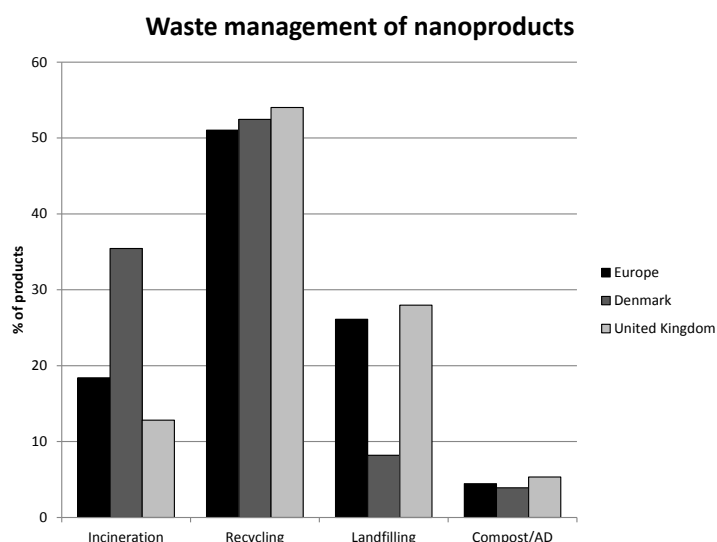


Figure 2 - Relative distribution of end-of-life nanoproducts into waste treatment options in the three analysed scenarios: Europe (EU), Denmark (DK), and the United Kingdom (UK) (taken from (Heggelund et al., 2016)).

2.1.4. Step 4: Combination of steps 2 and 3 to determine the distribution of NOAA routed to specific waste management options

Further analysis of the distribution of NOAA in the most common solid waste treatment technologies, i.e. incineration, recycling, landfilling and AD/composting, was performed by combining the waste material fractions (step 2) with waste treatment scenarios for a specific region (step 3). By doing that, we were able to identify important/critical loads of NOAA in waste treatment systems and the involved waste technologies. While in the present study we report on the NOAA distribution in Denmark, United Kingdom and Europe, a similar analysis can be performed for other specific regions or countries, as long as detailed data on the local waste management system is available.

2.2. Results

2.2.1. NOAA distribution in waste material fractions

The distribution of the NOAA (or combinations of NOAA, e.g. nanosilver and nanoTiO₂) expected to be found in individual fractions is shown in Figure 3, according to the number of products assigned to respective waste fractions. We found that the most abundant NOAA across all waste fractions is nanosilver (Figure 3). This is in line with the wide range of applications for nanosilver in consumer products. For example nanosilver is used in 20 different product subcategories corresponding to 210 products (nanodb.dk). In comparison, nanoTiO₂ is only used in 10 different product subcategories, although it is quite an abundant material used in 65 products (nanodb.dk). The largest waste fraction is “plastic packaging” (116 products) followed by “electronics” (88 products). Plastic waste from used product containers also comprises the largest variety of NOAA (18 different NOAA or combinations of NOAA). This might be caused by the fact that this waste material is generated from many different sources such as the automotive, food & beverage and home & garden sectors.

Minor waste material fractions include, for example, hazardous and metal wastes which are primarily generated from the home & garden and health & fitness sectors, respectively.

While more than 50% of the analysed nanoproducts are likely to end up in recycling processes (Figure 2), this will not be the final destination for these products, as it is expected that only a fraction of these will be routed back to production and manufacturing industries (Caballero-Guzman et al., 2015). Significant differences appear when looking at the incineration and landfilling treatment options. Europe and UK are quite comparable, routing 18% and 13% to incineration and 26% and 28% to landfilling respectively (Figure 2). Conversely, Denmark makes use to a large extent of incineration with energy recovery, resulting in 35% of nanoproducts ending up in waste incineration plants and only 8% in landfills (Figure 2).

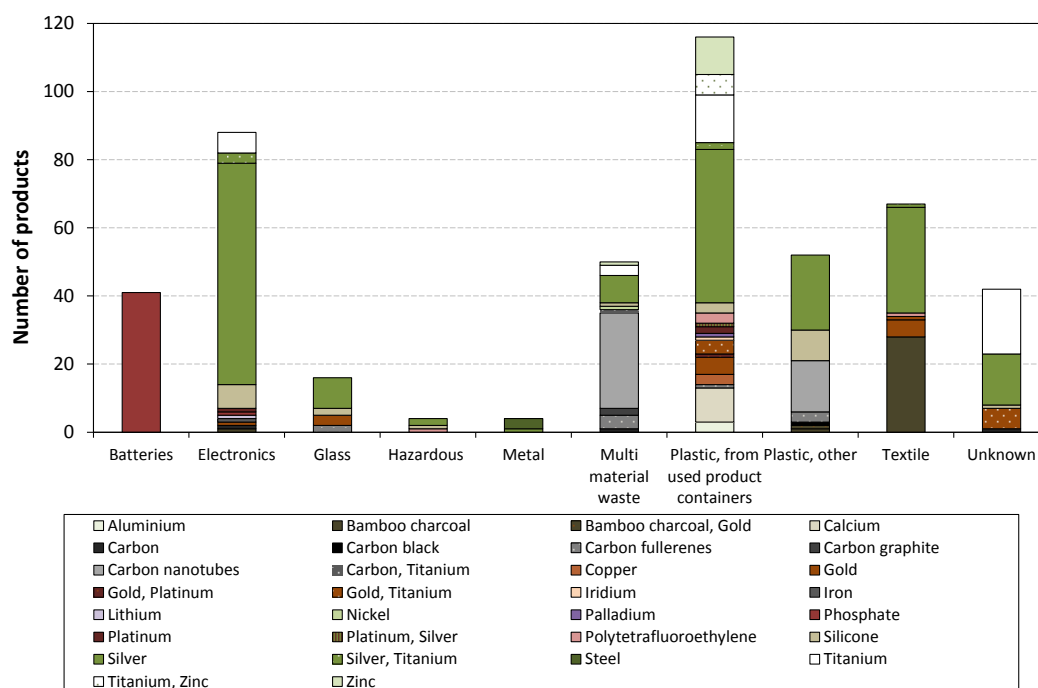


Figure 3 - Distribution of NOAA in the different waste material fractions according to data from nanodb.dk. The Y-axis represents the number of products containing a certain NOAA (taken from (Heggelund et al., 2016)).

2.2.2. NOAA Distribution to incineration, landfill, recycling and AD/compost

When we combined data from step 2, where we investigated the NOAA distribution in various waste fractions, and step 3, where we collected statistical data for waste treatment of these fractions in the EU, the UK and Denmark, we could assess to what extent individual NOAA would enter specific waste treatment technologies (i.e. incineration, recycling, landfilling and AD/compost), as shown in Figure 3. For example, 40% of EOL nano-enabled consumer products in Europe entering a waste incineration plant will contain nanosilver.

We found that, as expected, nanosilver is wide spread and comprises the largest fraction of NOAA in all treatment scenarios and regions (Figure 3). The distribution of NOAA in the different waste treatment systems is generally similar for Europe, e.g. the number of items containing silver, titanium and gold NOAA is more or less the same independent of the treatment scenario. If we compare incineration and recycling in Europe, it is clear that there is a higher percentage of phosphate NOAA in the recycling scenarios, which

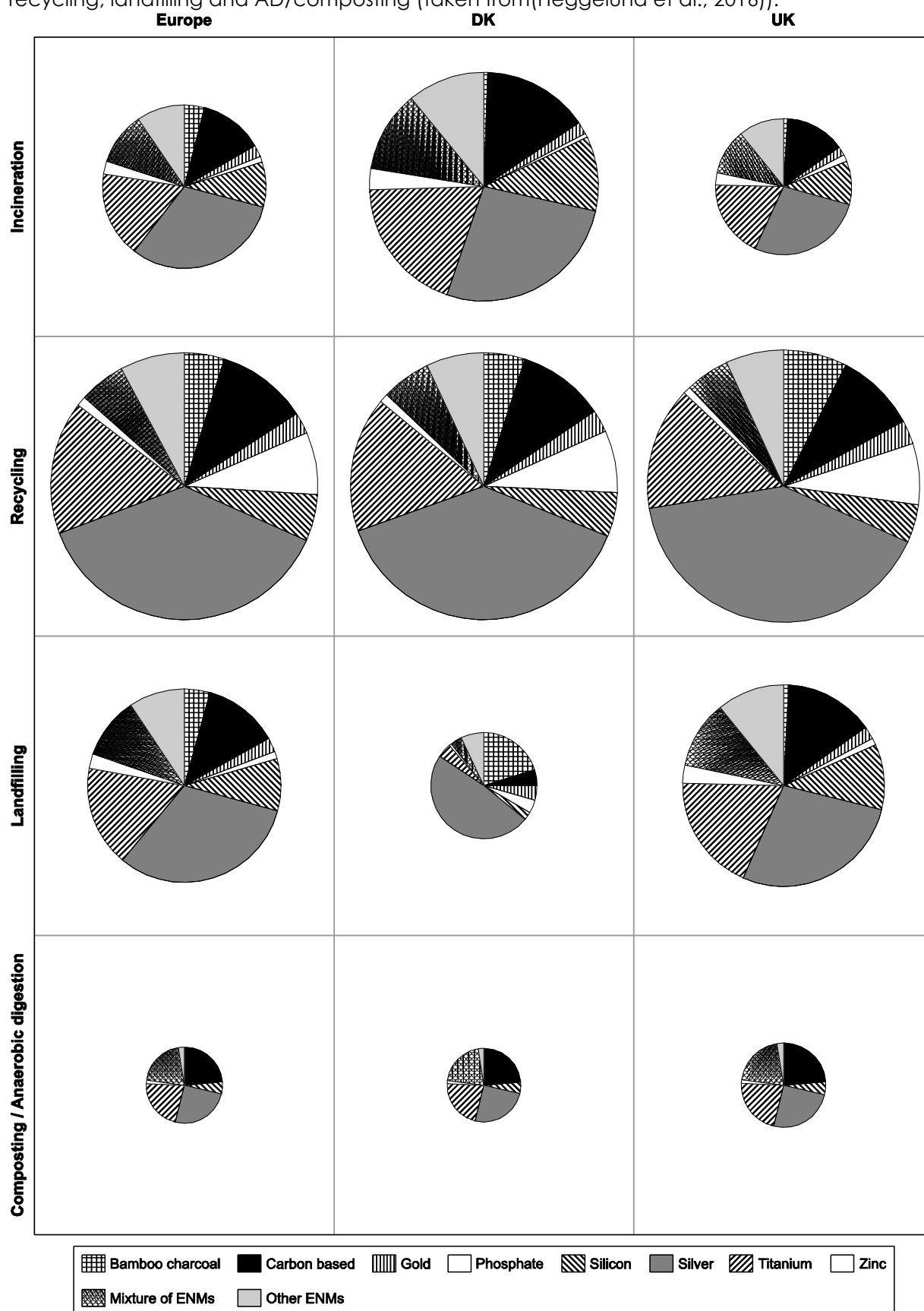
refers to the large amounts of batteries being recycled. On the contrary, we see a larger amount of carbon based NOAA entering waste incineration plants, which mainly originates from the multi material waste and waste from other plastic products.

To illustrate how different waste management systems can affect the distribution of NOAA, we included two additional regions in the analysis: Denmark (DK) and the United Kingdom (UK) (see figure 3). As we identified in the previous section (Table 2), the primary differences occur between the management of waste in Denmark and the two other regions. The fact that Denmark largely relies on waste incineration and makes only use of landfilling to a small degree, significantly affect the distribution of products and thereby the NOAA. Consequently, many more and more diverse products end up in landfills in the UK (135 products, 17 different types of NOAA) compared to Denmark (39 products, 11 types of NOAA).

Based on the products we identified, some main differences between Denmark and the UK occur specifically in the management of plastic packaging and textile waste (see Table 2). Because in the UK a larger amount of plastic waste (both packaging and other) is disposed of in landfills, the proportions of titanium and carbon based NOAA are higher in the UK landfill scenario. Whereas the Danish landfills, according to this data, receive larger amounts of textile which typically contains bamboo charcoal and gold NOAA. Also, batteries containing phosphate NOAA are also more prevalent in Danish landfills.

However, there is some discrepancy between the data we obtained from Eurostat, and the true Danish management of textile waste. In fact, it is not allowed to landfill textile waste in Denmark, which illustrates the importance of obtaining as accurate locale waste management statistics as possible.

Table 3 – Relative distribution of NOAA in the different waste treatment options: incineration, recycling, landfilling and AD/composting (taken from (Heggelund et al., 2016)).



2.3. Discussion

From our analysis, we found that nanosilver is ubiquitous in all the waste flows investigated. Further, recycling is identified as the compartment receiving most nanoproducts and thereby also a large variety of NOAA. It should be noticed that our analysis is based on the number of products appearing in a certain category/fraction of the Nanodatabase. The analysis provides no information regarding mass or volume of NOAA. Furthermore, many products in nanodb.dk contain an unknown NOAA, these products are not included in this analysis. Another source of uncertainty lies in the categorization of products. Some products are easily placed in a waste material fraction, e.g. electronics, textiles and metal, whereas others are not, e.g. hazardous and multi material waste.

2.3.1. Unknown NOAA used in the products on the European market

One of the major limitations of the present work relates to the fact that there is a great lack of information regarding the specific NOAA type used in many of the products available to consumers. This is reflected in the nanodb.dk inventory, where two out of three products contain a NOAA that is unknown. This illustrates the confusion and lack of regulation requiring labelling of NOAA of most nano-enabled consumer products, and also poses a major problem for our analysis. In this study we chose to exclude these products from our analysis, which decreases significantly the number of products we can analyse to approximately one third of the total number in the inventory.

2.3.2. Categorization of products into waste material fraction

In the initial step of the analysis, many considerations were made regarding the choice of material fractions and homogeneity of these fractions. A trade-off exists between having few simple categories and many detailed categories. On one hand, it is important for the routing of nanoproducts into waste treatment options that the waste material fractions are as homogenous as possible. On the other hand, the homogeneity may sometimes be compromised in order to decrease the number of individual material fractions, which would increase the complexity of the analysis. Another challenge is the handling of the custom-made waste fraction materials /categories such as “multi material waste” and “plastic, other”. We used statistics for treatment of MSW, in order to be able to analyse these products. However, as explained previously, these fractions are quite heterogeneous and to manage them as MSW might be a valid approximation for some but surely not for all products in the fractions. Alternatively, to increase the accuracy of the analysis, these products should be routed on a case by case basis, but this would make the analysis more time consuming.

Another challenge is the fact that the waste material fraction is difficult to identify for some products. This introduces some uncertainty in the analysis. An example is how to identify hazardous products, when labelling is not available (picture or description). Further, hazardous waste in this analysis is a very heterogeneous fraction. This is an issue since there might be large differences in how the individual product types would be managed, but in this analysis we are not able to separate them. For example, we would not be able to study the fate specifically of household paint waste. When we assigned each product in the database to a specific waste fraction based on the main matrix material, as opposed to dealing with general product categories and likely disposal of these, there is a risk of misplacing products. Some products carry a higher uncertainty because they are more complex and consist of different kinds of materials. This should be taken into account when interpreting the results of this analysis. The

complex products are often assigned to a waste fraction created for the purpose of analysing this product inventory (e.g. multi material waste), even though they can be very different, e.g. sports equipment, baby trolleys and camera lenses. This creates a problem when routing them to the waste treatment options. Finally, as some manufacturers do not provide information on the NOAA in their product and do not provide suitable photo and/or description of the product container/packaging, this means that we were forced to place these products in the waste material fraction “Unknown”.

2.3.3. Number based analysis and waste management statistics

In our analysis, we did not consider the mass of NOAA at any stage. Instead we use a number based analysis, which is very relevant since we deal with products actually available to consumers. However, it might easily be misleading, if the results are not carefully interpreted. For instance, we conclude that nanosilver is widespread across all waste fractions. However, one have to bear in mind that nanosilver is produced in very small amounts compared to e.g. nanoTiO₂. Also, we conclude that the largest waste material fraction is “plastic from used product containers”, but this fraction might contain the lowest amount of NOAA based on mass, since only a residual of the product, containing the NOAA, remains in the container by the time of disposal. While performing the analysis we also encountered a different problem concerning the chosen waste management data. First of all, the data we obtain from Eurostat is not detailed enough to describe exactly the waste fractions that we identified since we are dealing specifically with nano-enabled consumer products. Also, we found that the Eurostat database provided strange values in some cases (e.g. management of textile waste in Denmark), which are not corresponding with actual Danish practice. It might be a more useful approximation to utilize local waste management data if they are accessible and combine these with local expert judgements and estimates when statistics are incomplete. However, this approach compromises the comparability across countries or regions, which was also the aim of this study.

2.3.4. Our findings are in line with other research findings

Despite the limitations of our work, our findings are in line with other studies that have modelled the flow of NOAA from production, manufacturing and consumption into technical and natural compartments (Gottschalk, F., Nowack, B., Gawlik, 2010; Keller et al., 2013; Sun et al., 2014). The recent study by Sun et al. (2014) modelled the flow of selected NOAA based on yearly production volumes. The results for nanoTiO₂, nanosilver and nanoZnO are in line with ours, since they also identify recycling as a significant compartment for these NOAA. However, the importance of the compartment varies with the specific NOAA, e.g. for nanoTiO₂ the amount of material going to recycling corresponds to approximately 18% of the total production, whereas for nanosilver the proportion ca is approximately 37% (Sun et al., 2014). Overall, for the metal based NOAA investigated in the study by Sun et al. (2014), sewage treatment and primarily sewage sludge is expected to be the main EOL compartment, and from here they will most like be incinerated or applied to land. However, as is also highlighted in this study, the final concentrations of NOAA currently cannot be verified due to analytical constraints, which doesn't allow for the distinction between engineered and natural nanomaterials in the environment. Further, environmental concentrations are very dependent on production volumes and the conventional counterparts to NOAA are produced in far higher amounts, meaning that the environmental concentrations of these are expected to be one to seven orders of magnitude higher than for the NOAA

(Sun et al., 2014).

The study by Keller et al. (2013) was a first attempt to model the global environmental release of NOAA. The study combined NOAA market information and material flow modelling to assess the emissions of large production volume NOAA (e.g. silica, titania and alumina) into environmental compartments and landfills. It was estimated that 63–91 % of the global NOAA production in 2010 ended up in landfills. Waste incineration and waste water treatment were considered intermediate compartments since the authors assume that all slag and filters from waste incineration will be landfilled along with the sludge from sewage treatment plants, unless it's applied to land. Recycling is not considered in this analysis, which makes it difficult to compare with our results. The study by Keller et al. (2013) assessed the global production and disposal of NOAA by dividing the world into eight regions and extrapolating the available data for waste generation and disposal. This is different from our approach, since we try to illustrate the importance of the local waste management systems on the distribution of NOAA.

The most important findings of our study are that nanosilver is widespread across waste fractions and waste treatment options and recycling is identified as an important EOL compartment for nano-enabled products. Potential release at the EOL stage depends on the residual left in the product, which will vary according to use activities of the individual consumer and the product life time before disposal. A recent study by Wigger et al. (2015) found that up to 38% of nanosilver could remain in textiles after use (minimal release scenario). This study also investigated the influence of textile material and found that the matrix in part determines the important release point during use, e.g. for cotton, release during wearing and drying of the textile was much more significant compared to polyester. This implies that there is a need for a broad research strategy concerning EOL consumer products containing nanosilver since it may undergo a large variety of treatments ultimately affecting the environmental fate of the NOAA. In terms of number of products, recycling is still the most important compartment. This means that increasing focus should be put on the possible transformations and risks concerning nanosilver in recycling systems. Only one study was found (i.e. Caballero-Guzman et al. 2015), which looked specifically at the recycling of specific NOAA in different product categories. Here, the authors conclude that only a fraction of the NOAA in recycled products will end up going back to the production and manufacturing chain. Because most of the NOAA is associated with fractions that are not recovered in recycling processes most of them will be either incinerated or landfilled, which underlines the fact that recycling should not be seen as the final sink for these materials.

The present study provides a preliminary analysis of NOAA distribution in waste management systems. It points to an increased research effort on recycling and landfilling of nano-enabled products. But in order for our analysis to continue to be relevant, the product inventory (nanodb.dk) needs to be continuously updated both in terms of new products entering the market and products taken off the market. The usefulness of our work would also significantly improve if the mass or volume of NOAA as well as product items marketed and sold was made available in the open literature and therefore could be integrated into the flow analysis. However, this requires a lot of data from industry which is unlikely to be attainable.

Another apparent need in order improve the understanding and modelling of NOAA in waste management systems is data generation e.g. fate and behaviour of NOAA in

simulated waste treatment scenarios such landfill leachate solution or pilot scale incineration of NOAA containing products/materials. This could be achieved by performing standard waste characterization tests on nano-containing matrices and evaluate the potential release of NOAA. In this way, we would meet the need for evaluating the applicability of standard waste characterization methods to nano-enabled matrices, and possibly highlight safe by design features concerning EOL nanoproducts.

3. *Recycling*

3.1. *Process description*

Waste recycling is the reprocessing, by means of a manufacturing process, of a used material into a product, a component incorporated into a product, or a secondary (recycled) raw material (adapted from (ISO 18604:2013(en))). In this way, waste is used in the production of the same or similar products, originally composing the current waste (Christensen, 2010), thereby substituting for the use of virgin production. In several cases, because their quality is lower than the virgin ones, secondary materials can only be used in the production of different products. In this case, the term “downcycling” is typically employed. Reasons for downcycling could be multiple, for instance the:

- Presence of impurities originating from material misplacement during waste separation (e.g. a tin can is placed in the plastic bin).
- Natural worsening of the material properties (e.g. shortening of paper fibres, loss of structure when reheating thermosets plastics) due to recycling operations.
- Unwanted compounds affecting physical, chemical and mechanical properties of the material (e.g. flame retardants in plastics, alloying compounds in metals, ink in paper). When these substances are persistent and show an affinity for the solid phase, they tend to accumulate thereby hindering the further recyclability of materials (Pivnenko et al., 2015).
- Presence of hazardous substances (e.g. CCA in treated wood, PCB in construction and demolition waste) hindering recycling operations, because of their hazardousness for human health and the environment.
- Recycling operations typically involve different mechanical, physical, and chemical processes, such as sorting, washing, crushing, shredding, pulping, melting, extrusion, drying, refining, and reforming. The layout of a recycling facility varies significantly, especially depending on the feedstock material being processed. Material recycling does not include energy recovery and the use of the product as a fuel (ISO 18604:2013(en)).

3.2. *Presence of NOAA in waste recycling*

There is currently no evidence that the presence of NOAA can significantly affect recycling processes. The reasons for this may include:

- Recycling processes are in most cases include mechanical and thermal treatments while not including complex chemical transformation were the presence of NOAA could hinder the process.
- In most products analysed, NOAA are present in rather low concentrations.

However, the presence of NOAA may involve restrictions and specific safety procedures if environmental and human exposures are recognized being critical, thereby requiring changes in the management of recycling facilities.

3.3. Release of NOAA during waste recycling

Recycling processes has so far been very marginally investigated with respect to potential NOAA release. No studies have yet experimentally assessed whether and to what extent NOAA are released during recycling processes (Reihlen and Jepsen, 2015), or whether NOAA are transformed during these processes. Thus, for example, Caballero-Guzman et al. (2015) provide a probabilistic mass flow modelling of NOAA in recycling processes, where transfer coefficients are estimated based on typical distribution of bulky mass.

In the following, we provide a description of the potential factors affecting the release of NOAA during waste recycling. As recycling process are very different from each other depending on the material being processed, individual materials are presented separately.

3.3.1. Paper recycling

Paper recycling is a series of processes, where waste paper is pulped, de-inked and processed to produce new paper. The process generates solid waste (e.g. sludge) and liquid effluents that could carry a variety of contaminants, such as chlorinated phenolics, dioxins, furans and other chlorinated compounds, phosphates and suspended sediments. Both streams need further treatment and water contamination may be an issue. Air emissions (e.g. air emissions related with this process are sulphur dioxide, nitrous oxides, particulate matter, methanol, polycyclic organic matter, hydrogen chloride, formaldehyde, chloroform, phenol and chlorinated phenolics, dioxins, furans and other chlorinated compounds) may occur at different sub-processes of the recycling facility, as for example during the heat production and the chemical processing. Different APC systems are used, the complexity may be function of the size of the plant.

Pivnenko et al. (2015) recently investigated how the presence of a variety of chemicals could hinder the recyclability of paper, concluding that the two main factors are affinity for the air, aqueous or solid phase, and biodegradability (i.e. persistency). The same two factors could be employed for a first screening of NOAA present in paper to understand on their potential fate during recycling processes (Figure 4).

Despite the fact that no data is available regarding affinity of NOAA for the air, aqueous or solid phase, it could be speculated that most of the inorganic NOAA may have rather limited affinity for the air phase, meaning that they will end-up in the recycled product or in the solid and liquid by-products, thereby moving the problem to the further treatments, that could include landfilling and/or AD for the sludge, and WWTP for the liquid effluents. Regarding biodegradability, it could also be assessed that most inorganic NOAA would not be degradable (or only to a small extent), meaning that they may accumulate at each cycle.

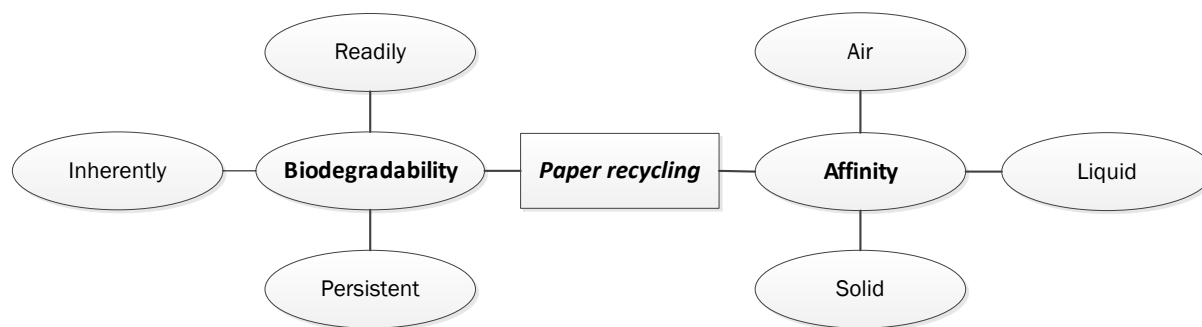


Figure 4 – Overview of factors potentially affecting the release of NOAA during paper recycling.

3.3.2. Plastic recycling

Plastics are typically classified into two main types: thermoplastics and thermosetting plastics. Thermoplastics are “the plastics that do not undergo chemical change in their composition when heated and can be moulded again and again” (Hansen et al., 2013), meaning they are to some extent more suitable for mechanical recycling. Examples of thermoplastics include polyethylene, polypropylene, polystyrene, polytetrafluoroethylene (PTFE), and polyvinyl chloride (Hansen et al., 2013).

Thermosetting plastics “can melt and take shape once; after they have solidified, they stay solid” (Hansen et al., 2013). In fact, during the thermosetting process, “a chemical reaction with the establishment of a tight crosslink between the plastic molecules occurs that is irreversible” (Hansen et al., 2013). Thermoplastics are thus less suitable for traditional mechanical recycling, while they can be reprocessed using feedstock recycling. This is however a yet not commonly employed technology, meaning that thermosetting plastics are in most cases sent for incineration in Europe.

Mechanical recycling of plastic typically comprises a series of processes such as washing, comminution, moulding, extrusion, and forming. Feedstock recycling refers to thermochemical processing (e.g. pyrolysis) of the plastic in order to degrade it to basic monomers to be used for manufacturing of new plastic.

Plastic does not only consist of plastic polymers, as during the manufacturing process a variety of compounds is added to modify/improve the mechanical performance (e.g. strength, stiffness, rigidity, elasticity) of the material, its aging properties (e.g. to prevent degradation in the environment due to UV rays, temperature, humidity, etc), its visual characteristics (e.g. colour, smoothness, shininess), the processing (e.g. to prevent the degradation of the polymer during moulding), and the economy. According to Hansen et al. (2013), plastic additives are typically divided into:

- Functional additives (stabilisers, antistatic agents, flame retardants, plasticizers, lubricants,
- slip agents, curing agents, foaming agents, biocides, etc.)
- Colorants
- Fillers (mica, talc, kaolin, clay, calcium carbonate, barium sulphate)
- Reinforcements (e.g. glass fibres, carbon fibres).

During manufacturing, use and disposal of plastic materials, different compounds contained could migrate from the matrix and eventually be released. Potential for release of different substances from plastics are seldom available (Hansen et al., 2013). Factors generally affecting the migration include the substances’ size, their

concentration, their boiling point, the vapour pressure and their solubility in both the plastic and the environment (Hansen et al., 2013). Small molecules have a low boiling point and tend to migrate faster, while large organic molecules have a tendency to migrate more slowly, while inorganic metal salts do not migrate. The age of the plastic material also plays a role, as plastic chains get shorter with age, thereby inducing a higher release.

During recycling operations, thermomechanical processing such as moulding and extrusion are used to melt the plastic to allow re-palletisation. The melting temperature of the polymer together with the melting/boiling point of substances could provide an indication about how prone NOAA are to be released to the gas phase. An overview of the melting temperature of different types of plastic is presented in Table 4. These temperatures are far below the boiling temperature of many inorganic NOAA (see section 4.3.2), thereby suggesting that release of NOAA to the gas phase should be rather unlikely. Other types of plastics (Table 5) may only be suitable for feedstock recycling, which would typically occur at much higher temperatures. In this case, consideration similar to those for waste incineration (see e.g. section 4.3.2) should be made. Table 4 finally shows that plastic recycling is not 100 % efficient, as some by-products are typically generated, originating either from impurities present in the waste or plastic residues. These materials will hence end-up in other treatment processes, such as incineration and landfilling.

Based on the considerations for different compounds and additives found in plastics, we identified a number of factors potentially affecting the migration and release of NOAA during recycling processes, as shown in Figure 5.

Table 4 – Overview of types of plastics typically recycled or potentially recyclable (adapted from Hansen et al. (2013)).

Type of polymer	Acronym	Melting T (°C)	Recycling rate (%)
Polyethylene	PE-LD/PE-LLD	125	92-93
Polypropylene	PP	165	90
Polystyrene	PS/HIPS	~240	90
Expanded Polystyrene	EPS/XPS	~240	>90
Acrylonitrile butadiene styrene	ABS	105	90
Polyvinylchloride	PVC	160	97.5 (rigid PVC) >90 (Soft PVC)
Polyester	PET	265	92-93
Polymethyl methacrylate	PMMA	160	>90
Polycarbonate	PC	155	>90
Polyamide	PA	233-272	>90

Table 5 – Overview of types of plastics typically not-recycled or potentially recyclable only via feedstock recycling (adapted from Hansen et al. (2013)).

Type of polymer	Acronym	Recycling
Polytetrafluoroethylene	PTFE	Possible, but normally not practiced (used as coating material)
Polyurethane	PUR	Feedstock
Epoxy		Feedstock

Unsaturated polyester	UP	Feedstock
Melamine	MF	Feedstock

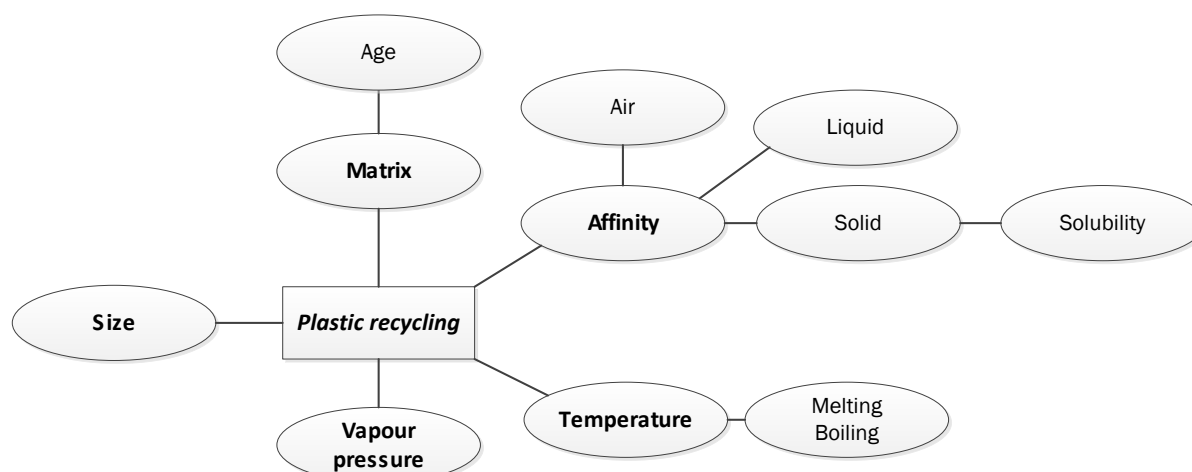


Figure 5 – Overview of factors potentially affecting the release of NOAA during plastic recycling.

3.3.3. Glass recycling

Glass waste can be recycled in different ways. When reused or used as inert aggregate, release of NOAA could occur in a similar way as for the initial products through abrasion and release to water phase, i.e. similarly to the user phase.

A main recycling route though includes the remelting of glass for the production of new glass products. The remelting occurs at high temperatures (e.g. 1400-1600 °C), which could induce evaporation of NOAA and subsequent release through exhaust gases. The magnitude of this phenomenon will depend on the boiling/melting point of the individual types of NOAA, which can be rather different. This aspect is further elaborated in section 4.3.2 with respect to waste incineration.

3.3.4. Metal recycling

Recycling of metal waste includes several different processing methodologies, depending on the specific metal in question. A common feature of most process is that metals may be processed (e.g. remelted) in high temperature ovens, the temperature depending on the metal. During this operation, evaporation of NOAA and airborne release may occur. As mentioned for glass recycling, the magnitude of this phenomenon will depend on the boiling/melting point of the individual types of NOAA, which can be rather different. This aspect is further elaborated in section 4.3.2 with respect to waste incineration. It should also be mentioned that most modern metal smelting facilities are equipped with advance flue gas cleaning systems, which may retain heavy metals and NOAA to a certain extent. The effectiveness of flue gas cleaning systems is further evaluated in section 4.3.3 with respect to waste incineration.

3.3.5. C&D

Construction and demolition (C&D) waste is generated during the building, repair, remodeling or removal of constructions such as roads, residential housing and nonresidential buildings. C&D waste includes different waste materials, such hard materials (i.e. concrete, tiles, mortar), wood, paper/cardboard, plastic, metals, other non-combustible.

Concrete, tiles, mortar represent the majority of building waste. After screening,

crushing and sieving, concrete and tiles are down-cycled into aggregates, potentially used as subbase for roads (unbound, main application), aggregates in concrete and asphalt, backfilling of excavations, filler in asphalt concrete.

Release of NOAA in connection to C&D waste recycling may occur through two main routes. Firstly, our experiments in task 3.4 (reported in D3.5) have shown that when hard materials such as concrete and tiles are crushed, significant amounts of fine and ultrafine particles are released to air, and non-negligible amounts of NOAA may become airborne. This may suggest that when NOAA are present in C&D waste, recycling operations may be conducted in enclosed environments making use of filtering systems for exhaust air.

Secondly, NOAA in C&D waste may be released from aggregates once these materials come in contact with water and leachate is formed. Factors affecting release of NOAA into the water phase at this stage may be similar to those listed for landfills in section 5.

3.3.6. Overall recycling

The main aspects to be considered when assessing potential release from recycling processes are summarized as follows:

- Hardness of the matrix. With hard material, there is a significant risk of dust formation and airborne release of NOAA in connection to shredding and size reduction steps.
- Temperature reached during the recycling processes: high T may induce evaporation of NOAA depending on the melting/boiling point of individual NOAA types.
- Affinity of NOAA towards:
 - Air phase: there is a risk of release in all cases
 - Solid phase: NOAA will tend to accumulate at every cycle of the material.
 - Liquid phase: risk of release in those processes where water and/or other liquids are used for remanufacturing of waste as well as in those cases where the secondary materials are employed in applications where the material may come into contact with water.

4. Incineration

4.1. Process description

Incineration is the (full or partial) thermal conversion of waste, operated with a surplus of air and leading to dramatic changes in the chemical and physical characteristics of the waste (Christensen, 2010). Incineration is a high-temperature process, where the combustion temperature is typically above 1000 °C in the central part of the combustion chamber¹.

¹ The flue gas temperature theoretically obtained upon complete combustion of the waste (i.e. the adiabatic temperature) is determined from mainly the heating value (HV) of waste, the air-preheating temperature and excess air level. Depending on the furnace design, a certain temperature drop is caused by heat transfer to the walls. To maintain the required 2 sec residence time at min. 850 °C in the afterburning chamber (i.e. after the last air-injection), the combustion temperature should be above 1000 °C in the combustion chamber above the grate. If the HV drops, air pre-heating must be increased to maintain the combustion temperature. When the HV drops so that 850 °C cannot be maintained with maximum air pre-heating, then a support burner must be used, else the waste firing must stop. There is a number of exceptions to the abovementioned, as the 1000 °C temperature is possibly not reached everywhere on the grate – particularly not on the first sections, where drying of waste occurs, and also not on the last sections where the slag is cooled. Also, furnaces designed for low CV-waste with extensive insulating bricks may not always maintain 1000 °C, whereas in a fluidized bed incinerator, the temperature is fairly uniform and stabilized by the sand, meaning that the temperature would be in the range roughly 850-950 °C.

A modern incineration plant – often called waste-to-energy facility when extensive energy recovery is employed – comprises several components (see Figure 6 and Figure 7), serving different purposes. Waste delivered to the plant is stored in a bunker until it is fed to the furnace, which could be based on different combustion technologies (e.g. moving grate, rotary kiln, fluidized bed). The following afterburning zone (also called secondary combustion chamber) is designed to ensure that all processes/reactions are completed. The combustion is supported with supply of primary (in the furnace) and secondary (in the afterburning zone) air. The reaction between the combustible components of the waste and the oxygen of the combustion air generates significant amounts of hot combustion gas (called flue gas), which contains high concentrations of several problematic substances (e.g. dust, heavy metals, acid gases, NO_x, organic pollutants, PCDD/F) that need to be removed before the flue gas is finally released to the atmosphere thorough the stack. The solid residue originating from the incombustible part of the waste is collected from the bottom of the furnace, and is thus called bottom ash. Having aggregate properties similar to those of natural aggregates, bottom ash can be used in different applications within the construction industry.

Before entering the air pollution control (APC) system, the hot flue gas must be cooled down. In modern plants, this is done in an energy recovery system, where the energy contained in the flue gas is used for energy production, e.g. electricity and heat. The following air pollution control (APC) or flue gas cleaning (FGC) system is system/set of technologies aiming at reducing target pollutants from the levels found in the raw flue gas down to acceptable concentration levels set by legislation. Typical components of an APC system include dust/heavy metals collection (e.g. electrostatic precipitators, fabric filters, bag house filters, cyclones, Venturi scrubbers), acid gas neutralization (e.g. wet/dry systems), PCDD/F abatement (e.g. activated carbon), NO_x reduction (e.g. selective catalytic or non-catalytic). Operation of the APC system generates different solid and liquid residues that need further handling, treatment, and final disposal.

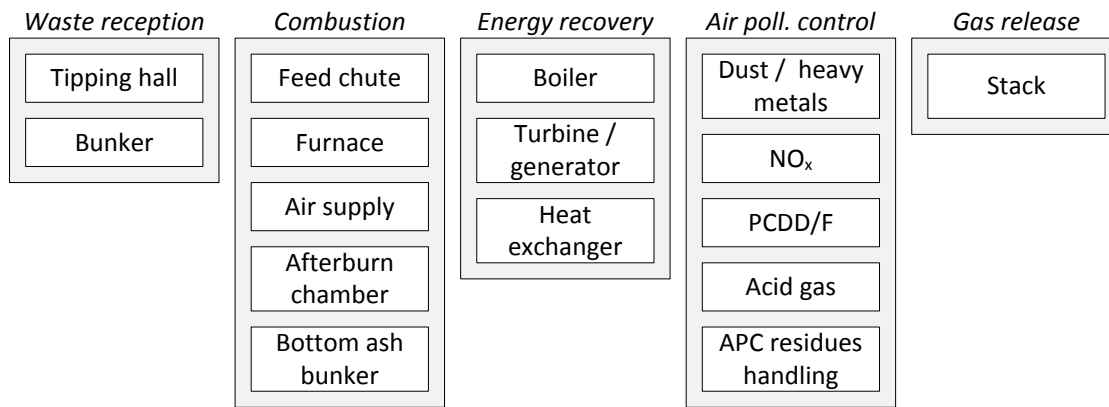


Figure 6 – Schematic overview of components of a waste incineration plant.

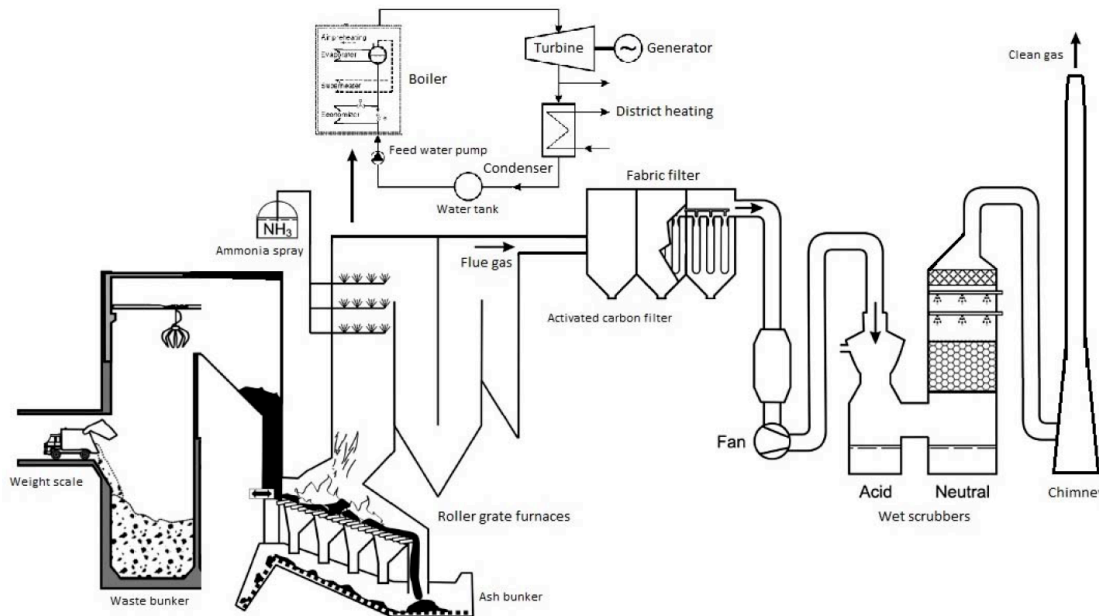


Figure 7 – Illustrative layout of an incineration plant with a moving grate furnace (adapted from Christensen (2010)).

4.2. Presence of NOAA in waste incineration

Within the current knowledge and considering the concentration and amounts of NOAA potentially delivered to waste incineration facilities, it is considered very unlikely that NOAA could have any effect on the combustion process as such. The presence of NOAA may however have some effect on the chemistry of the post-combustion, as described in the following.

4.3. Release of NOAA during waste incineration

Experimental measurements of NOAA release from up- or full-scale waste incineration facilities were so far only attempted twice, on CeO and TiO₂. The results from the two tests were quite similar, and can be summarized as follows (Reihlen and Jepsen, 2015):

- emissions on NOAA from the stack are not increased when nanoproducts are added to the waste feedstock;
- most of NOAA contained in the nanowaste end up in the bottom ash;
- a small, non-insignificant share of NOAA is routed into fly ash;

These results cannot however be extrapolated to all NOAA and all types of installations, as conditions may differ significantly.

According to Roes et al. (2012), NOAA could follow four different paths one entering an incineration plant:

1. NOAA that are present in nanowaste are destroyed in the grate furnace due to combustion.
2. NOAA are not destroyed in the grate furnace, and either
 - end up in bottom ash; or
 - move with the flue gas, are captured in the APC system and leave the system with the fly ash.
3. NOAA are destroyed (e.g. by oxidation or reaction with HCl) but form new NOAA (e.g. oxides, chlorides).
4. New NOAA are formed as a result of NOAA decomposition.

An overview of important aspects that could affect the release of NOAA in connection with nanowaste incineration is provided in Figure 8. Individual aspect will be described and discussed in details in the following sections.

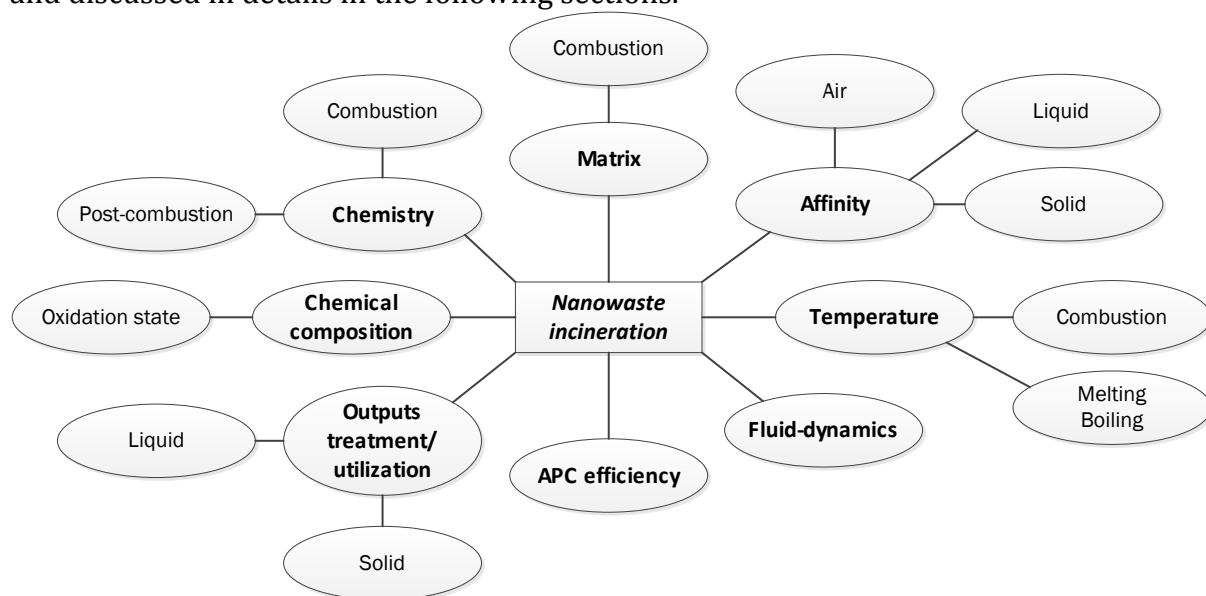


Figure 8 – Overview of factors potentially affecting the release of NOAA in nanowaste incineration.

4.3.1. Waste matrix and nanostructure

The composition of the waste matrix and the nanostructure (i.e. how NOAA are placed in the matrix) are determinant aspects for NOAA release during combustion. If the matrix is not-combustible and the NOAA are embedded in the solid, the NOAA will most likely end up into the bottom ash, as almost no NOAA have a chance to be liberated from the matrix. On the other hand, if the matrix is combustible, NOAA have higher chances of being liberated and end up in the gas phases.

4.3.2. Combustion temperature and melting/boiling points

The temperature of the combustion process plays an important role in relation to the melting and boiling point of individual NOAA, as this is likely to affect the distribution of NOAA between the solid and gaseous phases, or vanish due to complete burnout (Mueller et al., 2013). When the melting or boiling points of NOAA are lower than the combustion temperature, NOAA are more likely to be destroyed by melting and burning, while a higher melting or boiling point mean that less NOAA is destroyed. As an example, ZnO is reported to remain solid up to a temperature of 1500°C (i.e. the melting point), hence the major part of ZnO (37-86%) is found in the bottom ash

(Mueller et al., 2013).

When the surrounding temperature is between melting point and boiling point substances (but also NOAA) tend to vaporize and thus enter the flue gas stream (Mueller et al., 2013). Figure 9 compares the melting and boiling point of different bulk counterpart of many NOAA along with the minimum temperature of 850°C given by EU legislation and the actual combustion temperature. Silver NOAA are expected to enter the gas phase, because the melting point of silver is lower than the actual combustion temperature, while the boiling point is higher than actual combustion temperature Figure 9. Conversely, CNTs are expected to undergo complete burnout, because both the melting and boiling point is below both the actual combustion and the required temperature. On the contrary, TiO_2 has a boiling point of 2900°C (Mueller et al., 2013), while the melting point is 1843°C (AzoNano, 2013), which means that the nano TiO_2 is expected to distribute to the bottom ash. The melting points of ZnO and CeO_2 are found to be 1500°C (Mueller et al. 2013) and 2500°C (Walser et al., 2012), respectively, while boiling points could not be found in the literature. Information on the boiling point will however not change the expected distribution, as the melting points are already higher than the actual combustion temperature, meaning that ZnO and CeO_2 are expected to distribute to bottom ash.

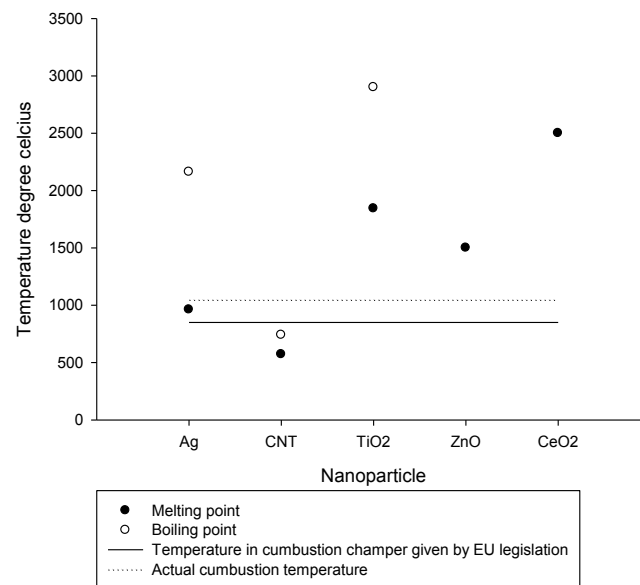


Figure 9 - Melting and boiling point of different bulk materials (AzoNano, 2013; Mueller et al., 2013; Walser et al., 2012).

Although, the boiling and melting points of the bulk counterpart may provide an indication of the possible distribution of NOAA into different compartments, it was shown that melting points of NOAA are size dependent. Generally, it seems that nanosized materials have a lower melting point compared to their bulk counterparts, and smaller sized NM tend to have lower melting points compared to larger sizes. Melting points of different NOAA at different sizes are shown in Figure 10, together with the minimum legal temperature (to be ensured during the combustion according to EU law) and the actual combustion temperature. Compared with the bulky counterparts (Figure 9), most melting points of NOAA are well below both the actual combustion temperature and the lowest incineration temperature, meaning that vaporization may be more significant.

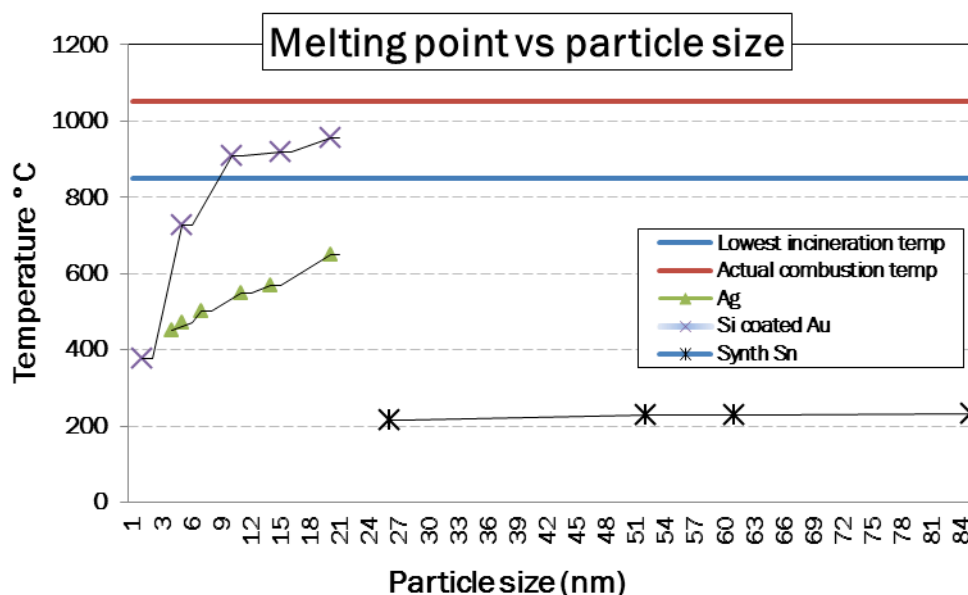


Figure 10 - Size dependant melting points of different NOAA (based on (Asoro et al., 2009; Dick et al., 2002; Jiang et al., 2006)).

4.3.3. Efficiency of the air pollution control (APC) system

The efficiency of the APC system plays a key role in removing the NOAA which are released to the gas phase during the combustion process. The choice of APC systems (cyclone, fabric filter, electrostatic precipitator (ESP), wet or dry venture scrubber, active carbon) differs from plant to plant, and thus the environmental exposure depends on which systems are present in each plant. The efficiency of the different APC systems depends on the size of the NOAA (see Table 6), while the electrostatic precipitator (ESP) and wet scrubber are not substance specific (Mueller et al., 2013), hence it is possible that the efficiency is as high for other NOAA than for nanoCeO₂. One study has studied the efficiency of ESPs on NOAA (Huang and Chen, 2002). Various sizes of particles both in nanosize and larger were tested in single- or two-stage ESPs, showing significant penetration of aerosol. In general, the single-stage ESP showed decreasing penetration of NOAA with decreasing NOAA size until a particle diameter of 15 nm, where the penetration would increase again. For two-stage ESPs, an increase in aerosol collection was seen around 50 nm. The authors suggest these findings of efficiency at small sizes to be due to (i) higher electrical potential in the ESP than the applied voltage because of high partial space charge density, (ii) a strong ionic flow could make particles, especially the ultrafine particles, to move toward the collecting plates and subsequently trapped because of a secondary flow of ions, or (iii) turbulence that increase the change for

particles to come in contact with the wires of the ESP. A part of particle diameter, the efficiency of ESP also depended on voltage and airflow through the ESP, with increased voltage leading to increased efficiency, and decreased air flow resulted in decreased aerosol penetration. The result of the study shows, that ESPs can be used for collection of NOAA in the APC system, but the efficiency is highly dependent on particle size, air flow rate, and applied voltage.

Table 6 - Reported NOAA removal rates in different parts of the APC system of an incineration plant.

Scenario	Removal technique	NOAA type	NOAA size	Efficiency	Reference
	Fabric filters		<100 nm	<80 %	Roes et al. (2012)
	ESP		3-10 nm		Roes et al. (2012)
	Wet scrubbers		100 nm	65%	Roes et al. (2012)
	Wet scrubbers		50 nm	50%	Roes et al. (2012)
Full scale	ESP	CeO ₂	80 nm	99.995%	Walser et al. (2012)
Full scale	Wet scrubber	CeO ₂	80 nm	99.9%	Walser et al. (2012)
Full scale	ESP	CeO ₂	80 nm	99.995%	Walser et al. (2012)

ESP: electrostatic precipitator

4.3.4. Residue management and exposure potential from ash and gas

Depending on the melting point and the boiling point of the specific NOAA, the NOAA can either be completely burned out, or end up in the gas or in the ash. If the NOAA is completely burn out during incineration, the potential for environmental exposure is none. If the NOAA is instead routed into the bottom ash or the solid/liquid residues from the APC system, the exposure will depend on the further management of these residues. These other solid residues (e.g. fly ash, slags) may undergo other chemical processing or final disposal in landfills and/or underground deposits; the potential for release is however not addressed in available literature. Liquid effluents (e.g. from scrubbers) may be routed to wastewater treatment plants (WWTP) and NOAA may end up in WWTP sludge.

If the combustion temperature is lower than the melting point of the NOAA, the NOAA may enter ash phase. The ash from incineration plants can either be deposited by special waste management (i.e. containing in big bags in indoor storage facilities) (Flyvbjerg and Hjelmar, 1997), landfilled (Hjelmar, O. & van der Sloot, 2010) or reused in construction work (e.g. road sub-base) (Danish Ministry of the Environment, 2010). The final disposal may in some cases be associated with long-term NOAA release as a consequence of rainfall exposure (Reihlen and Jepsen, 2015). This is especially the case with untreated residues, from which salts and heavy metals may be released and hence pose a significant risk of pollution to surface- and groundwater mobility for many years (Flyvbjerg and Hjelmar, 1997). Therefore, the bottom ash matrix should be treated before these actions of reuse, deposit, or landfilling to avoid leaching of NOAA and chemicals. The treatment can be solidification into i.e. cement, lime, or asphalt, vitrification where the slag matrix is changed into glassy matrix by heating (1400°C), or sintering where the residues are heated (900°C) which result in a denser matrix with less porosity (Roes et al., 2012). No studies have yet assessed the potential release of NOAA from bottom ash (Reihlen and Jepsen, 2015).

In addition to the above, bottom ash is often mechanically processed to recovery metal fractions and/or to achieve desired material properties (e.g. grain size). This mechanical treatment could certainly have a significant influence on the release pathways, similarly to what previous discussed with respect to recycling processes.

4.3.5. Chemical composition, size and oxidation state of NOAA

The type, chemical composition and size of NOAA entering the combustion chamber is a key aspect determining the fate of the NOAA. For example, if temperature is high enough, reduced particles (e.g. Al) may undergo combustion to an extent that depend on their size and aggregation state (Holder et al., 2013). Conversely, particles that are already oxidized and have high melting points (e.g. CeO_2) may exit the combustion zone essentially unchanged. Moreover, NOAA can be engineered to have certain properties, e.g. to be ionised. If the engineered property is present when the NOAA enters the APC systems, the NOAA will might not be trapped in the APC system.

4.3.6. Chemistry of combustion and post-combustion

Besides to the combustion of individual NOAA, another important aspect is the chemistry between NOAA and other substances present in the flue gas, as this may affect the transformation processes during and after combustion. NOAA could catalyse the formation of other pollutants (Reihlen and Jepsen, 2015). An examples is the de-novo formation of PCDD/F, which is known being catalysed by Cu and Zn. Another rimportant aspect is the aggregation of NOAA right after the combustion process, as this will affect the removal efficiency in the APC system (Holder et al., 2013), because it both affect the size and the physicochemical properties of the NOAA.

5. Landfilling

5.1. Process description

Landfilling can be defined as the dedicated use of land for disposing waste in an engineered facility (Christensen, 2010). The term landfilling includes a broad variety of technologies, ranging from open and uncontrolled dumps to modern and highly engineered facilities making use of advanced control and monitoring systems. A landfill can be seen as a complex reactor, where the physical and chemical processes involved in waste degradation can last for very long time (up to millennia for some materials).

The two main products or emissions of a landfill are leachate and gas. The amount/composition of generated leachate/gas and the conditions present in the body of the landfill are strongly interdependent: the amount/composition change over time as a result of the changing conditions in the landfill body, and vice versa (Table 7).

Leachate is produced by the infiltrating rain that seeps through the waste. Its formation rate is controlled by many factors, including climatic and hydrologic factors (i.e. rainfall, snow melt, groundwater intrusion), landfill design (i.e. vegetation, area, cover, sidewalls, liner material), operation and management (i.e. waste pre-treatment, compaction, irrigation, recirculation, liquid waste co-disposal), waste characteristics (i.e. refuse settlement, age, particle size, density, moisture content), and internal landfill processes (i.e. refuse settlement, organic matter decomposition, gas and heat generation, transport). Leachate contains extracted solutes, suspended solids and any other component of the material that are transferred (or released) from the solid phase to the liquid phase. The release of a given substance from a solid (waste) phase to a contacting

liquid phase involves both liquid/solid phase reactions and interactions and transport within the solid phase as well as in the liquid phase (Christensen, 2010). The release of compounds is typically controlled by the availability – i.e. the amount released is limited by the amount available in the solid matrix –, or by the solubility – i.e. the amount that can be solubilized in a given amount of water is the limiting factor –, or by a combination thereof. An important parameter determining the release is the so-called liquid-to-solid (L/S) ratio, which is the ratio between the cumulated amount of water (L) that at any given time has percolated through a system and the dry weight (S) of the material being percolated.

Landfill gas is mostly constituted of CH₄ and CO₂, and is the result of the anaerobic degradation of the organic part of the landfilled waste. The amount of landfill gas generated depends on the amount of organic matter in the waste, while the generation rate is controlled by the progressing of anaerobic processes. The anaerobic degradation is in turn affected/determined by a number of factors, including water content of the waste, temperature, oxygen, hydrogen, sulphate, nutrients availability, pH and alkalinity (pH 6–8 is preferred), and presence of inhibitors.

Table 7 – Overview of factors determining the amount/composition of leachate/gas generated in a landfill.

Leachate		Gas	
Composition	Amount	Composition	Amount
<ul style="list-style-type: none"> •waste composition •availability •solubility •landfill conditions (e.g. pH) 	<ul style="list-style-type: none"> •climatic and hydrologic factors (rainfall, snow melt, groundwater intrusion) •landfill design: vegetation, area, cover, sidewalls, liner material •operation and management: waste pre-treatment, compaction, irrigation, recirculation, liquid waste co-disposal •waste characteristics: refuse settlement, age, particle size, density, moisture content •internal landfill processes: refuse settlement, organic matter decomposition, gas and heat generation, transport 	<ul style="list-style-type: none"> •Water content •Temperature •Oxygen •Hydrogen •Sulphate •Nutrients •pH/alkalinity •Presence of inhibitors 	<ul style="list-style-type: none"> •waste composition •landfill operation: shredding, compaction, soil cover, recirculation of leachate, precomposting of bottom layer

Landfills are associated with a range of potential environmental impacts, related to emissions gas and leachate to air, soil and water compartment. These potential environmental impacts can have effects on a local (e.g. odor, noise, birds, rodents, insects, litter, dust, fire, vegetation damage, soil pollution), regional (toxic gases, surface water pollution, groundwater pollution), and global (e.g. greenhouse effect, ozone depletion) scale (Christensen, 2010). Because of the long lasting degradation processes, the potential for environmental exposure from may as well extend for long time frames, unless initiative are taken to quickly stabilize the waste in the landfill. In modern landfill, environmental release of gas and leachate are mitigated using different technological approaches, including bottom liners, leachate drainage, top covers, gas collection systems, and by placing that landfill as far as possible from sensitive environmental

receptors.

5.2. Presence of NOAA in landfills

Due to their inner nature, NOAA may have toxic effect with respect to the degradation processes occurring in landfills. For example, nanosilver which is the most prominent NOAA in waste (see section 2.2); it is well known that, for instance, nanosilver has biocidal effect. However, Bolyard et al. (2013) reported that nanoZnO and nanoTiO₂ had no inhibitory effect on aerobic and anaerobic landfill processes, thus possibly suggesting that, because of dispersion, the concentration of NOAA may be too low to have any significant effect. More investigation may however be needed before any solid conclusion could be drawn.

5.3. Release of NOAA from landfills

The behaviour of NOAA in landfills and the exposure potentials after landfilling is influenced by a range of different factors and processes as well as the actual NOAA being considered e.g. metals, metal oxides, carbonaceous NOAA. In general, it seems clear that NOAA have potential to diffuse to landfill gas, but the diffusion is regarded as less important when quantitatively compared to leachate and waste matrix, because the NOAA first has to be released from the product and not bind to the waste matrix or leachate if it should be a subject of diffusion to gas.

An overview of important aspects that could affect the release of NOAA in connection with nanowaste landfilling is provided in Figure 11. Individual aspects will be described and discussed in details in the following sections.

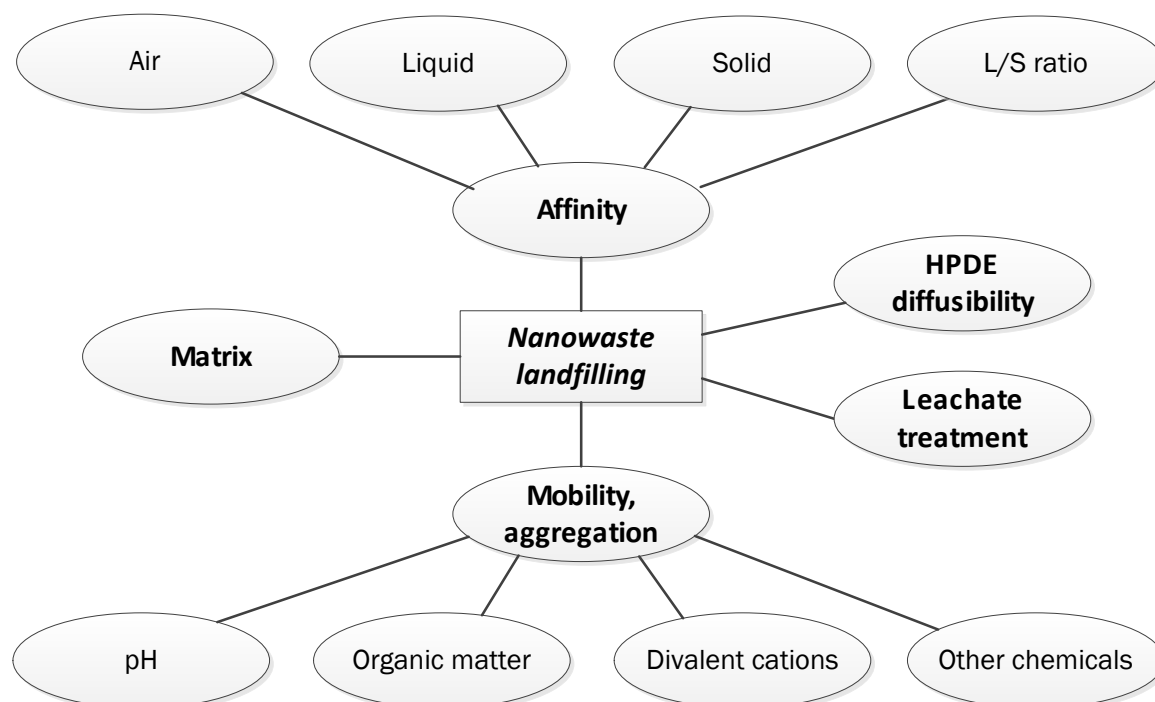


Figure 11 – Overview of factors potentially affecting the release of NOAA in nanowaste landfilling.

5.3.1. Product matrix, nanostructure and affinity to air, liquid, solid phases

The location of the NOAA in the product is a contributing factor for determining where

the NOAA can be found in the landfill (Reinhart et al., 2010), while the matrix material affects the release pattern over time, depending on its degradability. Surface bound NOAA are assumed to be readily available for landfill processes, because the product surface is in contact with the landfill surrounding, and therefore do not need first to be released from the product. It is assumed that NOAA suspended in liquids will settle to surfaces of other wastes in the landfill when the liquid has evaporated, and then be treated as surface bound particles. For NOAA suspended in solids, the mobility of the particles depends on a potential release from the product.

Once the location of NPs has been identified, the mobility has to be determined. No models or measurements are available for the behaviour and fate of NOAA in landfills. The literature is scarce as well for other well studied organic chemicals, but one available model is the mathematical mass flow model “Model for Organic Chemicals in Landfills” (MOCLA) (Kjeldsen and Christensen, 2001), which describes transmission products of organic chemicals and their emission and distribution to leachate, gas phase and waste matrix. The MOCLA model is used as an inspiration to describe the mobility of NOAA in landfill waste matrix. NOAA that do not sorb to the waste matrix will instead enter the leachate phase. The MOCLA model determines distribution of organic chemicals in landfills to air, soil, and water compartments and uses the equation $f_w = (\varepsilon \cdot C_w) / C_t$, where f_w is the fraction of chemical in water phase, ε is the volumetric content of water in landfill, C_w is the concentration of chemical in water phase, and C_t is the total concentration of chemical in the landfill) to describe distribution of the organic chemical to water phase.

Bjerg & Kjeldsen (2004) described the sorption affinity of compounds in a two-phase system, solid and water phase. Waste matrix and leachate from a landfill can be regarded as phases similar to solid and water. This can be used for NOAA, so the sorption of NOAA is described as the fraction of NOAA in leachate, which depends on the dry weight of a volume of waste matrix and the porosity of the waste matrix, and the partition coefficient of the NOAA. When saturated with water or leachate, the fraction can be expressed as $f_{\text{leachate}} = 1 / (1 + \rho / \Phi \cdot K_d)$, where f_{leachate} is the fraction of NOAA in leachate, ρ is the dry weight of waste matrix (kg/L), Φ is the porosity, and K_d is the partition coefficient of NOAA (L/kg). From this equation, it is possible to determine whether NOAA are mobile or if they rather sorb to the waste matrix in a landfill. For this purpose, the partition coefficient should be found in existing literature for experimental values.

In the MOCLA model, some scenarios on landfill parameters are provided, hereby illustratively used. The porosity is 0.4 (unit less) and the dry weight is stated to be 0.6 t/m³ (equals 0.6 kg/L) for landfill waste matrix (Kjeldsen and Christensen, 2001). It is important only to use experimental partition coefficients when the distribution to leachate is examined. For organic chemicals, the K_d value can be estimated from the equation $K_d = K_{oc} \cdot f_{oc}$, where K_{oc} is the distribution coefficient for a chemical between octanol and carbon phase (Kjeldsen and Christensen, 2001). However, the K_{ow} value is not useful to describe the partition between carbon and water. This is also applicable for K_{oc} . If no experimental K_d value can be obtained for a NOAA, the mobility cannot be predictively described by the NRCW framework.

In our evaluation, we used a fraction of 1% or more of NOAA that distribute to leachate and 99% or more is considered to be of relevance to further investigation of the

exposure potential of NOAA from both leachate and waste matrix, and a cut-off value of 99% where only leachate should be examined.

5.3.2. Mobility and aggregation

Different sources indicated that different factors could affect the mobility and aggregation of NOAA once they are in the liquid phase:

- The pH of the leachate (Andersen et al., 2014)
- The present and concentration of organic matter in leachate (e.g. (Lozano and Berge, 2012))
- The presence of divalent cations (e.g. Ca^{2+})
- The presence of other chemical that could interact with NOAA

In general, initial speculations indicate that typical landfill conditions – where organic matter and divalent cations (e.g. Ca^{2+}) are present – could enhance agglomeration and reduce the mobility of NOAA in leachate (Andersen et al., 2014).

5.3.3. Treatment of leachate

In low-tech landfills, the leachate generated as a consequence of water infiltration into the landfill body is not collected nor treated, meaning that leachate will infiltrate into the ground, where soil will be exposed to the presence of NOAA.

In more advanced landfills, the leachate may be collected and undergo further treatment. Leachate treatments include i.e. stripping, membrane, or biological processes, evaporation, membrane, adsorption, chemical oxidation, wetland, nitrification, and denitrification. Municipal treated or on-site treated landfill leachate is considered as wastewater by e.g. the Danish Law, and can be treated in public or private wastewater treatment plants (Danish Environmental Protection Agency, 2000). The products of sludge treatment in e.g. a wastewater treatment plant are sludge and “clean” waste water. The environmental exposure potentials of these depend on further management of the sludge or waste water. Water from wastewater treatment plants can be infiltrated to ground, discharged into lakes, rivers or sea, or sprayed on soil surfaces (Danish Ministry of the Environment, 2007), while the sludge could be incinerated or applied on land.

6. *NanoRiskCatWaste*

From the above, it is clear that a framework is needed that enables users to describe the exposure potentials of NOAA during and after the most common solid waste handling procedures i.e. recycling, incineration and landfilling. In the following, we will briefly describe NanoRiskCatWaste, a framework that we are developing to address this need. Outset is taken in solid waste stemming from consumer products that contain NOAA and the relevant physical and chemical properties of NOAA that contribute to the fate of the particles in waste management systems, most of which were described in previous sections. To evaluate the fate and exposure potentials in WMOs, roadmaps for each management system were developed. Roadmaps will be used as a basis for developing guidelines on safe-by-design, where relevant parameters/factors to be considered during the design phase to prevent adverse consequences during the waste management phase will be identified.

6.1. NanoRiskCat and NanoRiskCatWaste – an introduction

In light of the increasing concern and growing scientific literature on the adverse environmental health and safety aspect of NOAA, we previously developed a systematic tool called NanoRiskCat that enables users to communicate “what they know about the hazard and exposure potential of consumer products containing engineered NOAA. The final outcome of NanoRiskCat is communicated in the form of a short-title describing the intended use and five coloured dots. The first three dots refer to the qualitative exposure potential for professional end-users, consumers and the environment, whereas the last two refers to the hazard potential for humans and the environment. Each dot can be assigned one of four different colours: red, yellow, green, and grey indicating “high, medium, low, and unknown” respectively. An important aspect that is missing in NanoRiskCat is considerations about what happens with NOAA in solid waste and waste management operations and how this translates into an evaluation of the potential exposure of NOAA from a product in waste treatment systems to environmental compartments.

From the previous chapters, it is clear that exposure of NOAA originated from a consumer product that has become waste depends on three factors: the waste management option (WMO), the location of the NOAA in the product and physical-chemical characteristics of the NOAA. Taking this into consideration, we have developed a preliminary waste module that could potentially be added to the NanoRiskCat framework with the aim of describing the potential environmental exposure of NOAA stemming from solid waste. The NanoRiskCatWaste (NRCW) framework consists of a series of road-maps of NOAA fate during waste treatment and was applied to representative case materials. It is important to realize that only environmental exposure of NOAA from solid waste from consumer products was considered here. Hazardous waste was not covered although a potential exists for a households to dispose products with NOAA as hazardous waste (i.e. paint) because hazardous waste requires different treatment than non-hazardous waste. Recycling, as a waste treatment option, will be addressed at a later stage of the project.

6.2. Nanowaste roadmaps for incineration

The NRCW roadmap for nanowaste incineration is based on the following three overall basic assumptions regarding NOAA distribution in an incineration plant:

1. $T_{inc} < T_{melt}$ of NOAA \rightarrow NMs will enter ash phase
2. T_{melt} of NOAA $< T_{inc} < T_{boil}$ of NOAA \rightarrow NMs will enter gas phase
3. T_{boil} of NOAA $> T_{inc} \rightarrow$ NMs will undergo complete burnout

These three basic assumptions are based on the work by Mueller et al. (2013) who have utilized melting point and boiling point as input parameters in their model of NOAA flows during waste management.

The NRCW roadmap for incineration is shown in Figure 12. When applying the NRCW roadmap on a consumer product, the first decision is to determine the location of the NOAA in the product. Depending on the location, the NOAA will either be transformed due to heating, or distribute directly to bottom ash, if the NOAA is suspended in solids. The transformation of the NOAA is determined by the melting and boiling points, and the incineration temperature. The transformation results in a distribution to either bottom ash or flue gas, or a complete burnout of the NOAA. Residue management of

NOAA distributed to bottom ash or flue gas continues in Figure 13 and Figure 14. The orange box indicates that the NOAA is no longer in solid state and is no longer a NOAA but ions.

6.2.1. NRCW for flue gas

The roadmap for flue gas cleaning and management of APC residues is shown in Figure 13. The exposure potential of NOAA in air phase after treatment depends on the efficiency of the treatment. The solid fraction from cleaned flue gas can be reused, landfilled or solidified. The liquid fraction from wet venture scrubber can be treated in waste water treatment plants.

In general, we would argue the potential exposure to air is considered low if the cleaning system removes more than 99% of the NOAA. The 99% removal efficiency is similar to the needed efficiency of HCl, fly ash, Cd, Hg, and SO₂ after incineration and gas cleaning set by the European Union (Vehlow and Dalager, 2010). We would categorize a removal efficiency between 90-99% as medium and high, if less than 90% of the NOAA are removed from the flue gas by APC system. The APC removal efficiency influences the exposure potential for soil and water. If the APC system is very efficient (>99%), potential exposure to soil and water is considered low. If the potential exposure to air is medium, the potential exposure to soil and surface and groundwater is expected to be low, and medium if the APC system removes 90% or less of the NOAA. These potentials are based on the precipitation that the NOAA which is emitted to air must bound to soil or settle to the surface of water before it can have an exposure potential. While a nano-specific APC system is not developed and used in incineration plants yet, there is a potential that such cleansers can be developed if studies shows the need for it. Therefore the nano-specific cleanser is included in the framework.

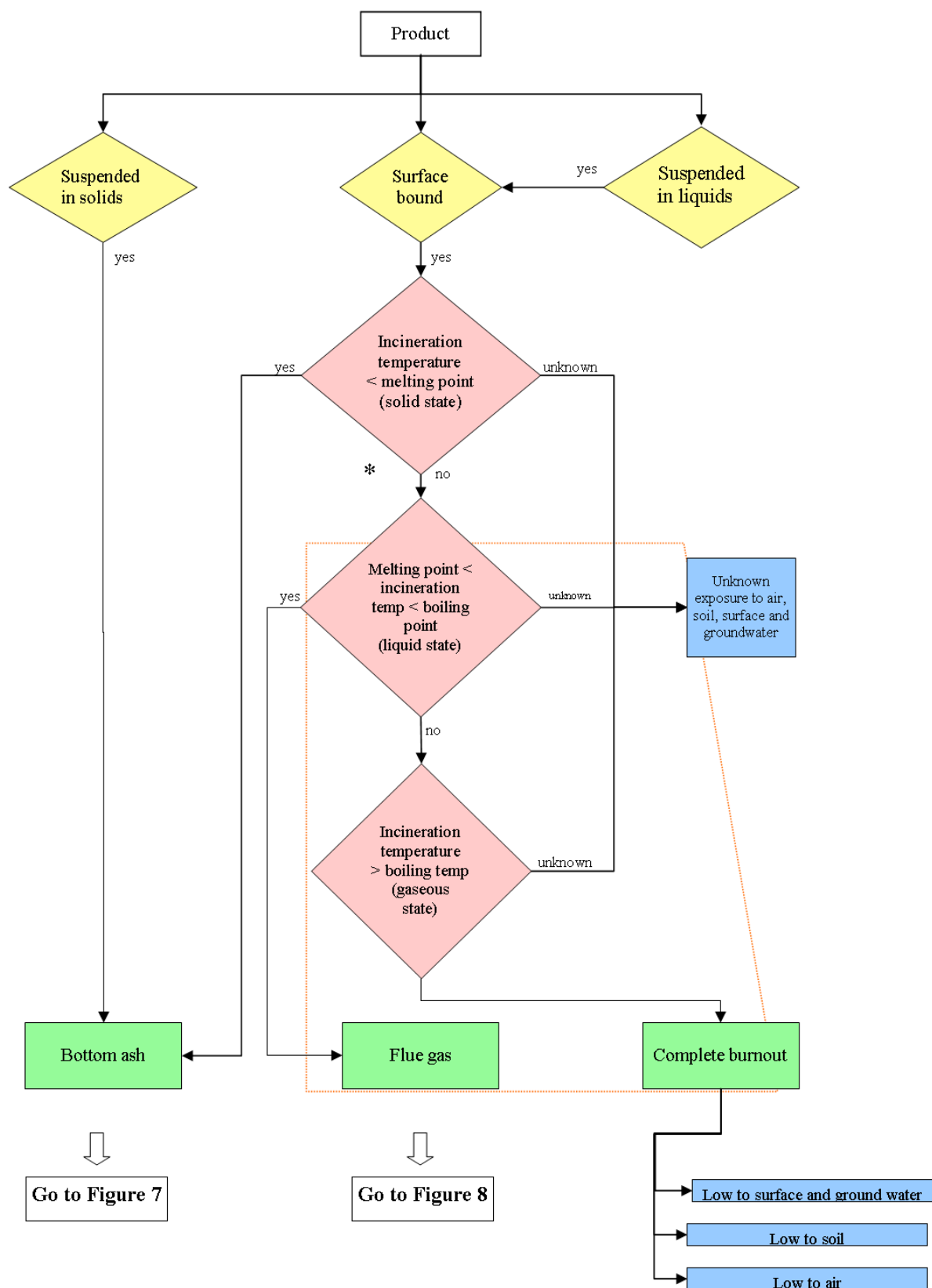


Figure 12 - NanoRiskCatWaste roadmap for nanowaste incineration. Yellow boxes are determination of location of NOAA in consumer product, pink are decisions on transformations, green are results of distribution media, purple boxes are possible residue treatment methods, and blue potential environmental exposure potential to either soil, air, surface and groundwater. White boxes refer to another NRCW roadmap.

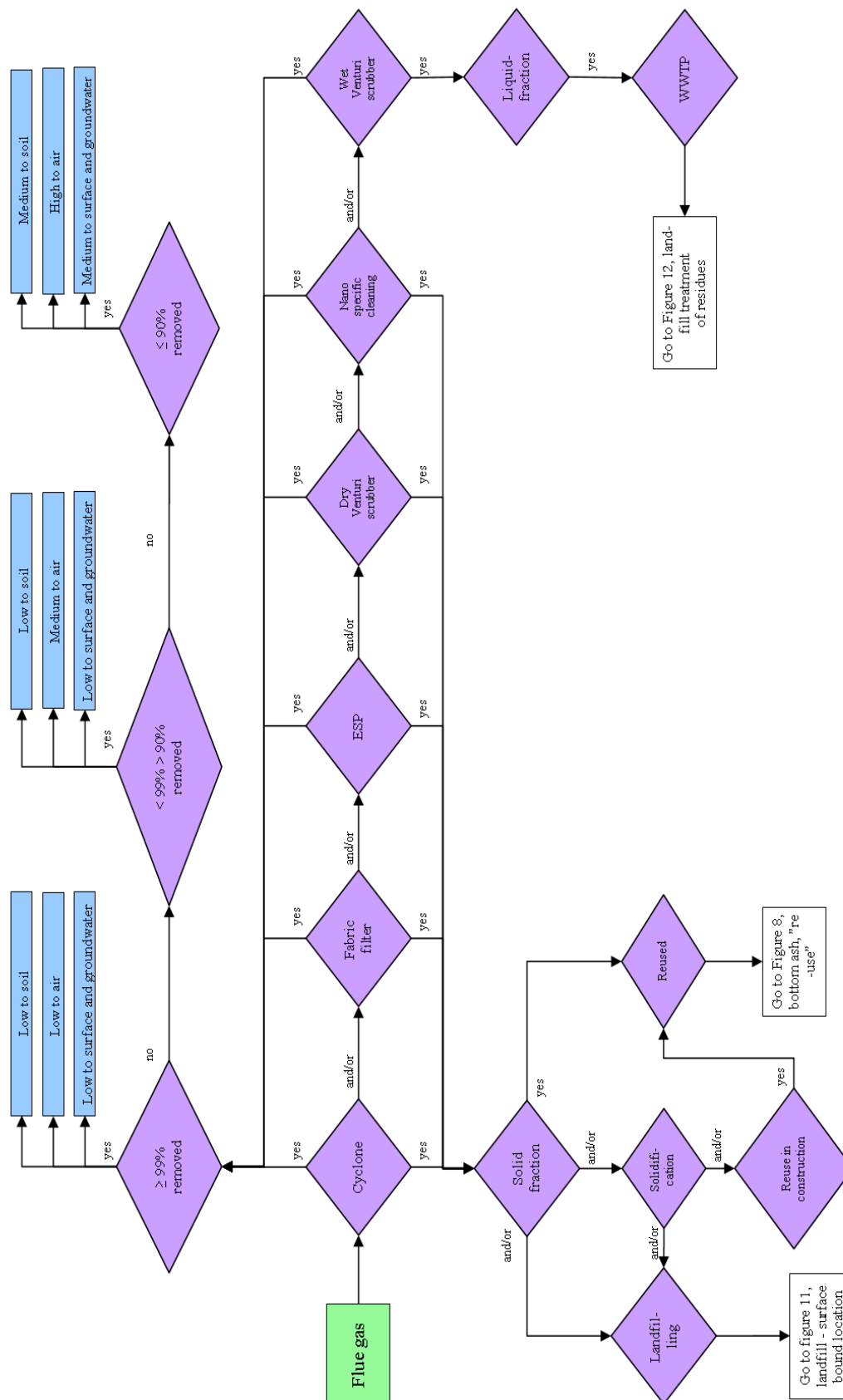


Figure 13 - NanoRiskCatWaste roadmap for flue gas cleaning and management APC residues. Green boxes are results of distribution media, purple boxes are possible residue treatment methods, and blue potential environmental exposure potential to either soil, air, surface and groundwater. White boxes refer to another NRCW roadmap. Flue gas can be treated in multiple APC systems.

6.2.2. NRCW for bottom ash

The roadmap for bottom ash management is shown in Figure 14. The solid residues from the incineration process can be reused, landfilled, or deposited in special closed systems. The potential exposure for individual treatments is shown, while for landfilled bottom ash the exposure potential can be determined by applying the roadmap on landfilling (Figure 15). In any case, the location of the NOAA will be “suspended in solids” no matter the origin location, due to solidification of the ash.

If the ash is deposited in closed, special systems, the potential emission to air, surface- and groundwater and soil is considered to be low, because the ash is stored in established, closed facilities with no contact to air, soil or surface- or groundwater. If the ash is landfilled in mineral waste landfills, the further fate depends on factors described in the section of landfills (see section 5.3). However, NOAA landfilled after an incineration process will be in a different kind of matrix (due to the heating process) than products that have become waste. This has to be in mind as this matrix might release the NOAA differently than wasted consumer products. Therefore, if bottom ash is landfilled, it is expected that NOAA are well encapsulated in the new matrix, and do not release from it. Use of residues of incineration ash, soil, building material etc. is allowed (Danish Ministry of the Environment, 2010). The regulation differentiates between uses with a solid coating and dense coating. When solid coated, a barrier of 1 m secures no contact with surroundings, and dense coated with a barrier that secures only 10% of the reused residues will be in contact with precipitation. The residues are allowed in roads, paths, squares, pipe trench, ramps and sound barriers with a maximum height and a minimum distance to i.e. water supply. When bottom ash reused for these purposes, the potential exposure to air is considered to be low, as the NOAA will have no or very limited contact to air. The potential exposure to both soil and surface- and groundwater depends on the coating. If solid coated, the potential exposure to all three environmental compartments is considered to be low, while dense coating allows some rainwater to come in contact with the NOAA which is expected in a medium potential exposure to soil and surface- and groundwater. The potential exposure to ground- and surface water and soil of reused ash depends on whether the NOAA are tightly bound in a matrix or not before use (Roes et al., 2012). There are different opportunities of such solidification and reuse, i.e. bound in cements or asphalt (Arm, M., Eighmy, T.T. & Christensen, 2010). Ash collected from APC systems can be used as mineral filler in asphalts or as a source of gypsum in wall board, while bottom ash can be used as unbound aggregates in i.e. road base or as a bound aggregate in i.e. cement, or as binding material in asphalt. In general, incorporation in cement is not sufficient to ensure no leaching of dry or semidry residues, as precipitation of residue chemicals can happen and over time the cement can degrade into powder (Flyvbjerg and Hjelmar, 1997). According to Roes et al. (2012), the ash can also be reheated to temperatures above 1400°C which will result in a glass like matrix. This matrix should also be able to avoid leaching of NOAA.

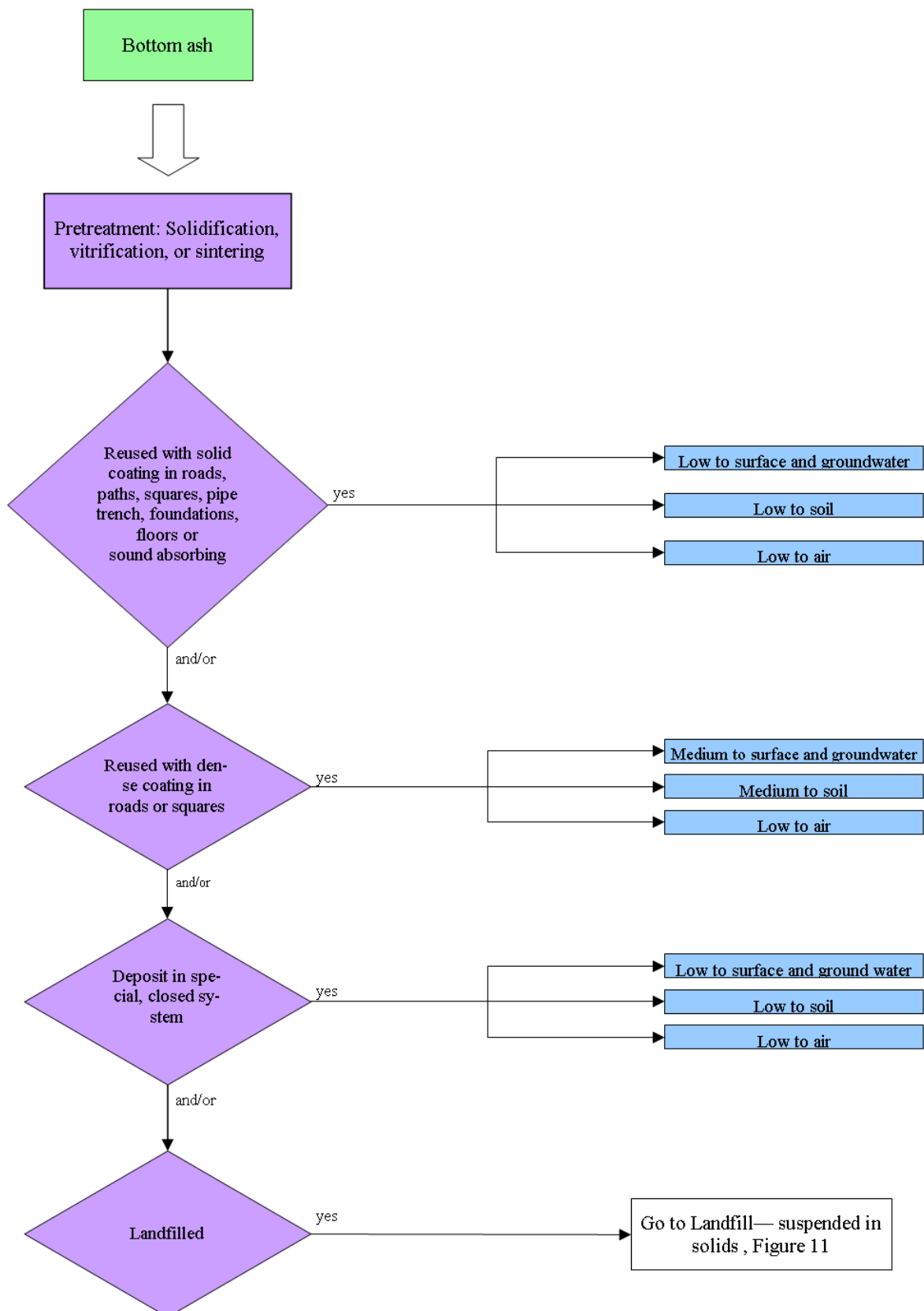


Figure 14 - NanoRiskCatWaste roadmap on incineration residue management of bottom ash. Green boxes are results of distribution media, purple boxes are possible residue treatment methods, and blue potential environmental exposure potential to either soil, air, surface and groundwater. White box refers to another NRCW roadmap.

6.3. Nanowaste roadmaps for landfilling

The roadmap for nanowaste landfilling is shown in Figure 15. NOAA suspended in solids of the consumer product can be assessed within existing knowledge regarding NOAA release from the product. Surface bound or NOAA suspended in liquids are immobile or mobile if the fraction of NOAA distributing to the leachate is $\leq 1\%$ or $> 1\%$ respectively. When immobile, NOAA distribute to the landfill waste matrix; if mobile, NOAA distribute to leachate, and the further treatment of leachate is studied wastewater treatment. If not possible to obtain a partition coefficient for the NOAA of matter, the mobility of the NOAA will be unknown. If the NOAA is associated with low mobility, the potential exposures to the three environmental compartments, air, soil, and surface and groundwater are also considered *low*. The landfill matrix is in fact well encapsulated and leakages of any material from the landfill are unlikely.

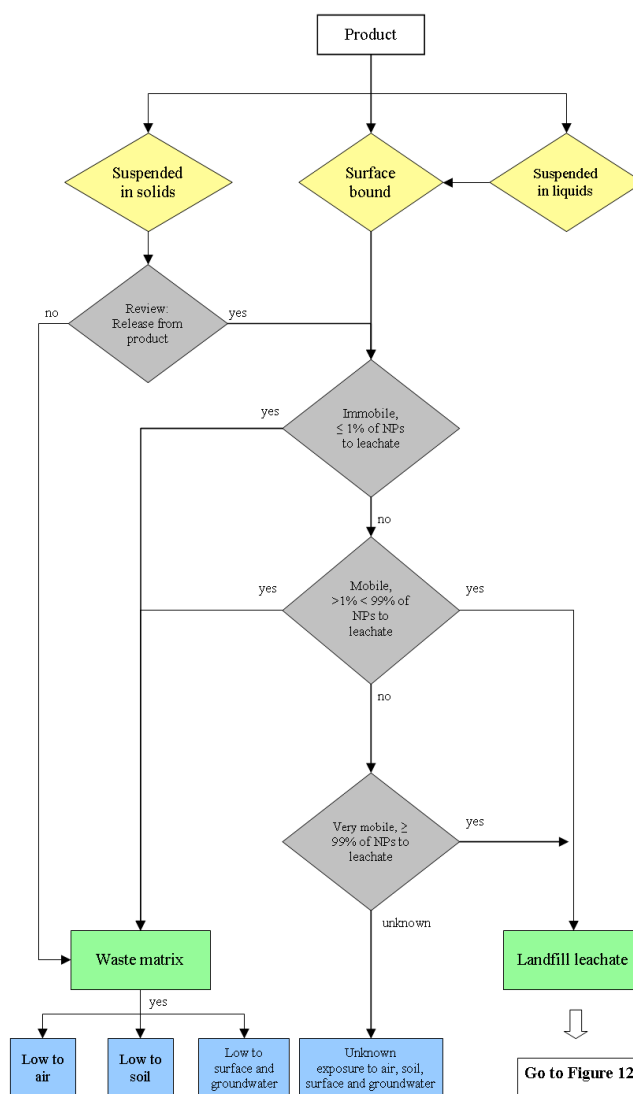


Figure 15 – NanoRiskCatWaste roadmap on landfill distribution. Yellow boxes are determination of location of NOAA in consumer product, grey are decisions on mobility, green are results of distribution media, and blue potential environmental exposure potential to either soil, air, surface and groundwater. White boxes refer to another NRCW roadmap.

6.3.1. NRCW for leachate treatment

The roadmap for leachate treatment is shown in Figure 16. Wastewater that infiltrates to ground has a high potential exposure of NOAA to soil, as the NOAA are in directly

contact with the soil. The potential exposure to air is low, because the NOAA most likely binds to soil rather than diffuse to air, and finally the potential exposure to surface and groundwater is high, because NOAA that distribute to leachate will neither bind to landfill matrix or soil. Therefore, NOAA can enter the water phase in soil and be moved to surface or groundwater. Discharge of wastewater to rivers, lakes or sea, is assumed to result in a low potential exposure to soil, because the NOAA are mobile, and thus stays in the water phase. The potential exposure to air is considered to be of medium potential as well as for wastewater infiltrated to ground. The exposure to surface and groundwater is expected to be high because of the direct exposure to surface water. Spraying of waste water to soil gives an exposure of high potential to soil, because the NOAA are directly sprayed onto soil. The exposure potential to air is considered of medium potential and of high potential to surface and groundwater as for waste waters infiltrated to ground.

Sludge from waste water treatment plant can be used for agricultural purposes or used in forest for fertilizing purposes, or it can be incinerated or landfilled. If the sludge is applied to soil, the potential environmental exposure is high to soil and surface and groundwater, because the NOAA is in direct contact with soil and the NOAA in sludge is mobile, so it can potentially move to surface and groundwater. The potential exposure to air is considered medium, because the NOAA can become in direct contact with air.

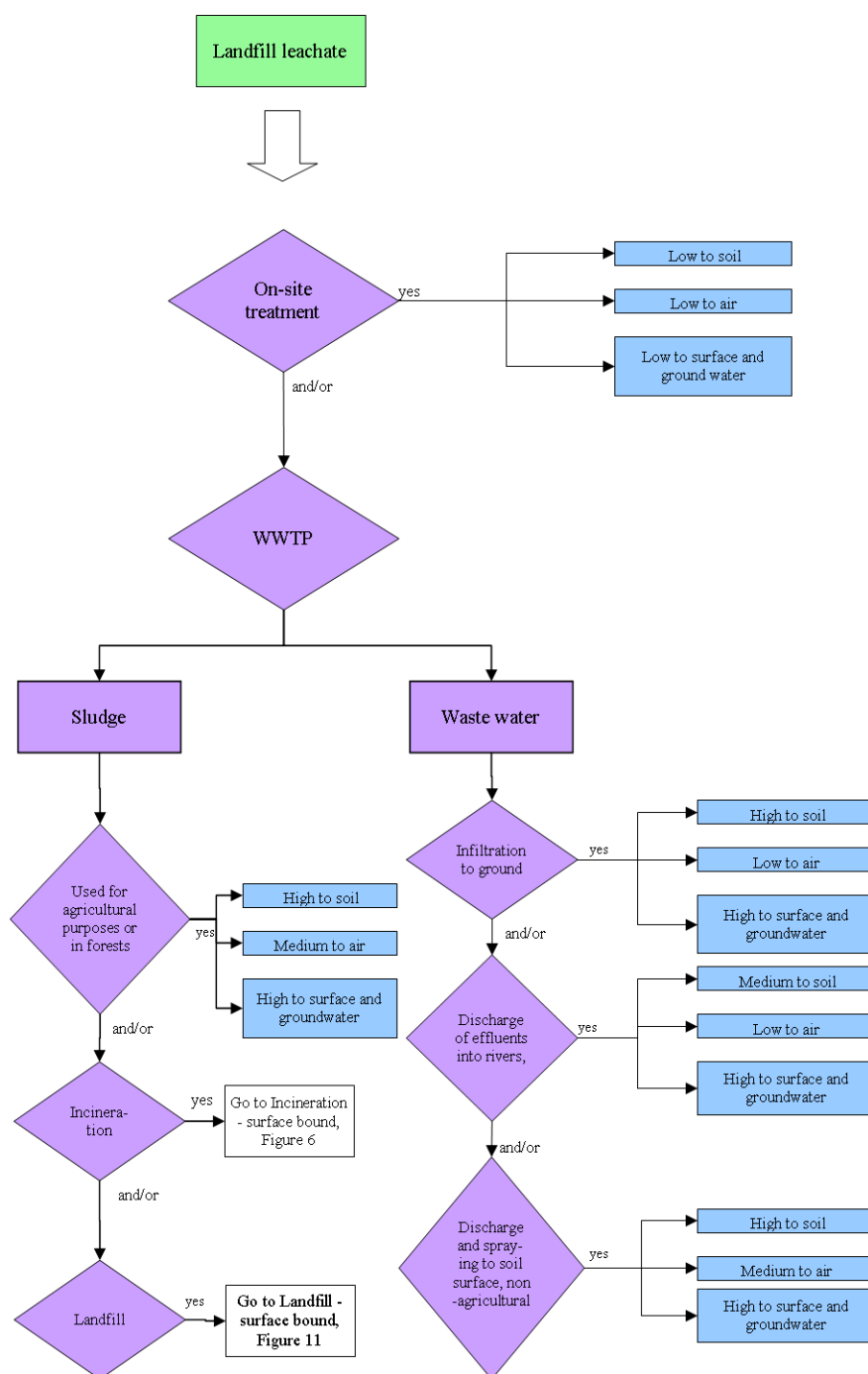


Figure 16 - NanoRiskCatWaste roadmap on landfill leachate treatment. Green box is distribution media, purple are treatment options, blue are environmental exposure potentials, and white boxes refer to different roadmaps. Landfill leachate can be treated as waste water on-site at landfills and/or at municipal waste water treatment plants (WWTP).

6.4 NanoRiskCatWaste for Nano-CuO Antimicrobial Wood Coating

Nano-CuO is surface bound to the wood, therefore melting point of nano-CuO is of relevance when it comes to incineration. The sizes of nano-CuO in treated wood is unknown, therefore melting points are found for different sizes of nano-CuO. Table 8 shows melting points of nano-CuO found in the existing literature.

Table 8 Melting point and boiling point of nano-CuO

Particle size	Melting point	Boiling point	Reference
40 nm	1326°C	-	(EPRUI Nanoparticles and microspheres Co, ltd.)
1-30 nm	1201°C	2000°C	(American Elements 2014)

It is assumed that the nano CuO treated wood is incinerated in regular incinerators, and that the flue gas combustion temperature is at least 850°C for two seconds, and likely around 1000°C. This means that nano-CuO most properly enter ash phase after incineration. Körner et al (2010) reports that compounds of treated wood distributes to ash phase during incineration, and furthermore that the ash is typically landfilled after incineration. Leaching abilities of Cu in ashes from incineration plants often exceeds limits (Körner et al 2010), indicating that Cu is mobile. Using the given information, the following NanoRiskCatWaste roadmaps for incinerated nano-CuO in treated wood emerge.

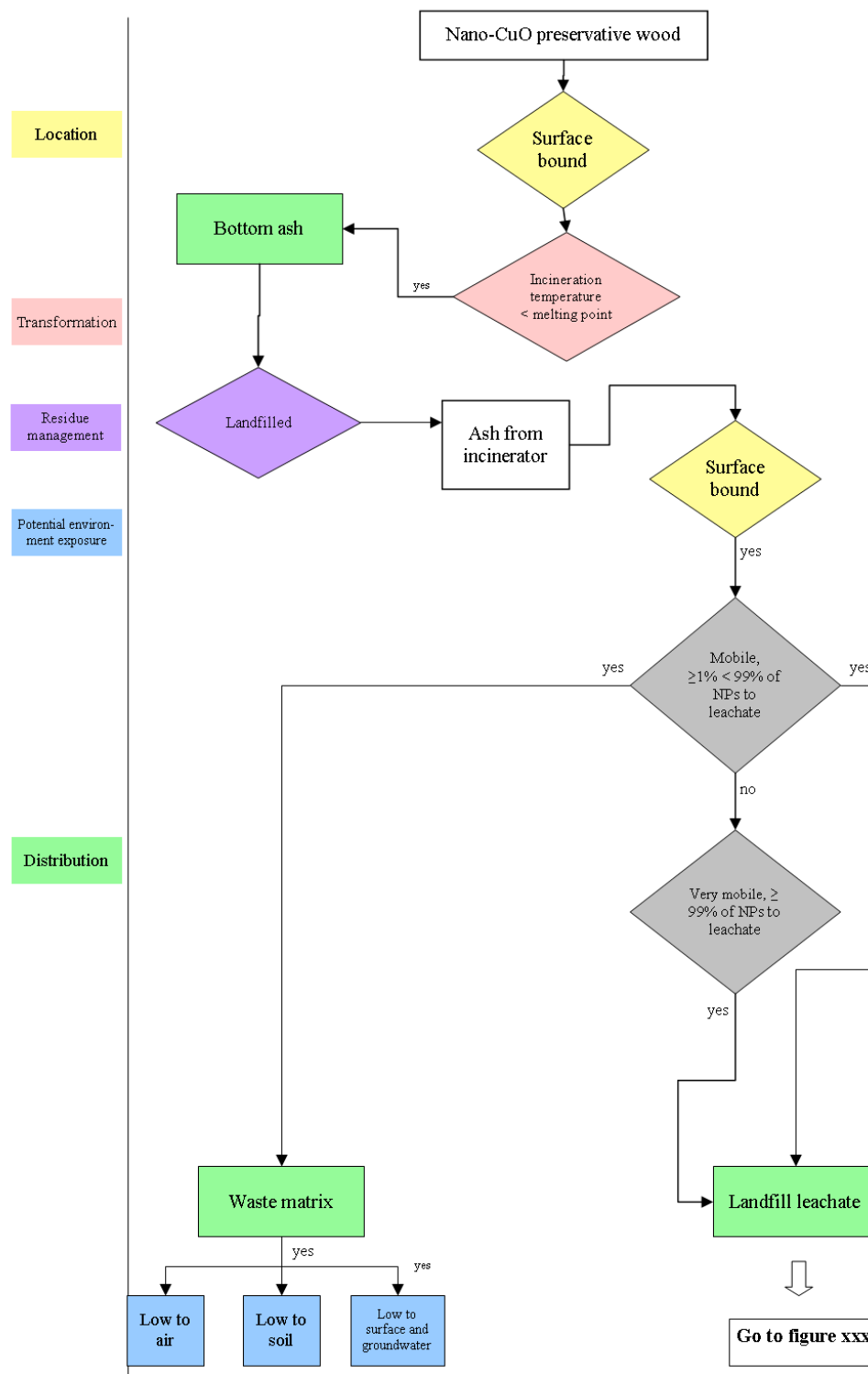


Figure 17 Incineration of CuO treated wood

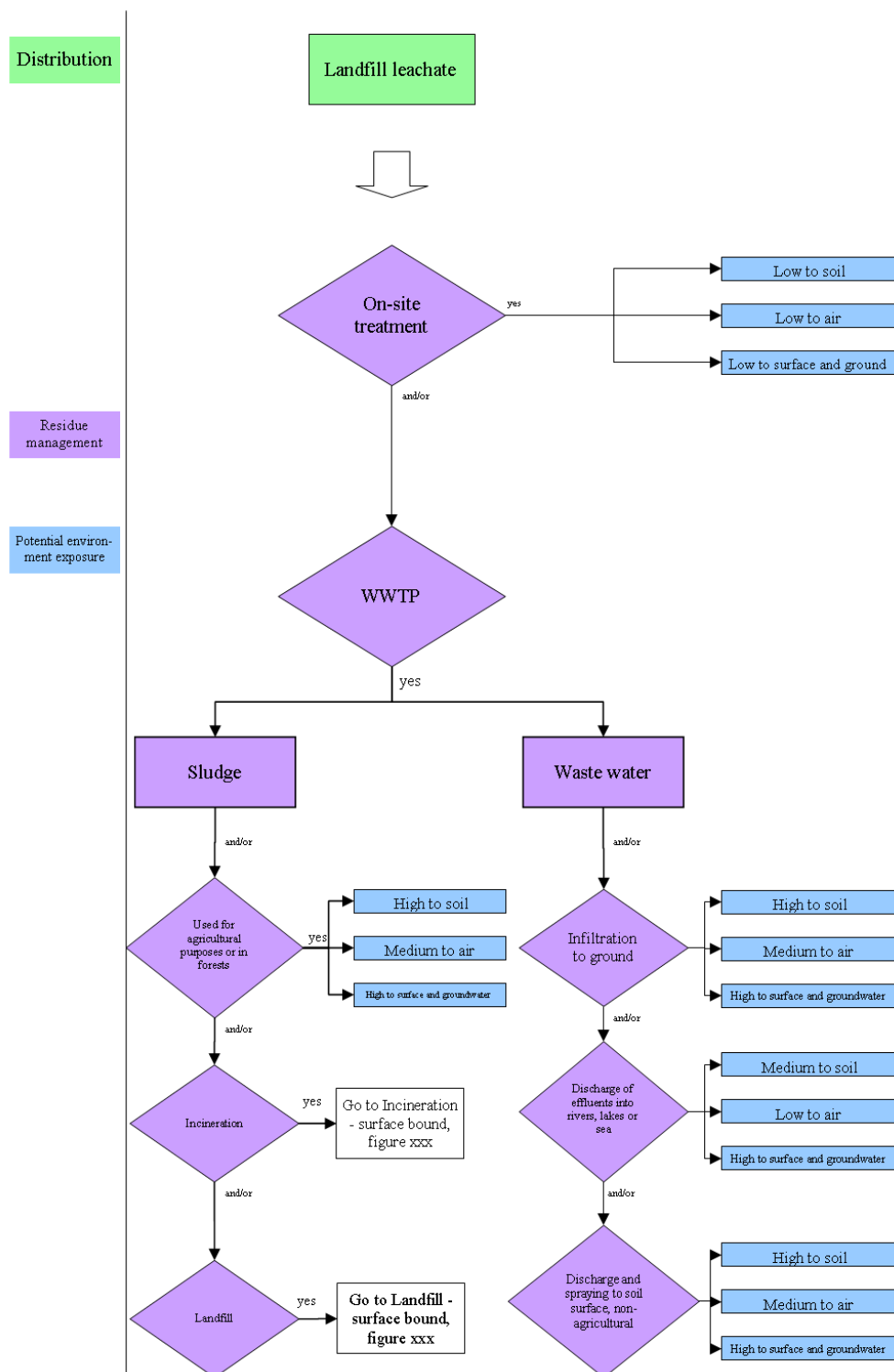


Figure XXX Leachate treatment of incinerated treated wood

For landfilling, the mobility of nano-CuO to the treated wood is important. According to Körner et al (2010), Cu does leach from the Cu treated wood, indicating that Cu enter leachate phase of a landfill, however, the K_d value for nano-CuO has not been found in existing literature and therefore it is not possible to perform accurate NRCW on nano-

CuO in impregnated woods.

Figure 18 Landfilling of CuO treated wood

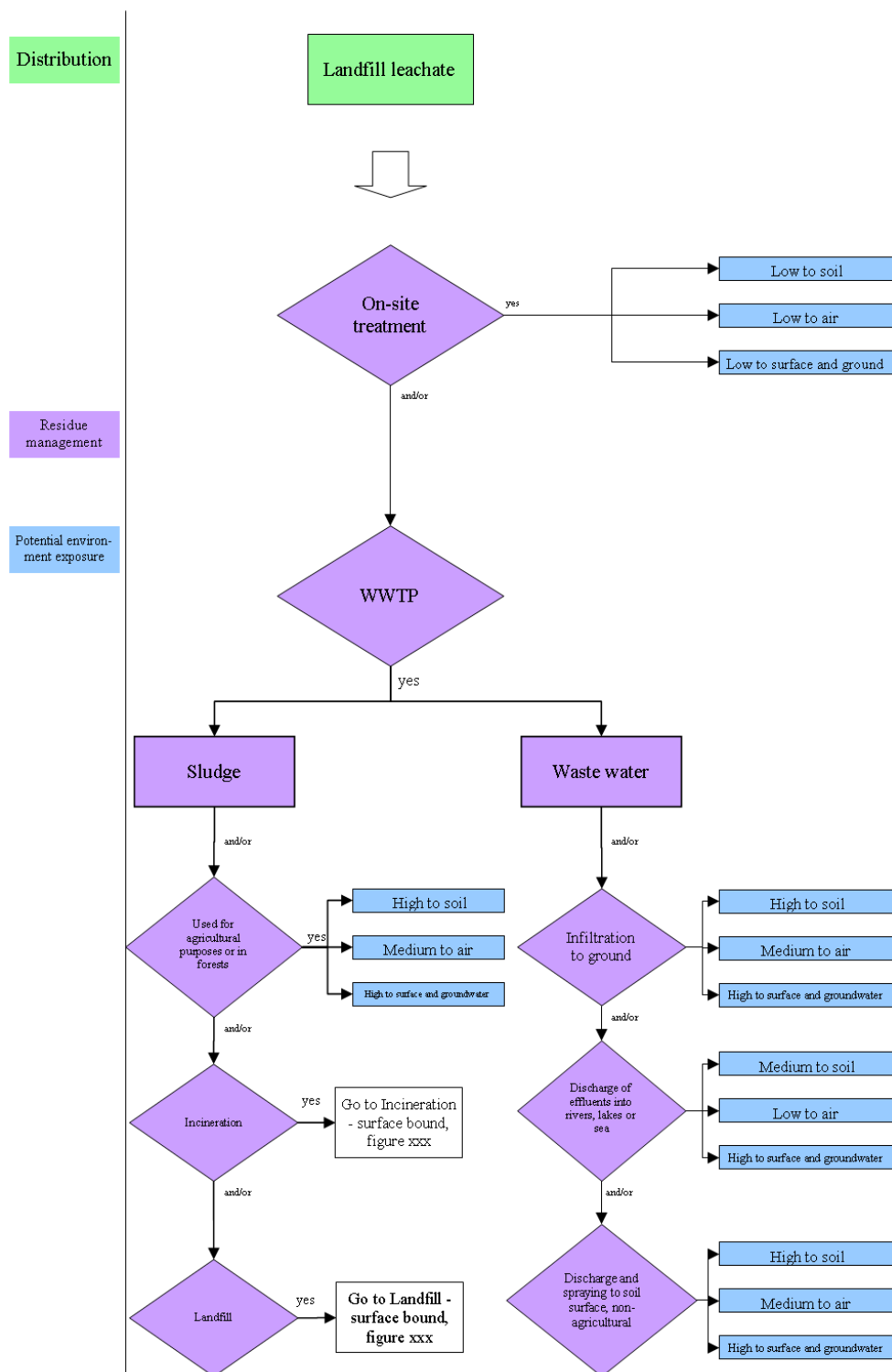


Figure 19 Leachate treatment of landfilled treated wood

6.5 NanoRiskCatWaste for NanoTiO₂ coated tiles

Nano-TiO₂ is used as coating for ceramic tiles for easier cleaning. It can be bought by households, and discarded together with regular consumer household waste.

The NRCW framework is suitable to describe the exposure potentials of nano-TiO₂ on ceramic tiles that have been discarded. This document will both focus on nano-TiO₂ discarded as the sold product, meaning the nanomaterial is suspended in liquids and discarded unused still the package, and as applied to ceramic tiles, where the location of the nano-TiO₂ then has become surface bound.

As the nanoTiO₂ is surface bound to the ceramic tile when used, the thermal stability is of relevance when regarding the incineration of the product. During incineration, the nanomaterial will settle to surfaces of other material, due to evaporation of the suspension, and the waste profile of both discarded products are similar. The melting temperature and boiling temperature is reported to be 1843°C and 2972°C, respectively (AzoNano 2013). When nano-TiO₂ is incinerated, the particles will be distributed to bottom ash. Bottom ash can be reused for infrastructural purposes with solid or dense coating given a *low* potential exposure to air, soil, surface and groundwater or a *low* exposure potential to air and a *medium* potential exposure to soil and surface and groundwater, respectively for solid and dense coating. If the bottom ash that contains nano-TiO₂ is deposit, the potential exposure for environmental exposure is *low* for the three compartments, air, soil and surface and groundwater. When landfilling the ash, the nano-TiO₂ is not expected to be released from the matrix, because the ash underwent solidification before landfilling. Therefore, the nano-TiO₂ will distribute to the landfill waste matrix, and the environmental exposure potential will be low to air, soil, and surface and groundwater.

Mueller et al (2013) modelled flows of nano-TiO₂ during incineration process, and came with the conclusion that 81% of nano-TiO₂ distributed to bottom ash and 19% to fly ash. This corresponds with the outcome of the NRCW roadmap, however the authors of the model also predicted nano-TiO₂ to enter the fly ash. No experimental studies on distribution of nano-TiO₂ are found during literature search.

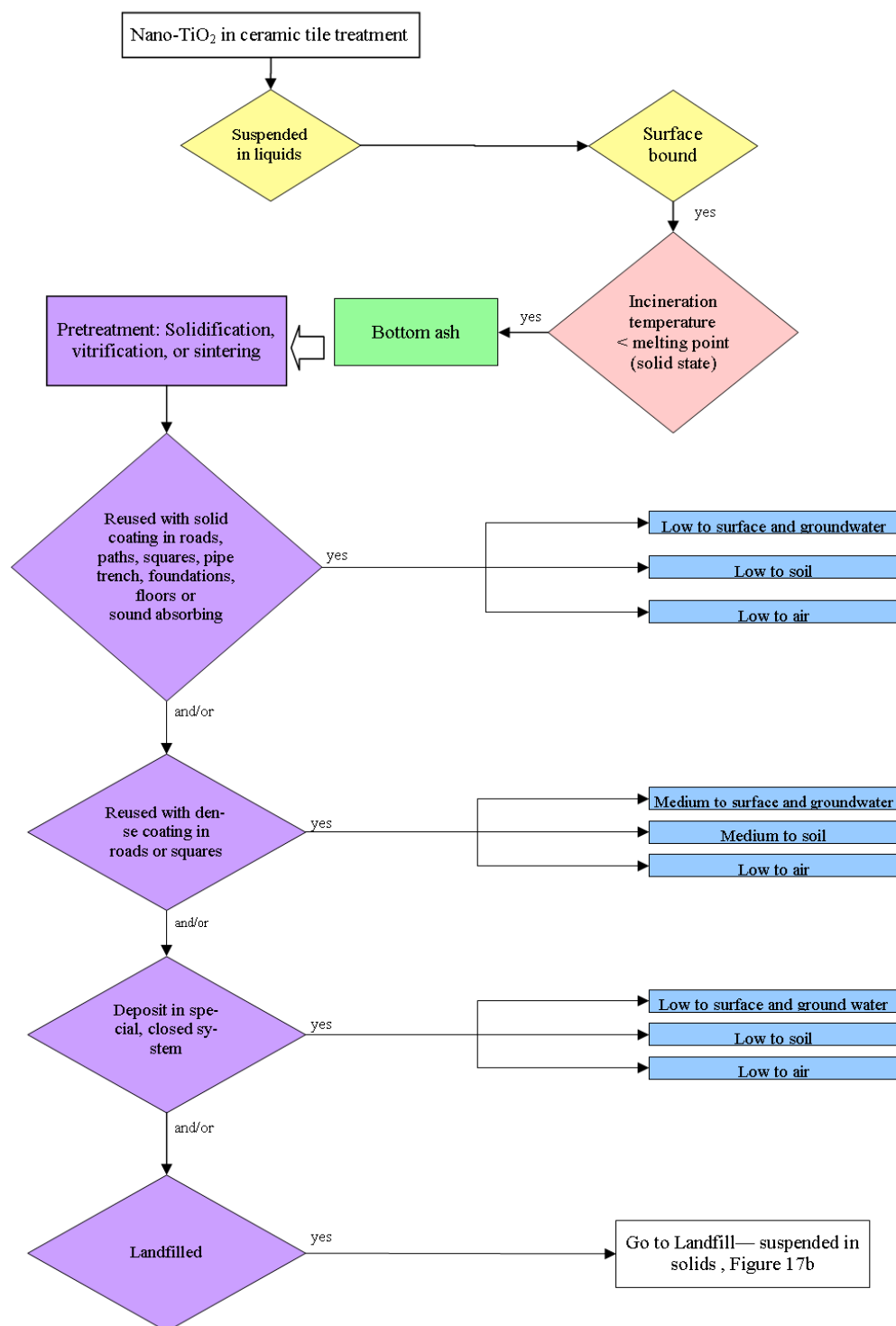


Figure 20 NRCW of incinerated suspension for ceramic tiles

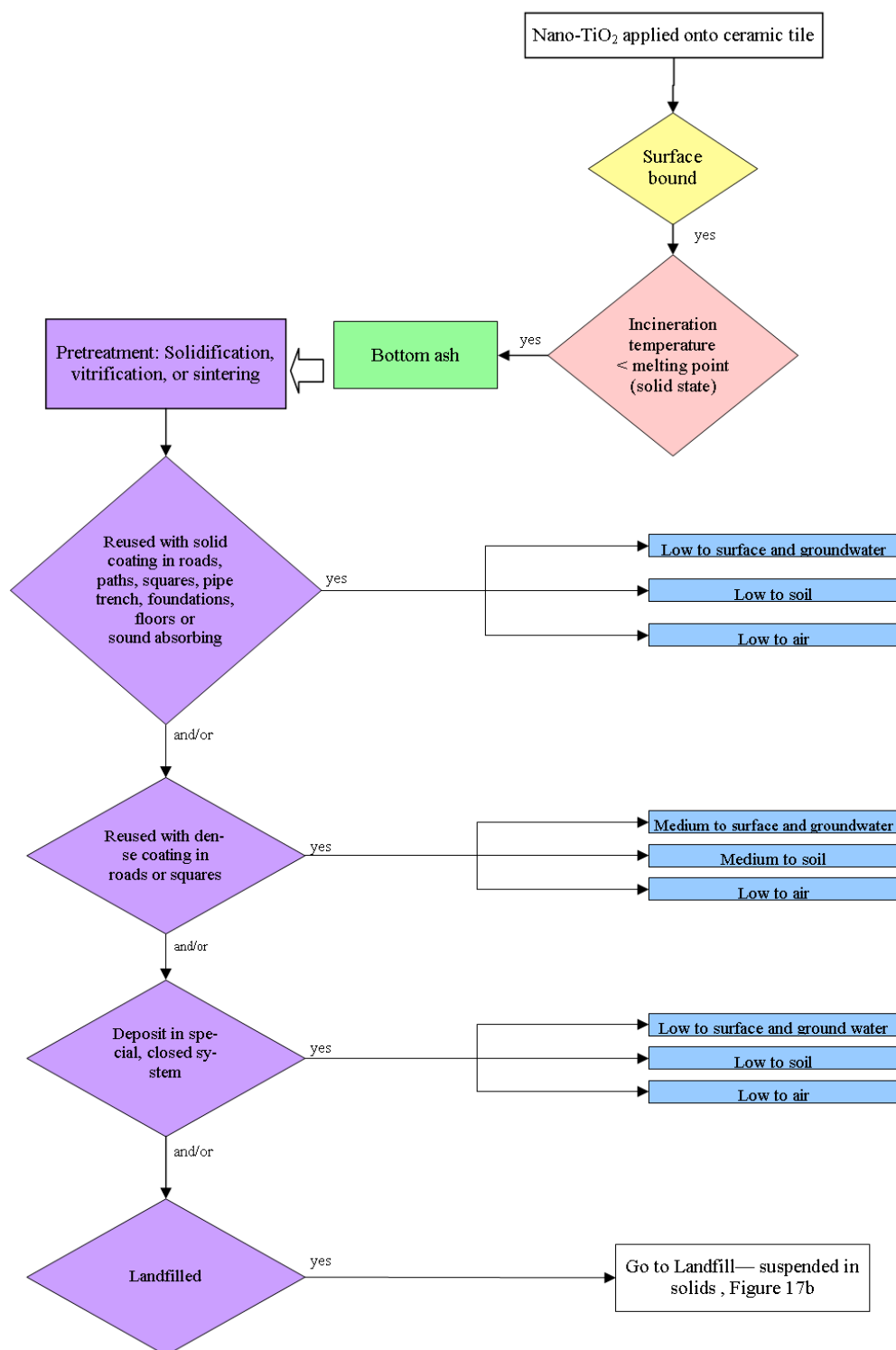


Figure 21 NRCW- incineration of nano-TiO₂ applied onto ceramic tiles

Because the nanomaterial is either surface bound or suspended in liquid and then become surface bound, the mobility of nano-TiO₂ in landfills is investigated. The

experimental K_d value in literature is found to be 740 L/kg, and the fraction of nano-TiO₂ that distribute to leachate is expected to be $f_{\text{leachate}} = \frac{1}{1 + \frac{0.6 \text{ kg/L}}{0.4} \cdot 740 \text{ L/kg}} = 0.0009$, and the nanomaterial is expected to stay bound to the waste matrix of a landfill.

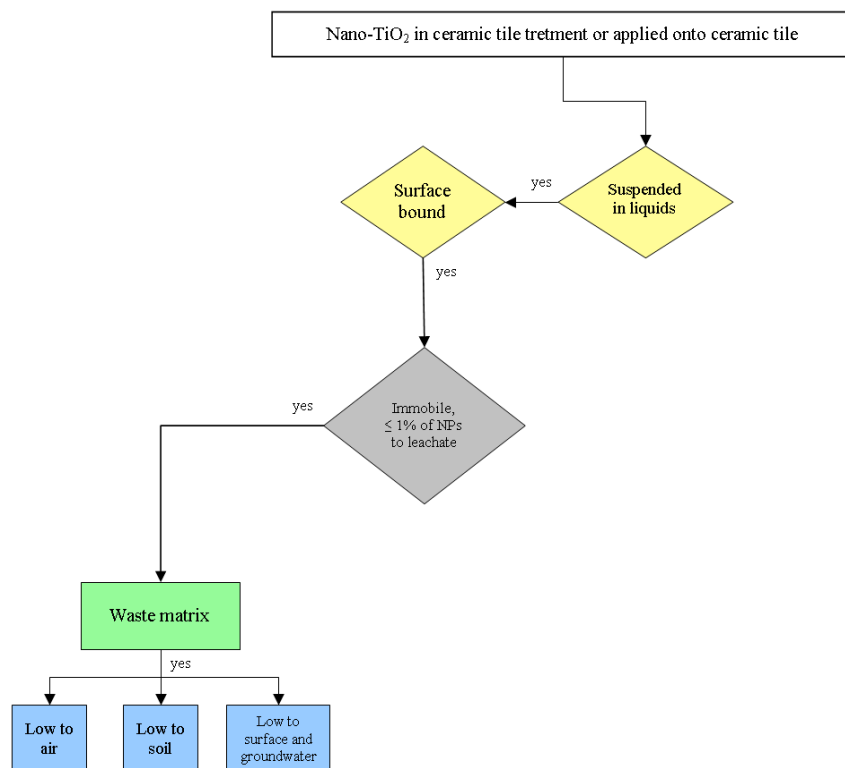


Figure 22

6.6 NanoRiskCatWaste for CNT in car bumpers

The CNT are suspended in solid plastic and are therefore expected to distribute to bottom ash after incineration. If the ash from incineration plant is reused for traffic applications with a solid coating, the exposure to air, soil, surface and groundwater is *low*. If the ash is reused with a dense coating instead of a solid, the exposure to air is *low*, however, the exposure to soil and surface and groundwater is *medium*.

The roadmap also states that the potential environmental exposure to soil, air, surface and groundwater is *low* if the bottom ash is deposited in a closed system. In case of landfilling of ash from incinerator bottom, it guides to the roadmap of landfilling.

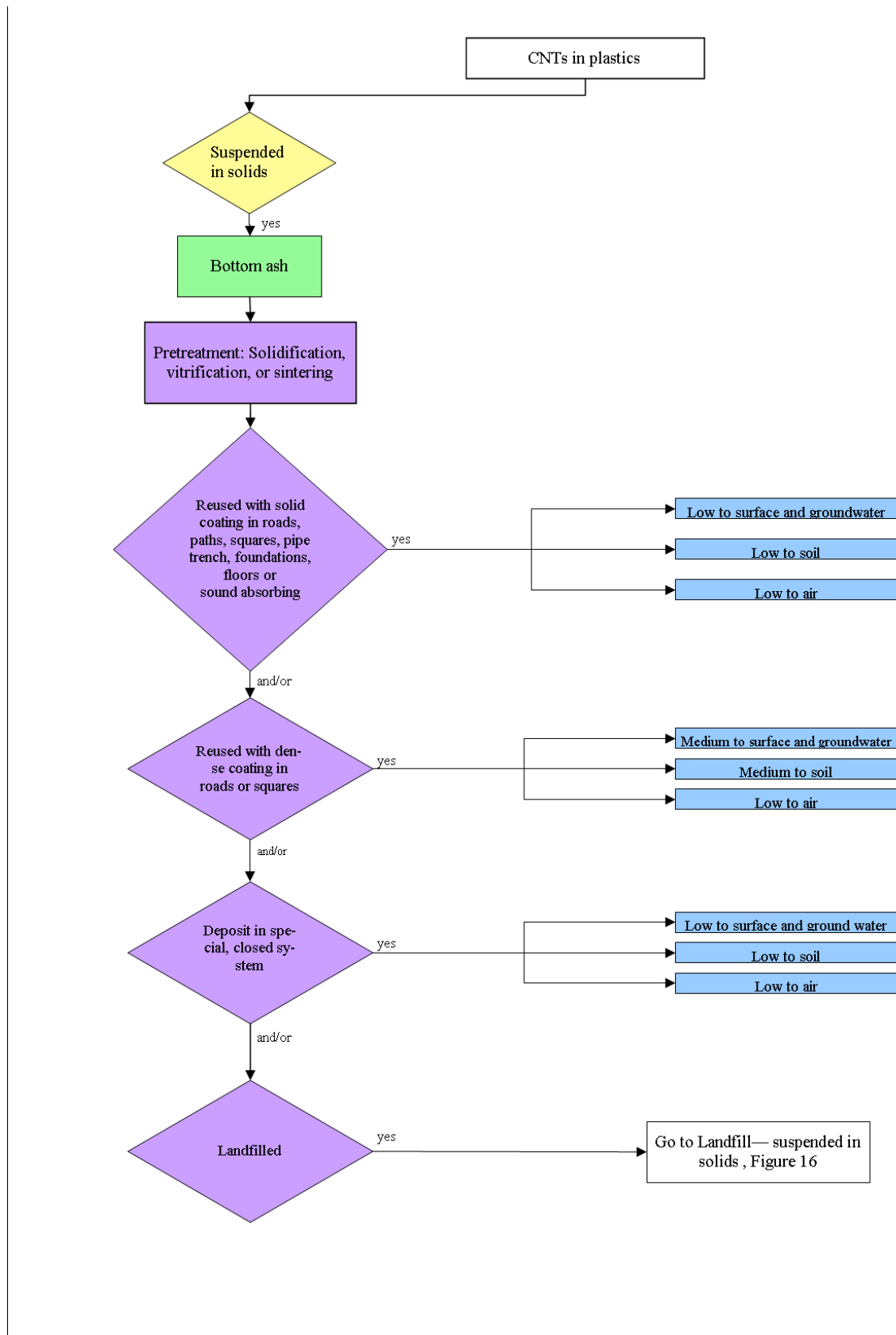


Figure 23

As the CNT is suspended in the solid plastic, the release rate of the CNT from the plastic is considered. One study tested the release of CNTs to water from thermoplastic polyurethane (TPU) (Wohlleben et al. 2013). The study showed that despite worst-case

scenarios with both impact of weathering and abrasion did not result in any release of CNTs. Therefore, the CNTs are expected to distribute and bind to the waste matrix of a landfill. According to roadmap Figure 9 the potential exposure of environmental exposure is *low* to air, soil and surface and groundwater, because the landfill matrix is well encapsulated.

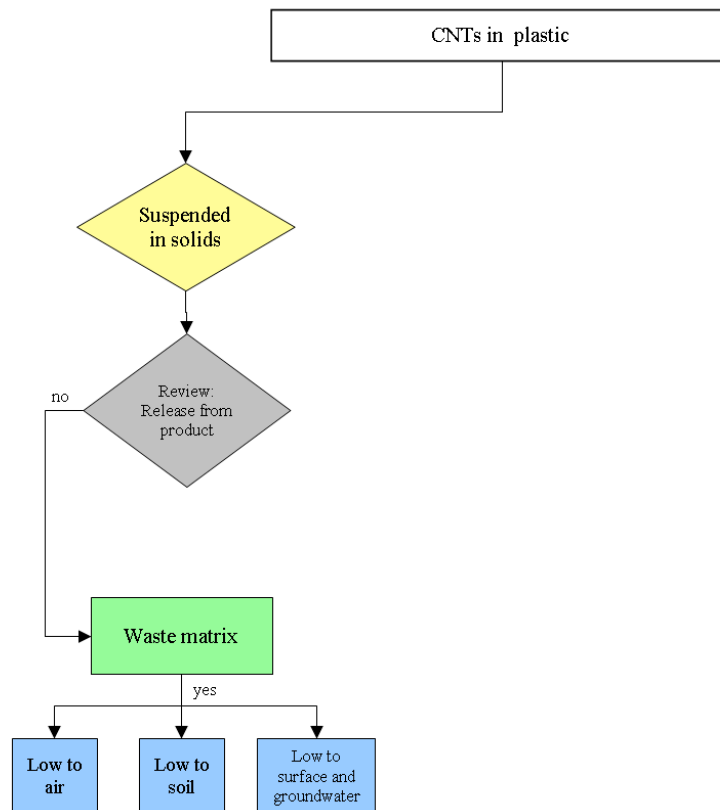


Figure 24

7. Guidelines for safe handling of waste flows containing NOAA Safe and safe-by-design recommendations

In this deliverable, we have assessed the waste material fractions expected to carry NOAA, the most prominent NOAA types, and the waste technologies involved in nanowaste handling. We have furthermore provided a brief description of individual waste treatment technologies, followed by a brief discussion on how the operation of individual processes may be affected by the presence of NOAA, and an overview of NOAA release pathways and important factors affecting it. Finally, we developed a series of NanoRiskCatWaste profiles for selected SUN materials. Based on this work a series of guidelines can be developed for the safe handling of waste flows containing NOAA.

7.1 Before nanoproducts are produced and becomes waste

First of all, it seems clear that minimizing nanowaste starts even before the nanomaterials and the nanoproducts are produced. Knowing the eventual pathway of the nanoproduct to be produced and waste treatment flows and practices in the country of eventual disposal is key as some countries have preferences for incineration whereas others prefer landfilling. In general, it seems that the low combustibility is to be preferred when it comes nanoproducts for which incineration is eventually to be expected, whereas a non-degradable matrix and a low affinity of the NOAA for the liquid phase may be important for nanoproducts that might end up in landfills. For nanoproducts that might be recycled, high melting/boiling point of the NOAA, non-hard matrix material and low affinity for the liquid phase are very important. Considering these aspects in the design phase would dramatically minimize release of NOAA in the waste phase.

7.2 Recommendations when it comes to recycling

When it comes specifically to recycling, the recycling processes for paper, glass, etc. are very different from each other and depend on the material being processed and hence general recommendations are hard to come by. For the individual waste material types, we recommend:

- Paper: limit the use of persistent NOAA, because of the clear risk of accumulation.
- Plastic: limit the use of persistent NOAA, because of the clear risk of accumulation; use NOAA with a low vapour pressure.
- Glass: use NOAA with a melting/boiling point higher than glass melting temperature.
- Metal: use NOAA with a melting/boiling point higher than glass melting temperature.
- C&D (in particular concrete, tiles, mortar): limit the use of NOAA because of the risk of airborne release during shredding and crushing operations; use NOAA with low affinity for liquid phase to prevent release into leachates.

7.3 Recommendations when it comes to incineration

During incineration can be liberated because the matrix is destroyed or because NOAA are volatilized. In order to avoid that NOAA are liberated from a solid matrix during incineration and end up in the gas phases, we recommend that NOAA are embedded in not-combustible a solid matrix when used. We furthermore recommend that NOAA are designed to have a melting or boiling point lower than the combustion temperature in

the combustion chamber so that NOAA are more likely to be destructed by melting and burning. In order to prevent release of NOAA into the environment, we recommend the use of state-of-the-art flue gas cleaning systems where at least fabric filters, wet scrubbers and activated carbons are present, to ensure that NOAA that might be released to the gas phase during the combustion process are captured. Here, care should be taken to design NOAA that do not have certain properties, e.g. to be ionised when the NOAA enters the APC systems as this might lead to the NOAA not being trapped in the APC system. We also recommend that any bottom ash matrix be treated before any reuse, deposit, or landfilling take place in order to avoid leaching of NOAA. Such treatment could include solidification into i.e. cement, lime, or asphalt, vitrification or sintering. As bottom ash is often mechanically processed to recovery metal fractions and/or to achieve desired material properties (e.g. grain size), great care should be taken to avoid release of NOAA.

7.4 Recommendation when it comes to landfilling

When it comes to landfilling, we recommend that nanoproducts are only landfilled in modern landfill where the environmental release of gas and leachate are mitigated using different technological approaches, including bottom liners, leachate drainage, top covers, gas collection systems, and by placing that landfill as far as possible from sensitive environmental receptors. We furthermore recommend avoiding landfilling nanoproducts that contain nanomaterials that display an inhibitory effect on aerobic and anaerobic landfill processes e.g. nanosilver. Leachate treatments should include e.g. stripping, membrane, or biological processes, evaporation, membrane, adsorption, chemical oxidation, wetland, nitrification, and denitrification.

7.5 Recommendations when it comes to identification and labelling of nanoproducts

Many of the recommendations that we have presented here, require that products that entail nanomaterials can be easily identified. In order to ensure easy identification of nanoproducts, we recommend that labelling is required.

3. Deviations from the Workplan

No deviations from the workplan

4. Performance of the partners

All partners performed in satisfactory time and quality.

5. Conclusions

The Steering Board deems this deliverable to be fulfilled satisfactory.

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