Materials Science & Technology

EMPA, Swiss Federal Laboratories for Materials Science and Technology

Nanomaterials in Landfills
Module 3:
Nanomaterials in Construction Waste

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Summary

Engineered nanomaterials (ENMs) used in the construction industry will enter recycling and construction waste through renovation and demolition of buildings. Information about ENM flows in these processes is currently insufficient, thus the potential release of ENMs into the environment from this waste and the implications of this release are unknown. The aim of this project was to investigate if there is currently any nano-relevance in construction waste from buildings (Hochbau). To provide an answer to this question, we evaluated the sources and the flows of ENM in construction waste, identified the potential exposure pathways of ENMs for humans and other organisms and then quantified the release of ENMs to technical compartments and the environment.

To obtain information about ENM applications and amounts used in the Swiss construction industry, we surveyed representatives from companies in this sector. Although the survey revealed a lack of knowledge about the application of ENMs in construction, it showed that ENMs are mainly used in paints and cement. The most frequently used ENMs in construction are nano-titanium dioxide (nano-TiO_2), nano-silicon dioxide (nano-SiO_2), nano-zinc oxide (nano-ZnO) and nano-silver (nano-Ag). We complemented this information with literature data and market reports to estimate the mass content of ENM-containing construction products and to calculate the amount of ENM in paint and cement sold in Switzerland for the year 2012.

With a bottom-up semi-quantitative approach, we estimated the flows of ENM contained in paints to recycling and landfills. The results show that waste from paint is contained in materials like concrete, bricks, gypsum or wood and the flows of the ENM are determined by the flows of these building materials. We estimate the ENM amount used in paints to be 14 t/y for TiO_2, 12 t/y for SiO_2, 5 t/y of ZnO and 0.2 t/y for Ag. A comparison with a top-down approach yields very similar results, supporting the validity of the obtained mass flows.

The main amounts of ENMs contained in paints will enter the recycling system from where they are finally transferred to landfills (23 t/y), followed by direct input into landfills (7 t/y) and waste incineration (0.01 t/y). We recommend performing the same type of analysis for other products containing ENM like cement or insulation materials, with a basis on detailed market data. Our results allow us to determine (at least qualitatively) the potential release of ENM in any of the technical or environmental compartments. The potential for release of ENMs is clearly high during recycling, so we recommend to place special emphasis in the future on this process due its importance for occupational health. Because landfills are probably the final compartment for most of the ENM, further analysis is required to determine the release potential to environmental compartments from these facilities.

Zusammenfassung

Künstlich hergestellte Nanomaterialien (ENM) in Bauprodukten gelangen nach dem Rückbau zum grössten Teil über das Baustoffrecycling auf Deponien. Das Wissen über die Flüsse der ENM in Bauprodukten- und Abfällen ist zur Zeit noch ungenügend und es ist wenig über eine mögliche Freisetzung und allfällige negative Auswirkungen für die Umwelt bekannt. Das Ziel dieses Projektes war es, nano-relevante Aspekte im Bauabfall (aus Hochbau) genauer zu untersuchen. Dazu untersuchten wir die Quellen und Flüsse von ENM in Bauabfällen in der Schweiz, identifizierten das mögliche Expositionspotenzial und berechneten die Massenflüsse von ENM in Bauabfällen.

Um Informationen zu den ENM-Anwendungen und Mengen in der Schweizer Bauindustrie zu bekommen, führten wir unter Vertretern von Firmen aus diesem Sektor eine internetbasierte Umfrage durch. Die Umfrage zeigte klar, dass wenig Wissen über die Anwendung von ENM im Bausektor vorhanden ist. Die wichtigsten Baumaterialgruppen, welche ENM enthalten, sind Farben und Zement. Die meistverwendeten ENM sind nano-TiO_2, nano-SiO_2. Nano-ZnO und nano-Ag. Wir ergänzten diese Information mit Literaturdaten, um die ENM-Gehalte und Mengen in Farben - als wichtigster Kategorie - für das Jahr 2012 in der Schweiz bestimmen zu können.

Mit einem „bottom-up" Ansatz schätzten wir die Flüsse von ENM aus Farben im Bauabfall ab. Pro Jahr werden ca. 14 Tonnen nano-TiO_2, 12 Tonnen nano-SiO_2, 5 Tonnen nano-ZnO und 0.2 Tonnen nano-Silber verwendet und gelangen so früher oder später in den Bauabfall. Ein Vergleich mit einem „top-down" Ansatz, welcher auf unabhängigen Ausgangswerten basiert, zeigt sehr ähnliche Mengen. Der Hauptanteil der ENM in Farben gelangt über das Baustoffrecycling auf Deponien (23 Tonnen/Jahr), gefolgt von direkten Eintrag auf Deponien (7 Tonnen/Jahr) und KVAs (0.01 Tonnen/Jahr). Wir empfehlen, ähnliche Abschätzungen auch für andere Produkte vorzunehmen, was jedoch bedingt, dass zuerst zuverlässige Angaben über den Markt gesammelt werden müssen.

Unsere Resultate erlauben es uns, qualitativ das Potenzial für Freisetzung der ENM in die Umwelt abzuschätzen. Dieses Potenzial ist einerseits während des Recyclingprozesses relativ hoch, was vor allem für die Arbeitsplatzsicherheit von Bedeutung ist. Da Deponien für die meisten ENM die endgültige Senke sind, braucht es zudem weitere Untersuchungen, welche das Potenzial der Freisetzung aus Deponien in die Umwelt genauer untersuchen.

1. Introduction

The construction sector comprises a wide variety of industries using different materials such as cement, metals, paints, glass, wood and others. During the last decade (2001-2010) the construction sector exhibited a positive evolution, with an average growth rate of 2.5%, a minimum of 0.1% in 2003 and a maximum of 4.4% in 2008 (FSO, 2014). On the other side, nanotechnology has steadily developed during the last decades in many industrial sectors, including the construction sector. Engineered nanomaterials (ENM) are produced in increasing amounts and have found applications in the construction sector in antimicrobial paints, self-cleaning windows, stronger cement, and more efficient solar panels, etc.

Despite the current relatively high cost of nano-enabled products, their use in the construction sector is likely to increase because of valuable properties impaired to the construction applications. The rapid development of new applications will decrease the cost of ENMs as they are produced in larger quantities. Recent studies suggested that workers in the construction industry handling nano-products mostly worked with cement or concrete products, coatings or insulation materials (SCAFFOLD, 2012).

However, developing technologies always raise concerns about their impacts on humans, other organisms, and the environment. This aspect is relevant not only from a safety and environmental perspective, but also from the economical side, because the long-term success of an industry relies to a large extent on the capacity to create safe products and applications.

There is an increasing effort to better understand the risks associated with nanotechnology. Risk information should be generated to provide elements to society, industry and regulators to improve their decisions and eliminate or reduce as much as possible the negative effects of a technology and at the same time maximizing its benefits. The characterization of risk associated with ENM or nano-enabled applications is still in its early stage and further advances are needed to achieve a comprehensive and realistic characterization of risk. However, the information currently available makes it possible to generate scientific information that provides the first elements to manage those risks.

The original question that we ask for this study was:

"***Is there any nano-relevance in construction waste?***"

To provide an answer to this question we need to assess the following elements:

[i] identify the sources and the flows of ENM in construction waste,

[ii] identify the potential exposure pathways of ENMs for humans and other organisms,

[iii] identify the release of ENMs to technical compartments and the environment and

[iv] identify the hazard they represent for organisms. After gathering all these elements, a characterization of risk is plausible.

In this work we focus on elements [i-iii]. We provide a definition of ENMs (Chapter 3) and then describe the potential use of ENM in the construction sector based on the available literature (Chapter 4). In Chapter 5, we characterize the waste flows for construction and demolition waste (C&DW) from buildings (Hochbau) in Switzerland (amounts and material types). We then describe the use of nano applications in the construction sector in Switzerland using the results of a survey we distributed among experts and a recent report published by the French government on ENM production (ANSES, 2013), (Chapter 6). Chapter 7 describes the behavior of ENMs in the construction sector. In Chapter 8, we provide a semi-quantitative assessment of the flows of ENM in C&DW based on assumptions that reflect the current situation in Switzerland, and it shows the potential release pathways that we identified. We finalize in Chapter 9 with a discussion on the assumptions used and the validity of the results, besides the limitations we faced and the most important uncertainties that need to be overcome in the future to provide more robust results. We close with the conclusions in the same Chapter.

Point [iv] is not covered here because the current state of the information makes impossible to characterize properly the risk of ENM in construction waste. Furthermore, our original intention was to model quantitatively the flows of ENM within the system, but this was not possible. Our main limitation was the scarcity market information that could be used to assess the amounts of ENM entering the Swiss system. Further limitations are discussed in Chapter 10.

The relevance of this report is to provide information that allows the identification of the main ENMs and nano-enabled applications present in the Swiss waste flows, so in the future toxicity and exposure assessments at critical points can be performed using analytical tools.

2. Motivation and goals of the study

Our motivation of this study is based on the results from the *Nanomaterials in waste incineration and landfills study* (Müller et al., 2012). This study aimed at identifying the total nano-fraction of fly ash (weight and particle number) from waste, wood and sludge incineration in Switzerland by particle size measurements and to compare it to the modeled amount of ENM as well as to the modeled amount of nanoparticles derived from conventional pigments. In addition, first measurements were made to analyze the size distribution of fly ash before and after acid washing. The results allowed a first estimation of the importance of ENMs for waste streams in Switzerland. The modeling showed that an important flow of nanomaterials goes directly from construction waste into landfills. These wastes were covered only very generally in the modeling because the main focus was on the flows through the waste incineration plant.

Based on that work, the present study aims to investigate nanomaterials in the construction waste flows in Switzerland. An estimation of the potential release during recycling will be made. The result of the estimation will be used to model the nanomaterial flows from buildings to landfills and the environment.

For this purpose, we focused on the following ENMs: TiO_2, SiO_2, ZnO, Ag, CuO, and $CuCO_3$. The following steps were taken:

1. Collection of **data on material flows of construction waste** to landfills, waste incineration and recycling.
2. Estimation of the **current amounts of nanomaterials in Swiss construction waste**.
3. **Modeling the nanomaterial flows** from buildings to landfills and the environment.
4. **Evaluation of the release potential** of nanomaterials.

3. Nanomaterial definition

Currently, there are several international definitions of nanomaterials, for instance ISO or EU:

- **The International Standards Organization** (ISO, 2010) defines a nanomaterial as a material with any external dimension in the nanoscale (size range from approximately 1 nm to 100 nm), called nano-object, or as nano-structured materials having internal structure or surface structure in the nanoscale (Figure 1).
- **According to the European Commission** (EU-Commission, 2011) a nanomaterial means a natural, incidental or manufactured material containing particles, in an unbound state or as an aggregate or as an agglomerate and where, for 50% or more of the particles in the number size distribution, one or more external dimensions is in the size range 1 nm - 100 nm.

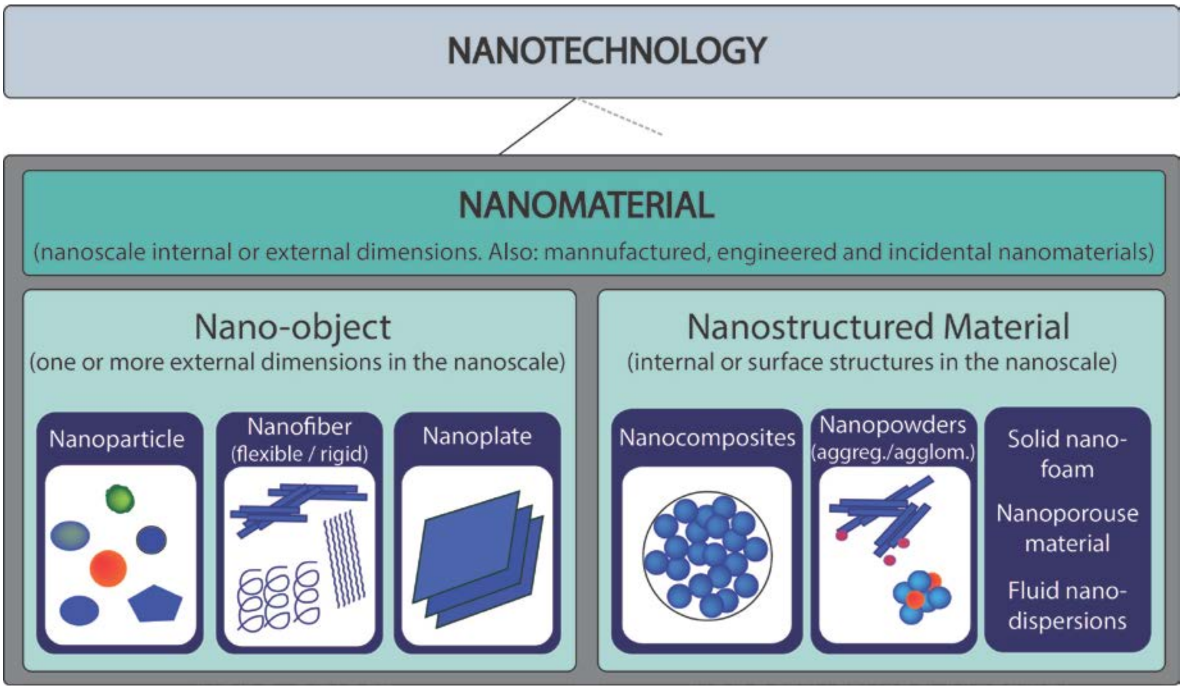

Figure. 1. Definition of nanomaterial according to ISO TS 27687 (ISO, 2008).

One has to take into account that some of these definitions are still in development. Several categories of nanomaterials (SCENIHR, 2010) have been defined:

- **Naturally occurring nanomaterials**: e.g. gas-phase condensation products, ashes, minerals, colloids,
- **Incidental nanomaterials,** e.g. by-products of human activities like ultrafine particles from high-temperature processes such as combustion and industrial processes and
- **Engineered or manufactured nanomaterials (ENMs)**, which are intentionally produced nanomaterials.

Although definitions of nanomaterials have just recently been coined, ENMs have been produced even prior to the development of the nanotechnology concept, for instance silica dioxide (SiO_2) (SASSI, 2008), silver (Ag) (Nowack et al., 2011) and carbon black (CB). Therefore, in some cases it may be difficult to say whether a given product contains nanomaterials or not. In some of these materials, e.g. SiO_2 or CB, primary nanoparticles of less than 100 nm are fused together to aggregates of more than 100 nm. Because the EU definition is based on the primary particle size, these materials may now be considered nanomaterials although they have been used as "conventional" materials for many decades.

4. Literature review on nanotechnology in the construction sector

In this chapter we provide an overview of nanotechnology in construction. In the first section we describe the driving forces behind nanotechnology and provide a list of applications, whether they are currently available or not into the market. In the second section, we provide a brief description of those that have been introduced into the market.

4.1 Introduction

The construction sector demands products like cement, steel, paints, insulation materials, window glass and many others. Nanomaterials are incorporated into those products to enhance their properties or to develop novel functionalities. The quest for sustainable development and the construction of green buildings in architecture also influences the innovation of products that exploit nanomaterial properties. Therefore it can be said that nanomaterials are used either

(i) to improve the quality of existing products, or
(ii) to innovate by developing novel functionalities in conventional products or through the creation of new products and applications.

Nanotechnology applications in the construction sector range from paints and coatings to solar cells. However, most of them have not reached a large commercial scale production, rather representing only niche segments of the markets (van Broekhuizen, 2009). In some cases, they have only being developed on a pilot scale or may only be found reported in the scientific literature as experiment results. Some reasons for the limited availability of nano-applications are

(i) their high prices compared to conventional products,
(ii) the uncertainty related to their safety, and
(iii) the uncertainty related to their technical performance (van Broekhuizen et al., 2011).

Outstanding properties observed at the nanoscale are intended to be introduced to construction materials. A special case are Carbon Nanotubes (CNT), which provide a strength 35 times superior to concrete, which make it suitable to be used in concrete structures (Hanus and Harris, 2013). So far it can be said that nanotechnology development could contribute to achieve some of the following ultimate goals (Lee et al., 2010; Teizer et al., 2012):

- Develop stronger concrete structures,
- Generate healthier environments,
- Protect the appearance and durability of structures,
- Reduce energy and other resources consumption.

The nanomaterials and their applications in construction that have been reported until now are shown in Table 1. Not all of them are currently available in the market. However, it is important to compile them so the reader becomes informed about all potential and actual applications, including the functionalization that is pursued.

Table 1. *ENM-based applications in the construction sector, ENMs used and properties or functionalities introduced. Sources: (Hanus and Harris, 2013; Kaiser et al., 2013; Lee et al., 2010; van Broekhuizen, 2009).*

Application	ENM	Introduced functionality or property
Cement	Al_2O_3, CNT, CNF, Fe_2O_3, SiO_2, TiO_2, ZrO_2	Improved structures durability Increased strength Abrasion resistance Air-purifying surfaces
Ceramics	CNT, SiO_2	Easy cleaning surfaces Enhanced mechanical and thermal properties Fire resistance
Coating	Ag, CeO2, CNT, Fe_2O_3, SiO_2, TiO_2, ZnO, carbon fluorine polymers, nanoclays	Biocidal activity Easy-cleaning properties Fire retardant Non reflection UV protection Anti-corrosion Wood preservation
Insulation material	SiO_2 (aerogel)	Glass insulation, Glass IR protection Heat losses reduction Noise barrier
Paints	Ag, SiO_2, TiO_2, ZnO, Cu	Easy-cleaning properties Biocidal activity Scratch resistance Air purification
Steel	Cu	Weldability; corrosion resistance; formability
Solar cells	CNT, Si, TiO_2	Effective electron mediation Glass glazing (heat losses reduction)
Windows	SiO_2, TiO_2, W_2O_3	Easy-cleaning Anti-reflection Anti-fogging Heat losses reduction
Structural monitoring	CNT	Nano electro-mechanical Systems (CNT-based NEMS)

4.2 Current applications of ENMs in the market in Europe

According to (van Broekhuizen et al., 2011), the main nano-applications in construction currently available in the market are cement, paints and coatings, and insulation materials. The same authors, based on a 2009 survey, claimed that nano-TiO_2, ZnO, Aluminum Oxide and SiO_2 are predominant in construction materials, and found no evidence for the use of CNTs. In this section we provide a brief description of those applications and the properties that ENMs impair to them.

4.2.1 Cement

Cement is a binder which is formed by the combination of silicates and carbonates. There are several types of cement, but the most used one is the Portland cement. It is produced by heating limestone (calcium carbonate) with small quantities of other materials in a kiln at temperatures above 1,000°C, in a process known as calcination. The resulting substance, called clinker, is ground with gypsum. Portland cement is the basic ingredient of concrete. Besides concrete, the production of mortar in masonry is other of the most common applications of cement (BCF, 2012) (PCA, 2014)

Nanomaterials are used in cement mainly to improve the quality and longevity of structures (Hanus and Harris, 2013). Nanoparticles can accelerate chemical reactions during initial hydration, leading to a more compact microstructure, decreasing permeability and improving mechanical properties such as compressive and flexural strength, and abrasion resistance, as a result of an improved interfacial bonding between the hardened cement paste and the aggregates (Raki et al., 2010). ENMs in cement can reduce corrosion and degradation of reinforcing steel and result in a lighter and more elastic concrete. Nanomaterials tested in cement include SiO_2, Al_2O_3, Fe_2O_3, ZrO_2, CNT and CNF (van Broekhuizen et al., 2011) (Hanus and Harris, 2013).

Only silica fume and nano-TiO_2 have been successfully incorporated into cement and commercialized (van Broekhuizen et al., 2011). Silica-fume is an aggregate of amorphous SiO_2 nanoparticles in the micro range (i.e. > 100 nm) and its application in cement seems to be a successful niche. The addition of 4 wt% of nano-Si resulted in compressive, split tensile and flexural strengths 1.7, 2.2 and 1.6 times higher compared to the non-nano version (Nazari and Riahi, 2011; Sanchez and Sobolev, 2010). Rough estimates in the EU consider that silica fume ultra high performance concrete (4 wt%) make up for 5% of the concrete market (van Broekhuizen, 2009). TiO_2 is mainly limited to bi-layer systems (i.e. when a second layer of concrete is laid over an original one (Zabulionis, 2005); the layers are made of: timber-concrete, concrete-steel (Zheng, 2013))[1] and in most cases microcrystalline particles (>100 nm) are used (van Broekhuizen et al., 2011). It should be noticed that the production process and the special equipment required to handle the nano-cement makes it more expensive than conventional one (Sanchez and Sobolev, 2010).

Photocatalytic properties of some nanomaterials are also exploited in cement-based applications (Hanus and Harris, 2013). They are used to create air-purifying and self-cleaning surfaces. Photocatalytic nanoparticles include TiO_2 and ZnO. The interaction of these particles with UV light creates free radicals which are capable of decomposing a wide range of microorganisms and organic pollutants that freely move in the air, including viruses, bacteria, fungi, NOx, SO_2, CO, C_6H_6 and other VOCs (Volatile Organic Compounds) (Chen and Poon, 2009). Photocatalytic surfaces can reduce the likeliness of spread diseases in hospitals and care centers, reduce atmospheric pollutants in cities and reduce the maintenance costs of concrete structures (Hanus and Harris, 2013). Nano-TiO_2 properties can also be

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[1] Bi-layer concrete systems are formed when a second layer of concrete is laid over an original one. Bi-layer systems may also refer to layer of different materials: timber-concrete, concrete-steel, for example Zabulionis, D., 2005. Stress and strain analysis of a bilayer composite beam with interlayer slip under hygrothermal loads. Mechanika 6, 5-12.. Bi-layer concrete surfaces are discussed by Zheng et el. (2013) in the context of pavement application.

exploited to enhance concrete's durability and to maintain whiteness throughout the entire lifetime of the construct (Raki et al., 2010).

Self-cleaning behavior of photocatalytic surfaces is also associated to the superhydrophilic properties of photocatalytic materials (Kazemi and Mohammadizadeh, 2012). This property is not only exploited in cement-based products, but also in windows and other surfaces (Hanus and Harris, 2013). Since superhydrophilic behavior is photoinduced, such films can be unstable in environments with low UV irradiation (Kazemi and Mohammadizadeh, 2012).

Cement applications include (reinforced) concrete, blocks, tiles, bricks, masonry mortar, pipe coatings, floors, pavements, roof tiles and many others (PCA, 2014). Reported applications of nano-cement include reinforced concrete for building structures (TiO_2 and silica fume) and road pavements in Italy, Belgium, England and the Netherlands (Hanus and Harris, 2013). Application techniques of photocatalytic materials in concrete road pavement include (i) application of a thin, photocatalyst-containing cement layer where the photocatalyst is added as a powder or a suspension during cement making; (ii) application of a photocatalyst-containing solution onto the concrete; or (iii) application by sprinkling the photocatalyst onto the fresh concrete.

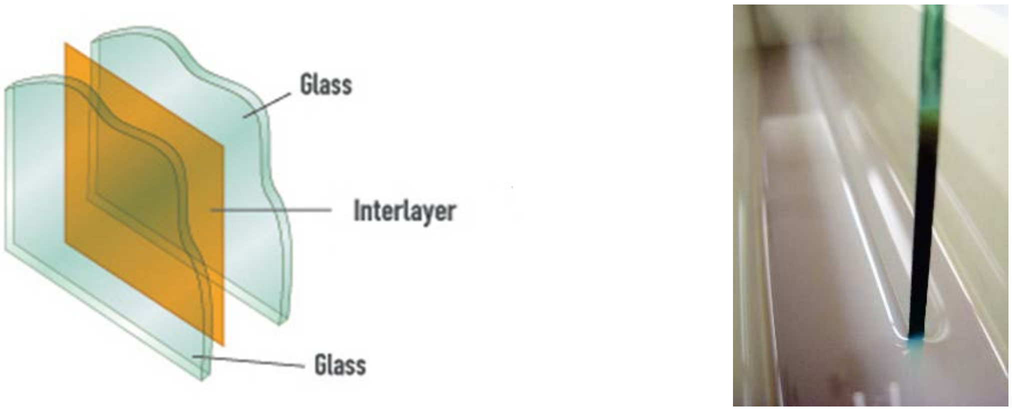
CNTs and Carbon Nanofibers (CNF) have also been tested in cement (Hanus and Harris, 2013). Improvements reported include an increase of 50% in compressive strength, 600% increase in Vicker's hardness and 200% increase in Young's modulus. However, there is no evidence of CNT-based cement available in the market mainly to its high price compared to the non-nano version, and the difficulty to achieve a homogenous dispersion of CNT throughout the cement paste, creating weak zones or zone of big stress which could result in structural deficits (van Broekhuizen et al., 2011) (Hanus and Harris, 2013).

4.2.2 Paints and coatings

The paint and coatings industry is one successful example for adopting nanotechnology (Evans et al., 2008; Kaiser et al., 2013) . Although there is an ongoing debate to determine whether or not the expected benefits of nanomaterials in these applications materialize in the long run (Kaiser et al., 2013), it is suggested that today they represent the biggest niche of nanotechnology in construction (van Broekhuizen et al., 2011) (NanoHouse, 2013).

The difference between paints and coatings may be subtle. However, making a distinction between them is useful to differentiate the properties that they are able to impair to surfaces. A coating is anything that covers a surface or substrate with a functionalization or decoration purpose. A paint is a coating type which is used mostly for decoration purposes (BCF, 2012). For instance coating is a more generic term that includes paints. The primary property of a coating is to be protective. Paint has the additional property of color along the protection (CQ, 2014).

It can be assumed that materials such as windows, solar panels and flooring materials use rather a coating containing ENMs than the material in the bulk. Especially in the case of windows a very thin interlayer -sometimes in the nano range- is used (Figure 2a) between the glasses for acoustic or thermal protection (UIUC, 2012). Also coatings containing nanomaterials can be applied on glass surfaces (Figure 2b). For instance a coating containing nano TiO_2 for self-cleaning effect could be also used for windows (the nanomaterial link itself with the glass and creates a hydrophobic surface) (FFF, 2014).



a. **b.**

Figure 2. a. *Thermo-acoustic interlayer for heat absorbing windows. Source: Nanohouse initiative Australia (UTS., 2002).* ***b.*** *Immersion bath for the deposition of pro.Glass Barrier 401 (special coating containing nano-ZnO) on a glass panel (Öko-Institut e.V., 2012).*

Coatings exist for a variety of purposes like protecting wood from being colonized by microorganisms, eliminating statics in fuel and electronic systems, avoiding biofouling in ship hulls, creating photocatalytic surfaces (pavements, walls and glass), preventing pipes from oxidizing, and many others (van Broekhuizen, 2009), (De Volder et al., 2013). In construction, paints and coatings are applied mainly on mineral surfaces (plaster, concrete, bricks, stone, tiles), wood (solid wood, panels, and particle or fiber boards), metal (mainly steel sheets) or glass. By definition, more properties can be impaired to surfaces through coatings than paints, as can be seen in Table 2.

Table 2. *ENM-based properties in paints and coatings used in the construction sector. Sources: (Lee et al., 2010), (van Broekhuizen et al., 2011), (Hanus and Harris, 2013), (Kaiser et al., 2013).*

Paints	Coatings
Biocidal activity	Biocidal activity
Easy cleaning properties	Easy cleaning properties
Scratch resistance	Scratch resistance
UV protection	UV protection
	Fire protection
	Non-reflectiveness

From a risk assessment perspective, the most relevant property listed in Table 2 is the biocidal activity. Easy cleaning property of some nanomaterials has already being described in the cement application subsection. The remaining ones will not be reviewed here.

Antimicrobial mechanisms of nanomaterials that generate the so-called biocidal activity is related to one or both of the following mechanisms (Chen and Poon, 2009): (i) the nanomaterials directly interacts with microbial cells affecting one of its component, killing it or avoiding reproduction. And second (ii), indirect interaction via the production of secondary products (reactive oxygen species or dissolved heavy metal ions) which cause some of the formerly mentioned interventions. Biocidal activity in paints and coatings may be used to create healthier environments, to avoid propagation of diseases, to contribute to keep the appearance of surfaces and reduce their maintenance costs (Hanus and Harris, 2013).

Metallic and metal oxides ENMs reported to have antimicrobial activity include Ag, TiO_2, ZnO, Cu, CuO, and MgO (Hanus and Harris, 2013). Bulk Ag has been used since ancient times for its biocidal effects (Nowack et al., 2011). Effective use of photocatalytic particles as antimicrobials is limited to environments where there is sufficient irradiation with light of the required wavelength, although incidental radiation from natural or some artificial light sources could be sufficient for TiO_2 activation (Markowska-Szczupak et al., 2010). Antimicrobial effectiveness in actual paints also depends on paint formulation, like total pigment volume concentration and type of acrylic dispersion used. Additionally, the morphology of the surface may influence biocidal activity (Hochmannova and Vytrasova, 2010). Currently, Ag, TiO_2, SiO_2 and ZnO nanoparticles are used in commercially available paints (van Broekhuizen, 2009)

Treated wood is a successful niche of nanotechnology, at least in the USA (Evans et al., 2008). The market for treated wood in North America is valued in $4.9 billion (gross sales) and every year around 20 million m^3 are treated with aqueous, mainly copper-based preservatives. Wood preservatives consist of copper carbonate particles and an organic co-biocide, both dispersed in water. New nano-copper preservatives compete with dissolved or complexed (non-nano) copper salts.

4.2.3 Insulation material

ENM-based insulation materials for buildings and houses can contribute to diminish greenhouse gas emissions by reducing energy consumption. Besides providing superior comfort, applying modern and efficient insulation would lower energy consumption and thus reduce global CO_2 emissions (Baetens et al., 2011). In 2005, buildings emitted more than 30% of global greenhouse gases in many developed countries, which was directly associated to heating and cooling purposes. New insulation materials based on SiO_2 have been recently developed. Often called Aerogel, "blue smoke" or "frozen smoke", SiO_2 has been used in the past in aerospace applications (Bheekhun et al., 2013). However, it has been implemented for insulation purposes to the construction sector only recently.

Silica aerogel consists of cross linked internal structure of SiO_2 nanoparticle chains that create air nanopores smaller than 100 nm (Baetens et al., 2011). It is considered the lightest solid material on the planet, possessing a high compressive strength, typically around 2,200 Kg/m^3 (Bheekhun et al., 2013). It is an excellent insulator because both the silica matrix and the filling air pores are poor heat conductors (Baetens et al., 2011). Currently available silica-based insulators have thermal conductivity as low as 0.027 W/mK (Hanus and Harris, 2013).

Applications in the construction sector include wall and windows insulation, which is positive if we consider that up to 60% of total energy lost from a building takes place through its windows (Jelle et al., 2012). Compared to conventional window glazing, silica aerogel may reduce heat losses between 55 and 80% (Hanus and Harris, 2013). Aerogel is ideal for windows insulation due to its high transparency.

Aerogel glazed windows were firstly introduced to the market in 2006, encapsulating granular aerogel between polycarbonate construction panels (Hanus and Harris, 2013). As long as aerogel is a very brittle material, the current maximum size of crack-free monolithic aerogel tile is about 0.6 m^2. Despite its potential, currently availability is rather limited. Some companies working in the development of these applications include Aspen Aerogels and Cabot Corporation (USA), Airglass AB (Sweden) and Fixit (Switzerland) (Baetens et al., 2011; Hanus and Harris, 2013). Other applications in construction of aerogel include sound barriers and insulation of solar collectors.

5. Construction and demolition waste in Switzerland

To investigate the flows of ENMs in the construction waste in Switzerland and their potential release, we first describe the disposal and recycling processes as they work nowadays according to the regulations in Switzerland. Construction and demolition waste (C&DW) is the result of changes in Switzerland's building stock and infrastructure (i.e. buildings –Hochbau-, and roads/underground constructions –Tiefbau-). C&DW also includes hazardous construction waste. However, C&DW from roads and underground construction (Tiefbau) and hazardous construction waste are not considered is this report; all results refer to C&DW from "Hochbau" activities.

5.1 Construction waste from buildings (Hochbau)

The quantities of C&DW shown here are for the year 2012 and are based on models developed according to the simulation of construction processes and the relevant material flows (Wüest & Partner (FOEN, 2001), (FOEN, 2008)). The C&DW is classified into different categories and each category compiles a set of different materials (Table 3).

***Table 3**. Construction and demolition waste categories (FOEN 2001, 2008)*

C&DW group	Materials
Road construction waste	Sand, gravel. Road construction waste
Asphalt	Asphalt
Concrete demolition	Concrete, cement[1]
Mixed demolition	Breakage, brick masonry. Artificial, natural stone (façades). Architectural membrane[1], coating & paint[1]
Combustible waste	"New" insulation[2], "old" insulation, plastics, textiles, taper, packaging, adhesives[1], sealants[1]
Wood	Construction, finish and residual wood
Metals[3]	Steel, light metals, aluminum, other metals (including steel)[1]
Mineral fraction	Roofing materials, ceramic, stoneware, slag, gypsum, plaster, glass, windows[1]
Mixed C&DW	Backfill (with construction waste), flooring material[1]

[1] Applications added from Survey on ENM in construction sector in Switzerland
[2] "New insulation materials" include both polystyrene as well as mineral wool, foam glass, etc.
[3] Includes iron armoring

Table 4 shows the waste produced in Switzerland according to the general C&DW categories and the disposal data for the different waste treatments. Not for all the materials mentioned in Table 3 data were available.

Table 4*. Source and fate of construction waste (Hochbau) in Switzerland in 2012, by C&DW group. Source: Author's extrapolations using the data available in (FOEN, 2008). Values in 1'000* ***metric tons (t)****.*

C&DW group	**By source[1]**				**By fate[2]**			
	Demolition	**New**	**Renewal**	**Total**	**Direct Recycling[3]**	**Recycling (REC)**	**Waste Incineration Plant (WIP)**	**Landfill (LF)**
Road construction waste	354	97	553	1'004	860	119	0	25
Asphalt	104	9	45	158	109	41	0	9
Concrete demolition	322	116	1'197	1'635	0	1'471	0	163
Mixed demolition	422	47	1'084	1'553	0	1'192[4]	0	361
Combustible waste	126	28	14	168	0	47	108	13
Wood	245	53	117	415	0	42	361	13
Metals	264	15	75	354	0	347	0	7
Mineral fraction	788	36	242	1'066	0	33	0	1'033
Mixed construction waste	1	0	103	105	0	0	26	78
Total	**2'628**	**401**	**3'429**	**6'458**	**968**	**3'292**	**495**	**1'703**

[1,] Extrapolation for the year 2012 based on the data given in the reports from Wüest & Partner (FOEN, 2001) and (FOEN, 2008), applying an annual average growth rate of 1.8% to the amount of residues (page 11, FOEN 2008).

[2] Unlike FOEN (2001), the report elaborated in 2008 (FOEN 2008) does not provide the values disaggregated by fate. To estimate the amount in each possible fate, we applied the following back-forward method:

1. We took the original values by source, i.e. demolition, new renewal and total (Table 6, page 92) of 1997 from FOEN (2001), and disaggregated them by fate (as in Tables and Figures, pages 102 – 103, same report).
2. We extrapolated the values from 1997 to 2008 using the annual average growth rate of 1.8% (in this case for 11 years).
3. We checked that the values matched with the information of 2008 in FOEN (2008). They were confirmed (Table 8.6 and Figure 6.2, FOEN 2008).
4. We extrapolated the values to 2012 using the same annual growth rate shown in FOEN (2008), i.e., 1.8% and the values match with Figure 6.1, page 26 (FOEN 2008).

[3] Direct recycling refers to the waste that is immediately re-used on the demolition (construction) site.

[4] On a first stage, the residues enter the recycling system. On a second one, a fraction of them may be transferred abroad (exported) for further processing (FOEN, 2012).

In 2012, most of the C&DW disposed in Switzerland was recycled (51%) or landfilled (26%). Only 8% was incinerated and 15% was re-used on-site (Table 4). This distribution varies depending on the specific material considered. According to the report from Wüest & Partner (page 92-103) (FOEN, 2008), no construction and demolition waste was exported to be treated abroad. However, based on (FOEN, 2012) we know that 37'000 t of mixed demolition were exported for recycling processing, that is 3% of the recycled mixed demolition waste (see flows in Chapter 8).

For the purpose of this report, we describe the fate of the materials which are covered with paints and coating (Table 5). Although these applications are separately assessed in Chapter 8, the fate of cement applications within the system is also included in Table 5.

The fate of paints and coatings with ENMs in construction waste depends entirely on the material where they are applied. Paints and coatings in construction are usually applied in mineral and concrete surfaces, wood and window glass. For the sake of completeness we also consider metallic surfaces (ferrous and non-ferrous separately), plastics and stone surfaces. Our primary source of information is the already mentioned report by (FOEN, 2008).

The transfer coefficients in Table 5 represent the fate of different applications with ENMs during the end-of-life stage of their life cycle and will be used later to determine the flows of ENMs within the Swiss system (Chapter 8). Cement and gypsum plasters are used to cover interior and exterior walls of buildings, which are usually coated with paints in succeeding stages. These materials correspond to the category "Mixed demolition" in Table 3. We calculated the coefficients by dividing the amount of waste in this category by the total amount of waste (Table 4). The numbers are then expressed in percentage in Table 5.

The same was done for concrete (both, paint and cement applications), wood, window glass, plastics and stone surfaces, which correspond to the categories "Concrete demolition", "Wood", "Mineral fraction", "Combustible waste" and "Mixed demolition" of Table 3 respectively. The resulting transfer coefficients for windows glass and concrete were corroborated through personal communications with experts of ETH Zurich and Eberhard Bau AG. Meanwhile, for metallic surfaces, we used the information provided by (Caballero-Guzman et al., 2015), who analyzed the fate of metals in the context of waste recycling in Switzerland. For concrete pavement, the data for "Concrete demolition" was also used.

Table 5. *Fate of construction and demolition waste from the most relevant applications used in the Swiss construction industry.*

Applications[1]	**Fate %**			
	REC	**WIP**	**LF**	**Exported**
Paint and coatings				
In stone surfaces (walls or façades) also cover with plaster	77	0	23	0
In concrete	90	0	10	0
In wood	10	90	0	0
In window glass	3	0	97	0
In ferrous metals	98	0	2	0
In non-ferrous metals	0	0	2	98
In plastics	28	64	8	0
Cement				
Concrete	90	0	10	0
Pavement	90	0	10	0

[1] Sources: Table 4 and (Caballero-Guzman et al., 2015). We calculated the coefficients dividing the amount of waste in this category by the total amount of waste (Table 4). The numbers are then expressed in percentage.

5.2 Final disposal of construction waste

In Switzerland the final disposal of construction waste is regulated by the technical ordinance on waste (TVA). Combustible parts of construction waste must be incinerated. Mineral construction waste is mainly either re-used on-site or treated and transformed to recycling products (Table 4). The residues generated during recycling and treatment processes of C&DW as well as overcapacities of construction waste are further disposed using the following facilities:

- **Landfills for inert materials** like construction wastes. In this facility no leachate is expected.
- **Landfills for stabilized residues**, i.e. solidified ash, flue gas cleaning materials. Leachate production is expected.
- **Bioactive landfills** receive incinerator slag. Gas and leachate production is expected.

If ENMs are present in the construction waste flows, a potential for release is possible due to the leachates and dust emissions from the above mentioned facilities. The major flows of ENMs from incineration plants (WIP) to the landfill are with the bottom ash. But if ENMs are enclosed into larger (vitrified) fragments of bottom ash, they may not be released any more (Müller et al., 2012). However, a release potential may exist if ENMs are loosely attached to larger particles after incineration (Walser, 2012).

For this report we consider the ENMs in hazardous construction waste as negligible because of the especial treatments this kind of waste received (i.e. in case of ENMs entering in the hazardous waste, they will be eliminated). We rather consider ENMs in conventional C&DW flows from buildings.

6. Market survey

The goal in this chapter is to get an overview of the use of nanomaterials in the construction industry in Switzerland based on a market survey performed among Swiss construction experts. We describe the methods used to carry out the survey and then present the results obtained and discuss them. We also evaluate additional market results available from European sources (e.g. (ANSES, 2013)) about the use of nanomaterials. With the evaluation these results we will achieve a better understanding of the flows of ENM in the construction and demolition waste (Chapter 8).

6.1 Survey of Swiss construction industry

The main goal of the survey was to obtain the feedback and experience from experts in the Swiss construction sector in terms of the use of nanomaterials in the construction industry in Switzerland.

6.1.1 Methods

The internet platform SurveyMonkey (www.surveymonkey.com) was used to design the survey, it was carried out between 12th February and 13th March 2014. The nanomaterial definition from the European Community (EU-Commission, 2011) was given to the industrial experts. The survey was sent to 60 experts from the construction sector. It consisted in a questionnaire of 5 questions about the estimates of ENM in the Swiss construction industry (i.e. use of nanomaterials, market size and share and amount of ENM used in the different construction materials). The final part of the survey was dedicated to further contact and comments from the experts to the survey.

The questions were related to the following nanomaterials: TiO_2, SiO_2, ZnO, Ag, CuO, $CuCO_3$ and to the following materials used in the construction sector: Adhesives, architectural membranes, asphalt, cement, coating, flooring material, geosynthetic barriers, insulation material, paint, pipes, sealants, solar panels, steel, other metals, windows and wood. The questions of the survey were:

1. According to your knowledge, are ENM used in the following products in Switzerland?
2. What is the annual market size of products containing ENM in Switzerland?
3. According to your knowledge of the whole market (including both, with and without ENM), what is the market share of products containing ENM in Switzerland?
4. Which ENM are used in the following products in Switzerland?
5. What is the mass content of ENM in the following products?

6.1.2 Results

A total of 18 experts answered the questionnaire (30% return). An evaluation of online surveys concluded that the response rates to such surveys vary greatly but most of them receive a 26% response (Hamilton, 2009), indicating that the response rate of this survey was good.

In relation to question 1: ***"According to your knowledge, are ENM used in the following products in Switzerland?"*** 94% (17 persons) of the respondents answered and 5% skipped the question. The majority of answers pointed out that in Switzerland ENM are most used in coatings and paints (78 and 72% respectively), followed by applications in solar panels (50%), flooring material (44%) and isolation material, sealants and windows (39%), see Figure 4.

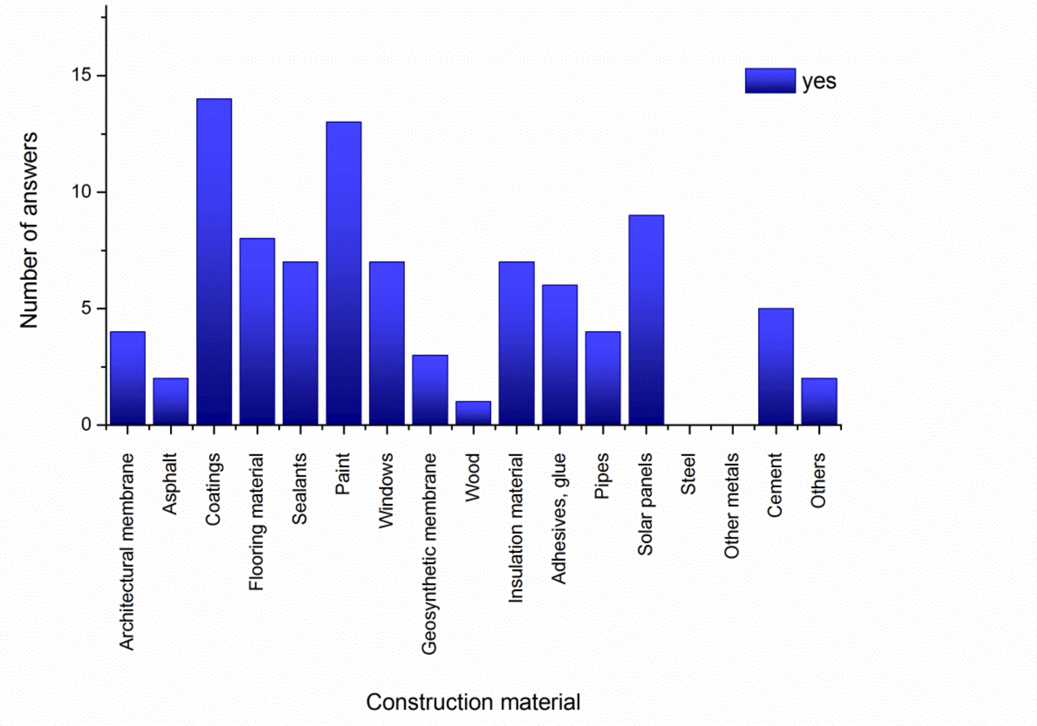


Figure 4. *Number of answers according to survey replies to the question 1: "**Are ENM used in the following products in Switzerland?"***

Figure 4, also shows that ENM are also used in adhesives (33%) and cement (28%). Other materials such as paving stones, floor slabs and products for garden development made of concrete were also mentioned.

In the question 2 "***What is the annual market size of products containing ENM in Switzerland?***", the participants could provide their answer in t, CHF and Euro or all of them, if they had the information. This question was answered by 22% (4 participants). In Figure 5 the answers of three respondents are shown.

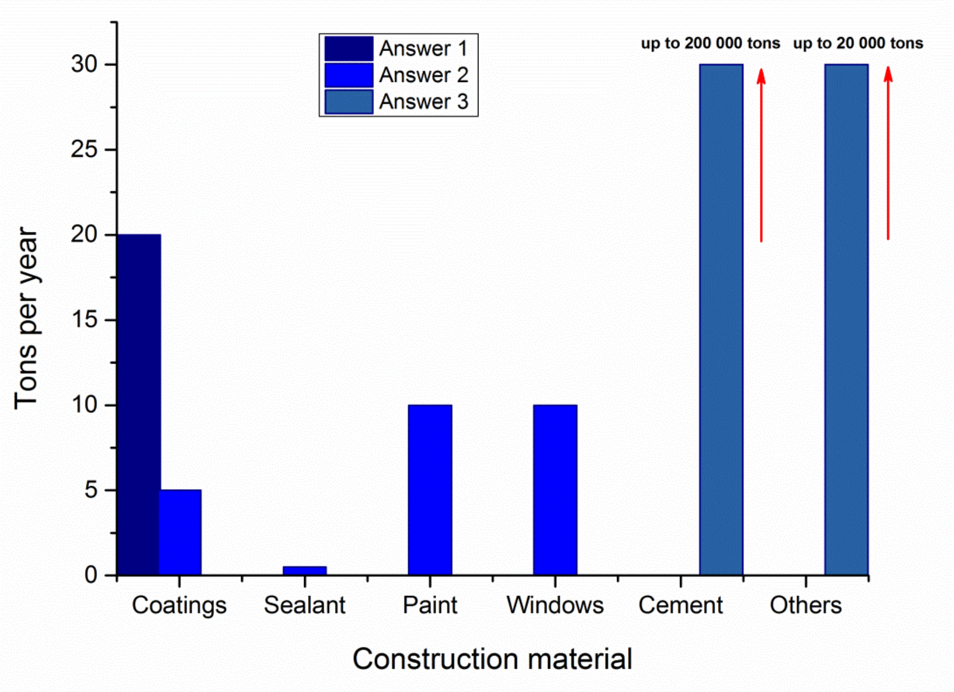


Figure 5. *Annual market size of products containing ENM in Switzerland based on the survey replies to the question 2: "**What is the annual market size of products containing ENM in Switzerland?"***

Participant number 1 reported an annual market size for coatings of 20 t/year; the second one reported 5 and 10 t/year for coatings and paints respectively, but also 10 t/year for windows and 0.05 t/year for sealants. The third participant gave values for cement up to 200'000 t/year and up to 20'000 t/year for other materials (but these other materials were not mentioned).

There was a last answer (participant number 4), who provided only the answer of zero (0) to those materials, which probably do not contain ENM yet or their market is unknown. These materials were architectural membranes, geosynthetic barriers and solar panels.

In question 3: ***"According to your knowledge of the whole market (including both, with and without ENM), what is the market share of products containing ENM in Switzerland?"*** the answers were provided in percentage (Figure 6). 28% of the participants answered this question.

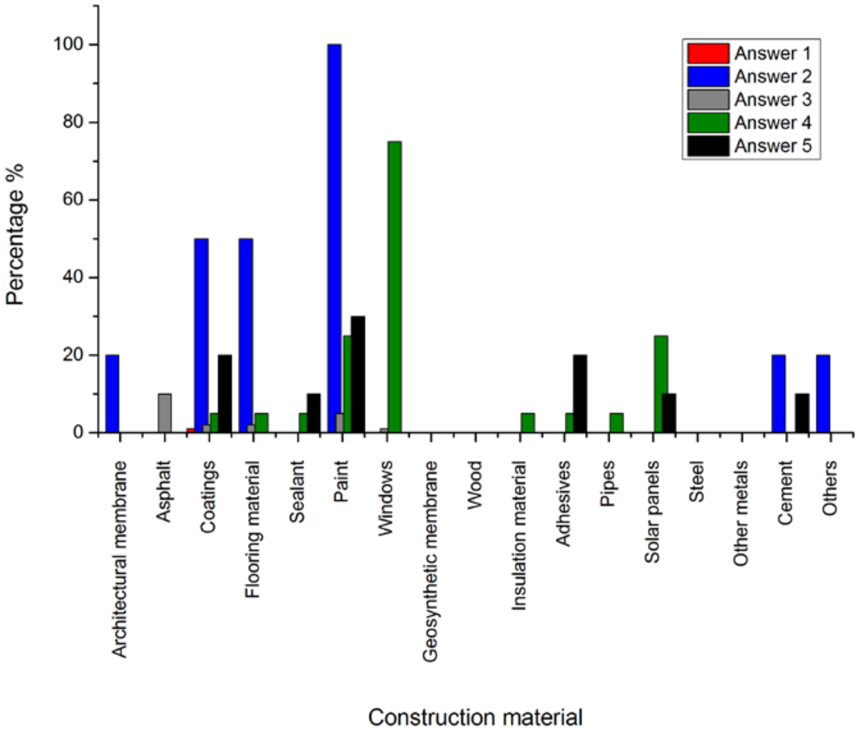


Figure 6. *Market share of construction products containing ENM in the construction sector in Switzerland. Based on the survey replies to the question 3: "**According to your knowledge of the whole market (including both, with and without ENM), what is the market share of products containing ENM in Switzerland?"***

Coatings and paints were the most mentioned material in this question about market size; these applications seem to share a big part of the market although the answers were very diverse. For instance participant 2 gave a value of 100% (i.e. all paints contain nanomaterials) while participant 3 provided a value of 5%. A similar result was observed for windows where participant 4 provided a share value of 75% and participant 3 a value of 1 % (see also the case of flooring materials in Figure 6). Other materials were also mentioned having a market share of around 20% (i.e. architectural membranes, adhesives, solar panels and cement).

Question 4: "***Which ENM are used in the following products in Switzerland?***" was answered by 17 % of the participants (3 respondents). Nano Ag was mentioned to be used for many products, even for architectural membranes, pipes and solar panels. The answers pointed out that all of the evaluated ENM are used in paints and again this is the most mentioned product. It is followed by flooring materials and windows, which use nano TiO_2, SiO_2 and Ag.

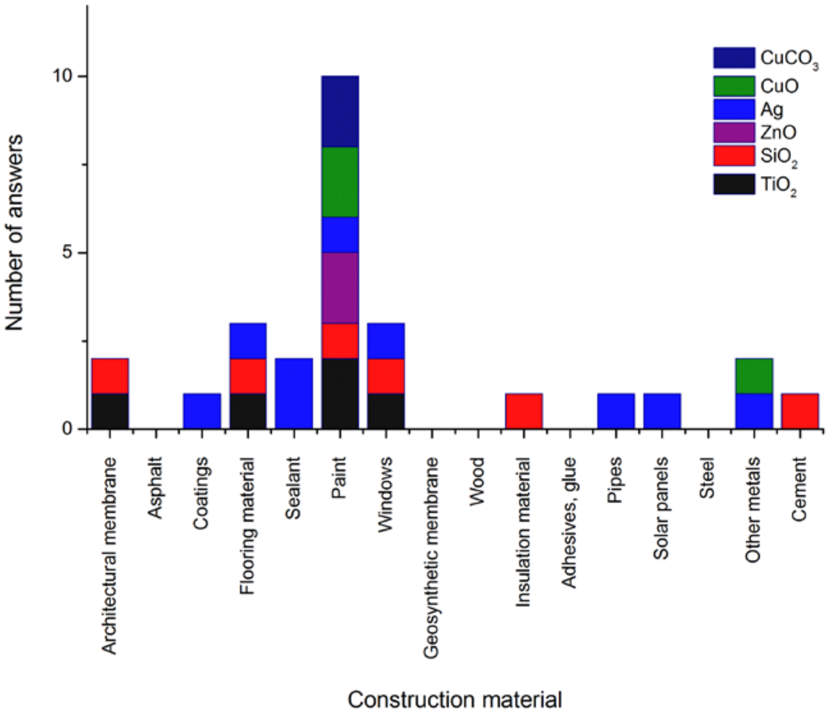


Figure 7. *ENM used in construction product in Switzerland. Based on the survey replies to the question 4: "**Which ENM are used in the following products in Switzerland?"***

Question 5: "***What is the mass content of ENM in the following products?***", was answered by 11 % of the participants (2 respondents). Figure 8 shows the little knowledge that is available among the experts about the amount of ENMs in the different products. However, the highest amount of ENMs in a product is given by nano SiO_2 (i.e. higher than 5 wt %) for cement. An amount of less than 1 wt % of nano Ag was reported for sealants. Between 1 and 5 wt % nano TiO_2 and SiO_2 was mentioned for other applications, but unfortunately these applications were not specified.

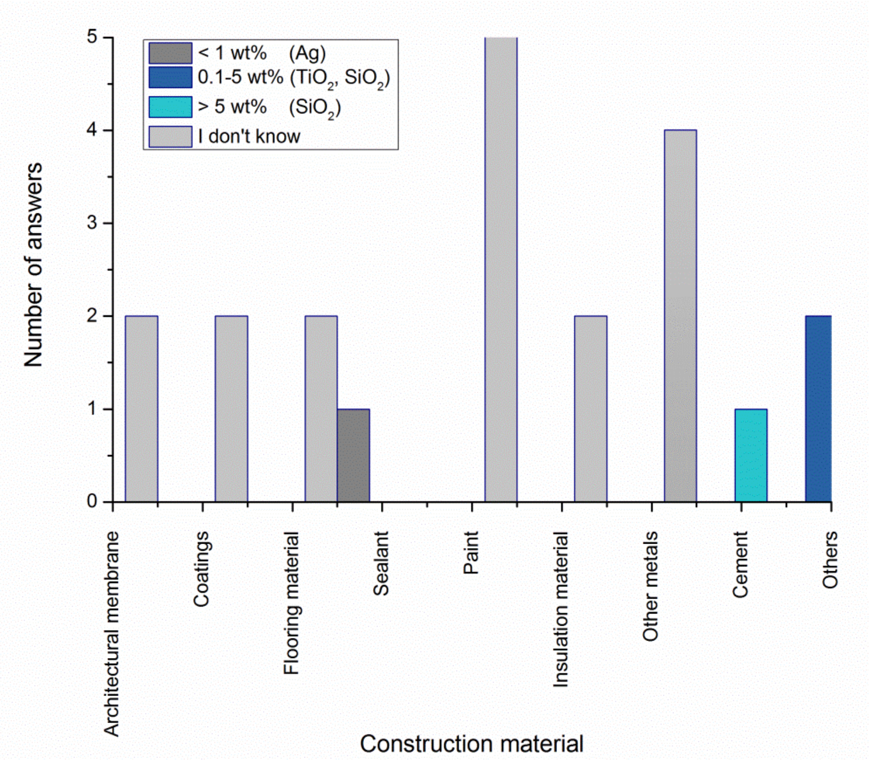


Figure 8. *ENM's mass content in construction materials, based on the survey replies to the question 5: "**What is the mass content of ENM in the following products?**"*

In the last part of the survey about contacts and comments, one participant was willing to provide a short interview to deepen information about ENM in construction materials. Four participants were interested in the final results of this survey. One comment was also received and it stated that:

"The use of nanomaterials in the construction sector is very critical because the costs compared to the benefits are usually very high, therefore their use for improving properties is usually not worth. Accordingly, the proportion of products containing nanomaterials in this sector is negligible. A meaningful way to use nanomaterials would be if they allow completely new uses or provide new properties (e.g. lotus effect for self-cleaning surfaces, but here the durability of a coating could be compromised)".

6.1.3 Discussion

The overall impression of the survey was that there is still a significant lack of knowledge about the uses of ENM, the market size and the amounts of ENM used in construction materials. This is evident by the few participants who were able to answer all questions and the diversity of their answers. On the other hand, the fact that only few participants answered some of the specific questions makes us confident to rely on their expertise for the construction sector they are managing.

Paints and coatings were the most mentioned construction products containing ENM. In the second place windows and solar panels were also reported. We presume that windows were mainly mentioned not because the window itself would be nano but rather the coatings containing ENM applied on the surfaces of the glasses or the interlayers place between the glasses of the window (see also Chapter 4).

Here the open question is whether these interlayers should also be considered as nanomaterial. Due to the mentioned issue about the uncertainty of application of coatings and or interlayers for windows, solar panels or even flooring materials, the answers to questions 1, 2 and 3 are very diverse. For instance the participants also mentioned that ENM are used in for windows, but it is unclear if the windows are **coated** with an ENM containing coating or if they rather have an interlayer in the nm range placed between the glasses. Nowadays almost all windows have this type of thermo-acoustic protection, probably that is the reason of providing a high market for windows "containing ENMs".

According to the survey, ENM are also used in adhesive, sealants, isolation materials and cement. Although in question 1, the use of ENM in cement was just mentioned in 28 % of the answers, in question 2 the size of the market seems to be very high (up to 200’000 t/y). From our point of view that could make sense because high amounts of cement are used to produce concrete for the different application in the construction sector. On the other hand if a material like SiO_2 is taken into account, then the market size is accordingly high, because this material is essential for the production of cement (see also question 4, where SiO_2 was mentioned to be used for cement). Another open question here would be whether this SiO_2 is actually in its nano form according to the review in Chapter 4.

Actual and accurate information of the uses and market of ENMs in the construction sector in Switzerland will serve to obtain better top-down and bottom-up results of waste flows. A declaration platform of ENMs, their uses and quantities might be one of the keys in order to cope with this lack of information.

6.2 Other sources for market information

6.2.1 ANSES report

According to the French Decree no. 2012-232 on the annual declaration on substances at nanoscale (MEDDTL, 2012), the French Agency for Food, Environmental and Occupational Health & Safety **(ANSES) has developed a portal dedicated to the declaration of these substances** (ANSES, 2013). The first attempt to collect the declaration on nanomaterials production and uses in France (i.e. information from producers, importers, distributors, users & distributors and remanufactures & distributors) was carried out between 1st January and 30th June 2013. During this period, a total of 670 French entities submitted at least one statement and 2’776 declarations were obtained. Carbon black (CB) and silica dioxide (SiO_2) were reported to be the most produced/imported substances in France in 2013 (274’840 t and 155'072 t respectively) (ANSES, 2013). For the interest of this study, Table 7 shows a summary for some materials found to be used in paint production and construction products.

Table 7. *Declaration of quantities of nanomaterial produced and imported in France for 2013 (uses in all industrial sectors) that are also used in the construction sector (ANSES, 2013)*

Nanomaterial		**Quantities produced and imported in France metric tons (t)**
Silica dioxide	**nano-SiO_2**	155'072
Titanium dioxide	**nano-TiO_2**	14'321
Zinc oxide	**nano-ZnO**	288
Copper pigments	**nano-Cu-Pigments**	100[1]

[1] According to ANSES report, the quantity material produced and imported for copper (which refers to pigments) in France varies from 100 Kg – 1 t, from 1 – 10 t and from 10 – 100 t. Here we choose the reported upper limit.

As given in the declarations, SiO_2 is the most used nanomaterial in the different applications related to the construction sector. Nano-Cu (from pigments) is most used for coatings, paints, solves and diluents. The declaration distribution of all nanomaterials produced, imported and used in France shows that almost all of the nanomaterials are also used in the paint and coating industry (Table 8).

Table 8. *Declaration distribution of reported materials and their uses in France. (ANSES, 2013)*

Applications related to the construction industry	**nano-Cu-Pigments**	**nano-SiO_2**	**nano-TiO_2**	**nano-ZnO**	**Distribution of reported declarations %**
Coatings and paints, solvents, diluent	xxx[2]	xxx	x	x	8.1
Buildings and construction works	x	xxx	x		1.4
Articles of stone, plaster, cement, glass and ceramics		xxx			0.5
Fillers, putties, plasters, modeling clay		x			0.5
Manufacture of other non-metallic mineral products e.g. plaster, cement	x	xx			0.5
Adhesives, sealants		xx			0.3
Metal articles	x	xx			0.2
Treatment of non-metallic surfaces		x	x		0.1
Biocides		x			0.1
Applications related to other industrial sectors[1]	n.a.	n.a.	n.a.	n.a.	88.3
Total					**100**

[1] For instance: Cosmetics and personal products, manufacture of chemical substances and vehicles, among others.

[2] The "x" means how often a nanomaterial was reported through the whole ANSES document according to its use (i.e. 3 "x": most reported for a specific application and 1 "x": less reported for a specific application.

n.a. means no data available

As shown in Table 8, SiO_2 and TiO_2 have the highest quantities of production and imports for the different uses (155'072 and 14’321 t respectively). These amounts are very high and it is difficult to know exactly which fraction corresponds actually to a new nanostructured or engineered nanomaterial (by design). An example of this situation is given for SiO_2, where the declared amounts of silica included: synthetic amorphous silica, silica gel, fumed silica, and silicon dioxide.

An interesting point was the absence of information about nano-silver. It was only declared for scientific uses (research and development) in a quantity range between 0.1 to 1 kg for the year 2013. This nanomaterial was not declared in any of the other sectors or applications. Taking into account results from a supply and demand study from the Silver Institute, France was one of the top ten importers of silver at global level with around 4.7 million ounces (Moz) or 146 t in 2010 (GFMS, 2011). This may mean that either nano-Ag is not a material that has any real use in applications or that it is imported in form of intermediate or final products (e.g. fibers, fabrics, textiles) that didn’t had to be registered.

Although the data of the ANSES register is rather limited due to the previously given reasons (i.e. many materials have probably by chance been taking into account as nanomaterial), the register represents the first approach to obtain ENM quantities and markets in one country.

Because no similar data are available for Switzerland, an adjustment of the amounts from France for Switzerland was carried out (Table 9). The adjustment was done using a conversion factor of 0.12 based on the population and of 0.17 based on the gross domestic product at purchasing power parity (GDP PPP). The GDP PPP means the value of all final goods and services produced within a country in a given year, divided by the average (or mid-year) population for the same year (World-Bank, 2012). For our purposes, the GDP PPP provides a better approximate to the reality than only using the population or the gross domestic product (GDP) of a country. For this reason and for further estimations (i.e. Chapter 8) we used rather the data given by the GDP PPP.

Table 9. *Adjustment of quantities of ENM produced and imported in Switzerland according ANSES report*

Nanomaterial		Quantities produced and imported in Switzerland (t)	
		Population[1]	GDP PPP[2]
Silica dioxide	**SiO_2**	19’021	26'362
Titanium dioxide	**TiO_2**	1’757	2'435
Zinc oxide	**ZnO**	35	49
Copper pigments	**Cu-Pigments**	12	17

[1] Provisional population in Switzerland at the end of 2012 (i.e. 8 M persons), (FSO, 2013).

[2] Gross domestic product at purchasing power parity GDP PPP for 2012 (i.e. 417 international dollars) per capita (World-Bank, 2012).

Besides the ANSES register, in June 2012 the **German Federal Environment Agency (UBA)** published "Concept for a European Register of Products Containing Nanomaterials" (UBA, 2012), it is based on the costs, benefits for all actors associated with such a register and the environment. The study concludes that the creation of a horizontal European register of products containing nanomaterials, which is built on present substance- and product-related regulations, is preferable to a separate register. In general it is recommended to acquire a better organization of the data, including for instance, mass content, market size of the nanomaterial in the specific product and sector. Also a differentiation between real nanomaterials and conventional materials should be clarified (e.g. silica and its nano functionalized version).

Although there is still a lack of information about applications of ENMs and their market, the results from the Survey and the analysis of the ANSES report helped us to visualize the behavior of nanomaterials in the Swiss construction sector and therefore ENMs in the C&DW flows.

7. Nanomaterial flows in construction waste

In this section we develop the theoretical framework to analyze the flows of nanomaterials in construction waste. We do it by describing the life-cycle of nanomaterials used in construction and apply the material flow framework to visualize the hypothetical flows and stocks within the Swiss system (Section 7.1). We then briefly define the approaches that can be implemented in practice to estimate the current flows of nanomaterials: top-down and bottom-up approaches (Section 7.2).

7.1 Theoretical framework

Life cycle is generally understood as the "life-span covering production, use and disposal of a material, chemical or product" (Som et al., 2010). The life cycle of an application used by the construction sector is as follow:

1. ENM is produced;
2. ENM is integrated into applications (i.e. paints and coatings or cement) used in the construction sector;
3. A nano-application is installed in a construction (i.e. building);
4. After some time of use, the application becomes waste due to the following reasons:
    a. When a building is demolished,
    b. During reparations taking place at buildings (e.g. broken windows or broken pipes),
    c. When a building is partially or completely renewed;
5. The waste is treated according to the regulation of a country. In Switzerland, the waste is incinerated, recycled, landfilled or exported;
6. If the waste goes into recycling, the fate of ENM depends on the material fraction where the ENM is allocated. During recycling, some ENMs can be eliminated or released to the air or to the water as wastewater, depending on the technical characteristics of the recycling process. The non-recyclable fractions are either disposed of or incinerated;
7. If the material is recovered, it is used once more as raw material for the production of an application (not necessarily in the construction sector);
8. If the material is disposed of into a landfill, then it can be considered to be finally stored in this compartment. Some transformations are possible depending on the type of facility. In Switzerland three types of landfills exist: (ISO.) for inert materials, (ISO.) for stabilized residues, and (ISO.) bioactive landfills (Müller et al., 2012) (see chapter 5). Some releases to air or transfer to soil or groundwater are possible;
9. If the material is incinerated, then the ENMs are transferred to the slag or the filters ash. ENMs could be released to the atmosphere if the filters are not efficient (but in Switzerland this is not the case due to a filter efficiency of at least 99.99% (Müller et al., 2012)), or transferred to recycling facilities where the metallic residues that will be recovered (Sun et al., 2014). ENMs can also be eliminated as a result of a chemical or physical transformation. Elimination in this context means that the material is not any more in the nano-range. This could occur for example if CNTs are burnt or a metal nanoparticle is dissolved.

We assessed the ENMs flows from the construction industry qualitatively in the Swiss system by means of a Material Flow Assessment (Figure 9).

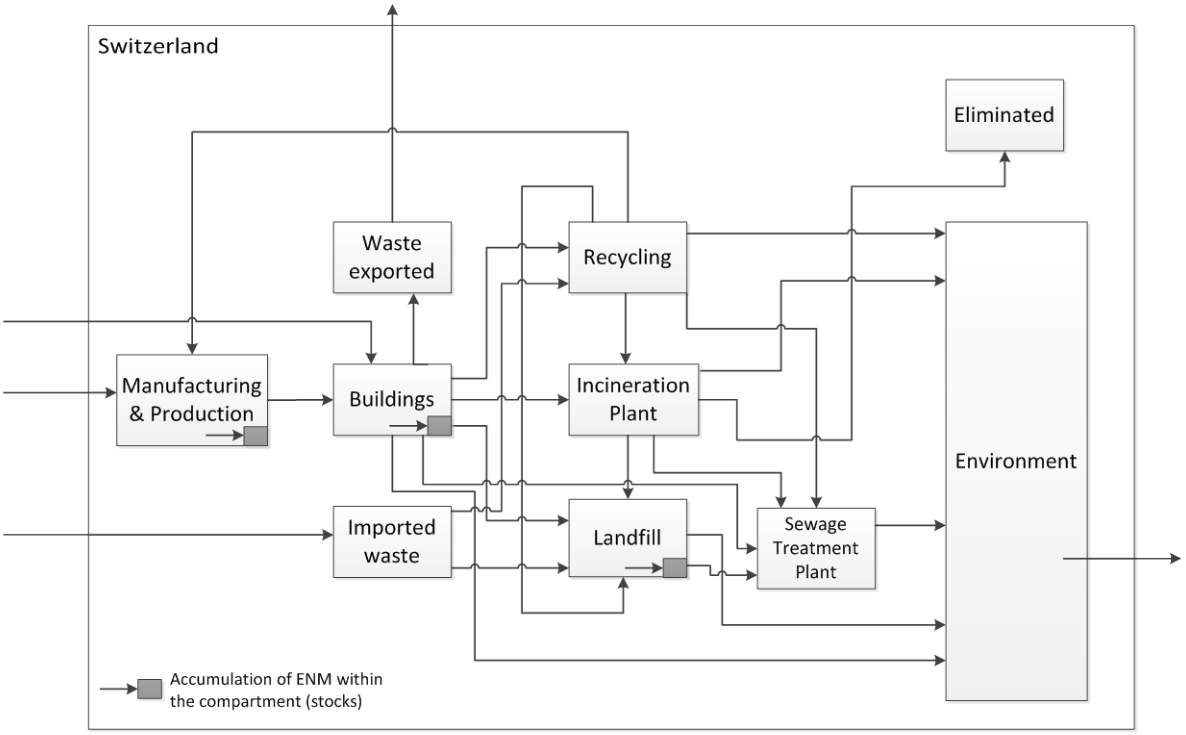


Figure 9. *Material flow diagram for ENMs in Construction and Demolition Waste (C&DW).*

Such a material-flow analysis is a valuable tool to identify where ENMs flow within a system and which compartments may be exposed to them. The evolution of the flows and stocks of ENMs in construction waste in the Swiss system depends on:

- The application type (ENM amounts used and allocation to the different applications);
- The waste management regulations;
- The technology used within the processes and facilities in the waste management system;
- The market dynamics: price and quantities sold (market penetration);
- The average age of an application before becoming waste.

Although nanotechnology has steadily grown during the last years, nano-applications in construction sector are still limited mainly to cement, paints and insulation material as shown in Chapters 4 and 6. The limited market penetration of these applications is related to their relatively high cost compared to traditional materials, the uncertainty about their risks to health, and the uncertainty about their real benefits (van Broekhuizen et al., 2011).

(Caballero-Guzman et al., 2015) stated that the way ENM is incorporated into the product is important to assess any potential release. An ENM can be released easier to water during a water-based recycling process if it is incorporated as a coating into the surface of a product, than if it is part of its material matrix.

7.2 Method to assess current ENMs flows in the system

This section describes in a simple way the system behavior of ENMs in applications used within the construction sector. However, to assess the current amounts of ENMs in the system is quite challenging, mainly due to the limited information available. There is no market information that allows us to determine the quantities currently used and the market share of these products. In a survey applied to the European construction sector in 2009, (van Broekhuizen et al., 2011) found that the market penetration is limited to specific niches, namely, concrete, paints and coatings, and insulation material. Other applications exist of course, but they are only commercialized to a minor extent.

The other important piece of information that is missing to assess the system behavior is the actual types and amounts of ENMs used in applications and the way they are incorporated into products. The information currently available has been obtained from the scientific literature, surveys and interviews. The reasons that no transparent and complete data are available from industry could range from industrial secrecy to the fear of manufacturers to clearly state that their products contain nanomaterials, especially when the safety for organisms and the environment is still subject of an ongoing research and debate. It is important to mention that from the risk assessment perspective, the more detailed information about the specific use of ENMs in applications is available, the better it is. At the nanoscale, knowing the amounts of ENMs used is not enough, because in this dimension there are other parameters that become much more relevant, like the particle size or if the nanoparticle was functionalized or not by the manufacturer, information that is usually not obtained through surveys or interviews, especially when even the information that experts in the area possess is rather limited.

To assess the flows of ENMs within the system, we considered a bottom-up approach (i.e. it begins with knowledge on actual nano-products) and a top-down approach (i.e. it begins with the total production of a specific ENM). In both cases the goal is to get an idea of the amounts of ENMs that will be transferred to the waste management system through the construction and demolition waste. Usually materials and applications in buildings have long lives, ranging from a couple of years (light lamps) to decades, or even centuries if we refer to historic constructions. In that sense, the nature of our analysis is prospective. We will not describe the dynamics of the system but rather how the ENMs that we think are currently present in the system will flow under the current legal and technological framework if they become waste. The value of this analysis is to foresee their flows to be able to perform a critical appraisal from the risk assessment perspective under the current system conditions. At the end, what is important is to understand if there is or will be a considerable risk to humans and the environment and this tool will provide a good starting point in that respect.

7.2.1 Bottom-up approach

In a **bottom-up approach** the starting point are the nano-applications. If the amounts sold of each application every year are known and we have information on the ENM-content in the product, we can estimate the amounts of ENMs in that application. With knowledge on the details of the incorporation of the ENM into the product we can determine the expected fate based on the current waste management regulations and the characteristics of the technical facilities used. The steps are described below:

1. Determine the quantities sold of each nano-application (market penetration);
2. Determine the ENM content of the application and, if necessary, the material fraction allocating the nanomaterial (metal, wood, glass, cement, etc.);
3. Determine the fate of the fraction containing the nanomaterial based on the waste management regulations;
4. If possible, determine any potential release of ENM in any of the technical or environmental compartments.

7.2.2 Top-down approach

In a **top-down approach**, the starting point is the total production amount of an ENM and the distribution of the mass between different applications. Once the amounts used each year in an application are known, the flows can be determined based on the current waste management regulations and the characteristics of the technical facilities used. The steps of this process are:

1. Determine the amounts of nanomaterial produced in a year;
2. Determine the amounts of nanomaterial used in a specific application;
3. Determine the fate of the fraction containing the nanomaterial based on the waste management regulations;
4. Determine any potential release of ENM in any of the technical or environmental compartments.

According to the described methods, **in Chapter 8 we will present the results of a semi-quantitative flow assessment for ENM specifically from paints in the Swiss construction waste stream**. With the information we obtained from the literature review and our survey we applied a bottom-up approach, where the amount of ENM in the construction applications was estimated. We compared these results with a top-down approach to determine the differences between the two methods and provide the best way to assess the ENM flows. Then we show the flow of different ENM in the construction waste and estimate their fate to the technical compartments. Then we evaluate qualitatively the potential release of ENM contained in paint residues from the technical and environmental compartments.

8. Semi-quantitative flow assessment of nanomaterials in construction waste in Switzerland

In this chapter we assess the flow of ENMs contained in paints when they become construction and demolition waste in Switzerland by using a bottom-up approach. We compare these results with a top-down approach and we investigate the amount of ENMs to recycling (REC), waste incineration plants (WIP), landfills (LF) and export and their potential release from construction waste. The ENMs flow behavior can be visualized in a semi-quantitative flow assessment. We focused on the ENMs contained in paint residues because paints are the most relevant current application in the construction sector in Switzerland according to the literature review and the survey (Chapters 4 and 6). Accordingly we focused on the most used ENMs used for paints: nano-TiO_2, nano-SiO_2, nano-ZnO and nano-Ag.

8.1 Bottom-up approach

The first step for this approach is to know the Swiss market of paints. We found data from the paint and coating industry about its market development in Switzerland for the year 2012 (Swiss paint and lack association) (VSLF, 2013). The market of paints is divided in different application segments (i.e. paint products for construction, plasters, industrial coatings, wood preservatives and wood coatings). The market distribution of the different segments is given in Table 10.

***Table 10**. Market distribution of paints in Switzerland for 2012. Source (VSLF, 2013)*

Application segment	Market distribution in Switzerland
Paint products and plasters for construction	76.26%
Wood coatings	3.34%
Others[1]	20.4%
Total	**100%**

[1] Other segment includes: industrial coatings, wood preservatives and printing inks

The majority of the sales from the paint industry are destined for the construction sector. Data about the volume of paint sold in Switzerland was not available. We estimated these amounts from a market study carried out by (McCulloch, 2012). In this study the market behavior of the 25 top paint manufactures in Europe was analyzed in different regions. It was found that for the EMEA region (i.e. Europe, Middle East and Africa) in 2012, the market size of paints was 18.7 Billion Euro, which correspond to 9'860 million Liters (m L) of paints sold (McCulloch, 2012). Based on this information and on the GDP PPP[2] data (see chapter 6.2) from both regions, an adjustment from EMEA to Switzerland was made and we estimated the size of the Swiss paint market to be 0.274 billion CHF and the amount of paint sold

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[2] GDP PPP: gross domestic product at purchasing power parity per capita. EMEA and Swiss GDP PPP (34'399 and 417 International dollars respectively) in 2012 (World-Bank 2012).

to be 45 m L in Switzerland. This volume was distributed according to the Swiss market of paints (see Table 10). Subsequently the amount of paints sold (in t) related to the different applications of paints in the Swiss construction sector was obtained (Table 11).

Table 11. *Estimation of the quantity of paint sold in Switzerland in 2012 based on sales in EMEA from (McCulloch, 2012) and Swiss market distribution from (VSLF, 2013)*

Application segment	**Market distribution in Switzerland (%)**	**Amount of paints sold in Switzerland[3] (t[4])**
Paint products[1] and plasters for construction	76.26%	139'445
Wood coatings	3.34%	6'107
Others[2]	20.4%	37'302
Total	**100%**	**182'854**

[1] Paint products for construction include: indoor and outdoor paints, other coatings

[2] Other segment includes: printing inks, wood preservatives, industrial coatings

[3] Estimation based on GDP PPP data, the conversion factor from EMEA to Switzerland was 0.0121

[4] Density of paint is assumed to be 1.5 Kg/L according to (ISO, 2011) and several product data sheets from paints

The values shown in Table 11 also agree with the amounts reported by (Burkhardt and Dietschweiler, 2013), where the Swiss market is assumed to be 10 times smaller compared to the German one (based on statistics by (VdL, 2012)). That means for paint products and plaster for construction sold in Switzerland, an amount of 90’000 t was estimated (we obtained 139’000 t, due to market size uncertainties and also to the paint density used for our estimations, see Table 11). Although these values are comparable, we provide rather a range of the amount of paints products and plaster for construction sold in Switzerland (i.e. from 90’000 to 139’000 t). We used the highest value of the range to continue with the next estimations. In (VdL, 2012) 6’200 t were reported for wood coatings, we obtained almost the same value (i.e., 6’107 t).

With the estimated amounts of paint sold in Switzerland (Table 11), we investigated the market of paints which contain ENMs (i.e. nano-paints) and determined the quantities sold of this nano-application (market penetration).

8.2 Quantities sold of a nano-paint (market penetration)

To determine the quantities of nano-paint sold in Switzerland, we used the results given in Table 11 and an assumed a market penetration for nano-paints of 1% for the year 2012 and 20% for year 2020 according to (Meili, 2013). This assumption also agrees with the observations of consumer behaviour of paints and cement containing ENMs provided in our Survey (chapter 6). With this analysis, we were able to estimate the sold amount of paints containing ENMs (t) in Switzerland for 2012 and 2020 (Table 12).

***Table 12**. Quantity of paint sold in Switzerland in t/y containing ENM taking GDP PPP data and assuming a market penetration 1 and 20%*

Application segment	**Nano-paints sold in Switzerland (t/y)**	
	Nano-paint market penetration of 1%[3] in 2012	**Nano-paint market penetration of 20%[4] in 2020**
Paint products[1] and plasters for construction	1'394	27'889
Wood coatings	61	1'221
Others[2]	373	7'460
Total	**1'829**	**36'571**

[1] Paint products for construction include: indoor and outdoor paints, other coatings

[2] Other segment includes: printing inks, wood preservatives, industrial coatings

[3] 1% is assumed due to the uncertainties in the market share of nanomaterials

[4] 20% is assumed according to (Meili, 2013) and our survey observations

The total amount of paints containing nanomaterials used in the construction industry in Switzerland in 2012 was 1'767 t/y. With these results we could determine the ENM mass used in the application (i.e. nano-paint).

8.3 ENM content of the nano-paint

To determine the amount of ENMs in paints, we use the estimated quantity of nano-paints sold in 2012 in Switzerland (Table 12) and the mass content of ENMs that normally is reported for paints. The mass content of ENMs was obtained from different sources for instance in the European Project NanoHouse (NanoHouse, 2013). Here the mass content of nano-TiO_2 in the analyzed paints was about 3 wt%, nano-SiO_2 was 5 wt% and nano-Ag was around 0.1 wt%. In the case of nano-ZnO a value between 1-5 wt% was recommended by the participants in the NanoHouse Survey. Likely, in our survey for this study, the same ranges of mass content were provided (i.e. for nano- TiO_2 and nano-SiO_2 between 1-5 wt% was reported and for nano-Ag less than 0.1 wt%). The mass content values used for this study are given in Table 13.

***Table 13.** ENM content in paint given by their mass concentration (wt%)*

Application segment	**Mass concentration of ENM in paints (wt%)**			
	TiO_2	**SiO_2**	**ZnO**	**Ag**
Paint products and plasters for construction	3%	5%	1%	0.1%
Wood coatings	-	-	-	0.1%
Others	n.a	n.a	n.a	n.a

The (-) means that none of this ENM is used in the application segment. n.a. means no data available

Combining Tables 12 and 13 we obtained the amount of ENM in paint in Switzerland. Due to the lack of and the uncertainty of market information, we show here the worst-case scenario. That is assuming that the complete mass of nano-paint is only nano-TiO_2, or nano-SiO_2, or nano-ZnO or nano-Ag for the years 2012 and 2020 (Table 14).

Table 14. *Amount of ENM (t/y) used in paints according to the mass of ENM usually applied in a nano-paint. For each ENM the numbers represent a scenario where only this ENM is used.*

Application segment	**Amount of ENM in paint for 2012 (t)**				**Amount of ENM in paint for 2020 (t)**			
	TiO_2	**SiO_2**	**ZnO**	**Ag**	**TiO_2**	**SiO_2**	**ZnO**	**Ag**
Paint products and plasters for construction	42	70	14	1.4	837	1394	279	28
Wood coatings	-	-	-	0.1	-	-	-	1
Others	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
Total	**42**	**70**	**14**	**1**	**837**	**1394**	**279**	**29**

The (-) means that none of this ENM is used in the application segment. n.a. means no data available

In order to obtain a more realistic scenario, we used the distribution of ENM amounts used in paints from our survey (Chapter 6, question 4). The ratios used for the distribution according to the answers of the survey were for nano-TiO_2 and ZnO (0.333) and for nano-SiO_2 and nano-Ag (0.167). The ENM content in the nano-paints in Switzerland is given in Table 15.

Table 15. *Distribution of the amount of ENM (t/y) used in paints considering the distribution between the various ENM according to our survey.*

Application segment	**Amount of ENM in paint for 2012 (t/y)**				**Amount of ENM in paint for 2020 (t/y)**			
	TiO_2	**SiO_2**	**ZnO**	**Ag**	**TiO_2**	**SiO_2**	**ZnO**	**Ag**
Paint products and plasters for construction	14	12	5	0.2	279	234	93	5
Wood coatings	-	-	-	0.01	-	-	-	0.205
Others	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
Total	**14**	**12**	**5**	**0.2**	**279**	**234**	**93**	**5**

The (-) means that none of this ENM is used in the application segment. n.a. means no data available

The material fraction where the nanomaterial was allocated to was done for nano-paints according to the application segments (plasters, paint products and wood). These amounts of ENMs serve to estimate their fate when entering in the waste management system. And also to compare with recent results from a top-down approach (Sun et al., 2014).

8.4 Comparison top-down approach vs bottom-up approach

We compared the amount of ENM in paints obtained for Switzerland in the year 2012 in section 8.3. with a top-down approach by (Sun et al., 2014) to see how close or how far we are from the amount of ENM in paints estimated (Table 16).

***Table 16**. Total amounts of ENM in paint in Switzerland, modelled by (Sun et al., 2014) and comparison with the quantities obtained in this study*

	Sun et al., (2014)			This study
	total in CH (t)	Share in paint	ENM in paint in CH (t)	ENM in paint in CH estimated in this report (t)
nano-TiO_2	337	8.9%	30	14
nano-ZnO	48.4	14.3%	7	5
nano-Ag	1.1	3%	0.03	0.2
Others	13.3	1%	0.2	n.a
Total	**399.8**	**28%**	**37.23**	**19.3**

The main conclusion of this comparison is that the actual ENM mass in paints obtained in this report is in the same order of magnitude as the estimates from (Sun et al., 2014), except for nano-Ag, which is 7 times higher in our estimate. However, the use of nano-Ag seems to be often overestimated. The following table shows the required market share of nano-paints to reach the ENM-amounts of Sun et al, 2014.

***Table 17**. Required market penetration to achieve the ENM amounts from Sun et al., 2014 in Switzerland*

ENM	Penetration market to reach Sun et al., results
nano-TiO_2	2.1%
nano-ZnO	1.4%
nano-Ag	0.2%

For the case of ENM in paints our results from the both bottom-up approach were comparable with a top-down approach. That means one can apply one or the other kind of approach depending of the available information for each specific nano-application.

The next step of this analysis is to determine the fate of the fraction containing the nanomaterial based on the waste management regulations.

8.5 Waste flows of ENM contained in paints

With the data obtained about the amount of ENM in nano-paints for year 2012 (Table 15) and data from the destination routes of paints in the Swiss waste management system (Table 5, Chapter 5), we can estimate the fraction of ENMs contained in paints applied to walls or façades (also cover with plasters) and woods (due to the lack of specific data for every waste sub category) to recycling, incineration, landfills and export (see following Table and Figure 10).

Table 18. *Estimation of the fate routes for containing ENMs paints and coatings in the construction and demolition flows in Switzerland*

Applications	**Fate (t)**				
	ENM	**REC**	**WIP**	**LF**	**Exported**
Paint					
In stones, masonry surfaces (walls or façades)	**TiO_2**	10.74	0	3.21	0
	SiO_2	9.01	0	2.69	0
	ZnO	3.58	0	1.07	0
	Ag	0.18	0	0.05	0
Wood surface	**Ag**	0.001	0.01	0	0
Total		**23.50**	**0.01**	**7.02**	**0**

The flows after recycling are distributed according to (Hischier et al., 2015), where about 85% of the residues go directly to the landfill and the rest (~15%) go back to manufacturing process. In Figure 10a, we also included the export of mixed demolition waste to recycling process (i.e. 37’000 t in 2012) as in (FOEN, 2012). To estimate this flow, a transfer coefficient of 3% was obtained according to the total mixed demolition waste going to recycling (see Table 4).

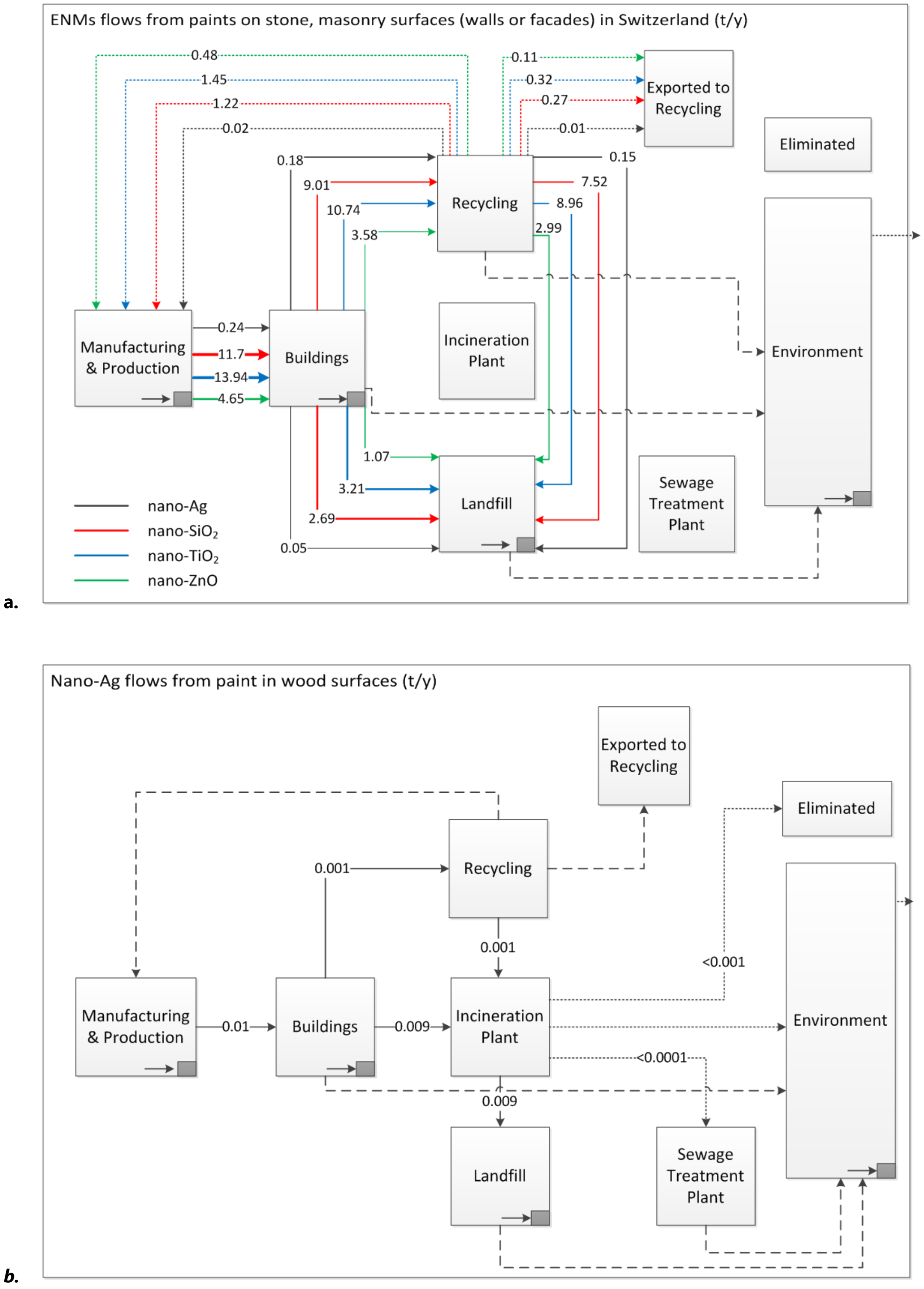

Figure 10. *Nanomaterial flows in Switzerland of ENM used in paints and coatings:* ***a.*** *on stone or masonry (i.e. walls or façades) surfaces.* ***b.*** *on wood surfaces. Dotted long-lines represent flows that either are potential, or that exist but no data is available. Dotted short-lines represent flows that are small.*

In Figure 10b, we used a transfer coefficient of 100% as in (Caballero-Guzman et al., 2015), to calculate the flows from recycling to incineration. Although it is likely that some wood with nanocoating is recycled (dotted line from Recycling to Manufacturing and Production), officially all painted wood should be incinerated. To estimate the flows from incineration to elimination, sewage treatment plant and landfill, we used following transfer coefficients respectively: 2.5%, 0.02% and 97.48%, according to (Müller et al., 2012).

The fraction of ENMs entering the flow of C&DW in Switzerland due to the use of paints and coatings containing ENM is 30.5 t/y. The majority is going to recycling, followed by landfills processes. Nano-TiO_2 and nano-SiO_2 are here the main source on ENMs in the recycling. In the case of nano-SiO_2 fraction, this ENM is also expected to increase the flows mainly in the recycling system and to contribute to the flows to landfills because concrete and pavement are recycled to 90% and landfilled to 10%. Figure 10a. and b. also shows that after recycling, most of the paint residues containing ENMs are going to landfills.

We recommend performing the same type of analysis for other products like cement or insulation materials, when data on the actual market size become available. This is needed so that we can sum up all fractions and are able to obtain a complete picture of the behaviour of ENMs in construction waste.

The results obtained from the analysis of the fate of ENM from nano-paints in the construction and demolition waste in Switzerland make it possible to determine (at least qualitatively) the potential release of ENM in any of the technical or environmental compartments.

8.6 Potential release and exposure of ENM contained in paint from waste in technical or environment compartments

During the end-of-life stage of nano-applications, release of nanomaterials may occur within technical (recycling facilities, landfills or incineration plants) and environmental compartments (air, soil and water). Release to the environment usually takes place after nano-applications are handled and processed in technical compartments, as depicted in Figure 10. Release of embedded nanomaterials in products and applications can take place due to mechanical, thermal or chemical degradation of composites hosting the nanomaterials described by Froggett (2014a).

In this section the release potential of nanomaterials within the technical compartments previously described is shortly discussed based on the literature available. Release potential is defined as the possibility that a nanomaterial added to form a nanocomposite may become separated from the composite matrix and released to the surrounding environment (Froggett, 2014a). It should be considered that release potential is dependent on the scenario and the environment, as described by Nowack et al. (2013).

8.6.1 Release during renovation and demolition

One pathway that way not included in the flow modelling is the possible release during renovation or demolition. These activities are normally producing quite a lot of dust and therefore also release of ENM is possible. This is important especially for occupational exposure but also for environmental release, as the dust will settle on soils or is washed into sewers with water. A quantification of these flows is currently not possible.

8.6.2 Release during recycling

In contrast to landfilling, were only three different types of landfills exist; recycling is an activity that involves many different processes depending on the waste type and the materials that are treated. Therefore, release during recycling is dependent on the technical characteristics of the processes that treat the material recycled. Construction waste that is recycled includes mineral residuals (cement production), concrete (aggregate production), metals and wood. Only release of fine particles during concrete recycling has been analyzed in depth. Kumar and Morawska (2014) found that more than 90% of the particles released during handling concrete aggregates are below 100 nm. However, it is important to note that this was a standard concrete that did not contain any nanomaterials and the results can therefore serve as a baseline for production of a nanoparticulate fraction during recycling operations. For the remaining materials, for which no studies on release exist, we can expect too some releases: for cement production, during the handling of mineral material which is used as additive and for particleboard production (wood recycling) during chips production. Release during metal recycling is less likely, although some leakages would be normal during the metal handling.

The use of water in the production processes aforementioned is not expected, so the likeliness of release of ENMs to water is unlikely. The transfer to the soil would be through the deposition of the airborne nanoparticles.

8.6.3 Release during landfilling

Currently, the information about the release potential of ENMs in this technical compartment is almost inexistent. Mueller et al. (2013) modelled the flows of nanomaterials during waste handling in Switzerland, but were not able to quantify any release from landfills to the environment. Nanomaterials in construction waste could be transferred to landfills mainly through the mineral material containing nano-paint or through the concrete which is not recycled. However, an indirect route is through the bottom ash of incineration plants after wood with nanomaterials is burned. In both cases some release to air, soil or water (leakages) are possible, although further studies are necessary.

It is likely that the landfill conditions will promote the release of nanomaterials. However, the changing conditions in them will also influence the ENM behavior and composition. If released, their characteristics must be determined to ensure that the relevant state for toxicity is evaluated (Reinhart et al., 2010). Whether they are transferred to the air is uncertain, but likely. Another possibility that should be further analyzed is the transfer of nanomaterials to the groundwater through the leachate. Leachate is primarily formed as water percolates through the waste layers, where organic and inorganic components from the waste are solubilized and incorporated within it (Lozecznik et al., 2010).

8.6.4 Release during incineration

Incineration plants are currently been subject to intense study under the context of nanomaterials. The aforementioned model by Mueller et al. (2013) found that, in most cases, the major flows of nanomaterials from incineration occur to bioactive landfills through the bottom ash. In the output of their work, little release of nanomaterials is observed to the air and to the waste water treatment plants. In an experimental set up with nano-Cerium Oxide (CeO_2) on a full-scale incineration plant in Switzerland, (Walser, 2012) found that wet scrubbers filter up to 99,9% of the nanoparticles in the flue gas. The authors concluded that no relevant emissions to air are expected from plants equipped with state-of-the-art flue gas cleaning systems. Although the previous studies point out to a low level of emissions to air and water, their results cannot be generalized to every plant, as technical variations between plants may play an important role in the amount of nanomaterials released to the environment.

8.6.5 Potential release of ENMs

Based in the little literature available on release and our expertise, we can qualitatively asses the release potential in the technical compartments previously analyzed (Table 19). During recycling, some release to the air is likely during the handling of the mineral material for the production of cement, during the production of aggregates from concrete and during the sawmilling of wood chips for the production of particle boards. The transfer to the soil would be through deposition of airborne particles. Release to water during recycling is less likely as long as no water is expected to be used during those recycling processes. During landfilling, release of ENMs to air, soil and groundwater is likely. ENMs transferred to landfill include mixed demolition waste and concrete by a direct way, or bottom ash from incineration plant by an indirect route (wood incinerated). Nanomaterials in landfills may be carried to the air by the wind and to the groundwater by the leachate. During incineration, the degree of likeliness of release to air, soil and water is strongly dependent on technology used. The studies carried out so far in Switzerland show very low levels of release of nanomaterials to air, soil and water, although the results should not be generalized.

Table 19. *Release potential of ENMs during recycling, landfilling and incineration of construction and demolition waste.*

Technical compartment	Potential release		
	Air	**Soil**	**Water**
Recycling	Likely	Likely	Unlikely*
Landfilling	Possible	Possible	Possible
Incineration	Unlikely	Unlikely	Unlikely

* Unless wet processes are used.

9. Discussion and conclusions

This report focuses on current applications in the construction sector. However, the reader should keep in mind that the ENM-flows will ultimately depend on the market dynamics, something that is not possible to assess. Innovations frequently disappear after some years of unsuccessful trials to penetrate the market. This will depend on the prices and that the expected benefits materialize compared to conventional materials and applications. As it can be seen in paints and coatings (biggest application of ENMs in the construction sector), if benefits do not materialize, then it is expected that in the near future this application will disappear, and thus no more nano-Ag and nano-TiO_2 will be transferred through the C&DW. Even if we decide to study the applications that are currently trying to penetrate into the market, we don't know how they will evolve in the future.

Some reasons of why ENM-applications represent only niche segments in the market are according to (van Broekhuizen et al., 2011) the high cost of the ENMs, uncertainty on safety and uncertainty on technical performance.

9.1 Changes related to the report on nanomaterials in waste incineration and landfills

According to our last report on Nanomaterials in waste incineration and landfills (Müller et al., 2012), one of the conclusions of the modelling was that the second most significant input of ENMs into landfills is the direct disposal of construction waste in landfills. From these materials that may be crushed and compacted on-site, release of ENM might be possible. The model describes a direct disposal of C&DW to an inert landfill.

We saw in our work that for Switzerland residues from construction are in the first instance recycled (59%), 31% are disposed in landfills for inert materials (mainly the mineral fraction, i.e. roofing materials, ceramic, stoneware, slag, gypsum, plaster, glass, windows) and the rest is incinerated. Thus, assuming direct disposal of C&DW to inert landfills can overestimate the ENM fractions of nanomaterials in the models, for instance in (Müller et al., 2012). On the other hand, during recycling the ENM may end up in fractions that are disposed in landfills and therefore the overall flow to landfills might be higher.

9.2 Current limitations to model flows and releases

The flow assessment needs to be refined and be done based on an application-by-application basis whereas our work assesses the "average application". However, differences in incorporation of ENM into products during manufacturing may have an effect on the release potential. The main lack of information is related to the actual usage of ENM in products on the market and their market penetration. Future assessments should therefore focus more on the market information than on the nano-specific assessment of flows.

In our flow assessment we did not consider any transformation or destruction of ENM during use, aging, release, recycling or disposal, only to a limited extend, based on previous modeling by (Müller et al., 2012) and (Caballero-Guzman et al., 2015). Transformation reactions can completely eliminate the nano-properties of the ENM, for example if they are dissolved. This has been observed for example for SiO_2 in paints where in laboratory experiments significant dissolution of nano-SiO_2 was observed during release (Al-Kattan, 2015). Also nano-Ag can undergo dissolution reactions, resulting in destruction of the nano-identity of the material. After such transformation reactions, the materials are no longer nanoparticles and thus no special assessment has to be performed anymore. It is therefore important to get more real-world data on transformation and degradation reactions of ENM in order to decide if a nano-relevance exists with a certain product or not.

Even if the ENM are released untransformed, there still may be no nano-relevance. Most of the work performed about release of materials from nano-products has shown that the released materials are mainly present of matrix fragments with embedded ENM (Froggett, 2014a). This was for example observed for paint containing nano-TiO_2 where mainly paints fragments containing nano-TiO_2 embedded in the polymer matrix were released (Al-Kattan, 2014). These fragments have different effects compared to the pristine and free materials – in vitro tests the particles contained in paint had no observable effects whereas some of the pristine particles did affect cell growth (Kaiser, 2013). This clearly shows that it is not sufficient to know about the overall flows of ENM in the construction sector but also have information on the fate of the particles.

A major limitation of our current evaluation is that we did not consider the time, especially the long time any ENM in a building is fixed inside the building, thus resulting in a very long delay of a possible release. Most of the flows estimations were done for year 2012 and assuming a market penetration of ENM of 1% for the same year. It is unlikely that now (year 2014) relevant waste flows are already coming from nano-applications. The ENMs will start to distribute to the different compartments decades from now. Furthermore in the future recycling processes are going to be more used and improved, probably resulting in different flows than today. Also the regulation about ENM will probably develop and they must be taking into account for further analysis.

9.3 Conclusions

- The survey showed that there is still a significant lack of knowledge about the uses of ENM, the market size and the amounts of ENM used in construction materials.
- Paints seem to be the major current use of ENM in the construction sector.
- **We were able to perform a semi-quantitative flow assessment for ENM specifically from paints in the Swiss construction waste stream.**
- Using a bottom-up approach we estimate the ENM mass used in paints to be 14 t/y for nano-TiO_2, 12 t/y for nano-SiO_2, 5 t/y of nano-ZnO and 0.2 t/y for nano-Ag.
- A comparison with a top-down approach yields very similar results, supporting the validity of the obtained mass flows.
- For these paints the main overall mass flows are going to recycling and landfills.

- The flow assessment can form the basis for assessing the potential for release.
- Occupational exposure may mainly be possible during recycling operations.
- Potential release from the technical compartments where waste is treated is likely during recycling and landfilling.

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