

Supporting Information

Prospective Environmental Life Cycle Assessment of Nanosilver T-shirts

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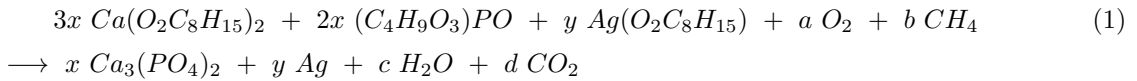
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S.1 Process Descriptions of Nanoparticle Production

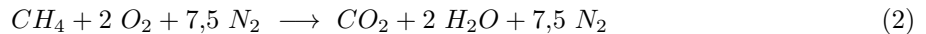
S.1.1 Flame Spray Pyrolysis

In the following chapters, nanosilver and nanoAg-TCP are only distinguished if necessary, otherwise, information is the same for both compounds.

If the reader is interested in more information about the nanoAg-TCP production technology than provided below, we recommend the studies of Kammler et al. (2001) [51] and Dreesen et al. (2009) [26]. The material and energy requirements of most of the chemicals which are used for the production of nanosilver are found in the ecoinvent database v2.2 [20]. For precursors not found in the database, the inventory has been established, which is presented below in more details. Electricity is the main energy carrier and hence its production is influencing the environmental impacts of the nanoparticle processes. The direct electricity use of the processes was measured on site and the UCTE-electricity mix was chosen as default. The amount of precursors for *nanoAg-TCP* and *nanosilver* is presented in Table S.1. The dry, gas-phase process converts oil-like mixtures (2-ethylhexanoic acid, xylene, calcium hydroxide, tributylphosphate, silveroctanoate) and gases (oxygen, methane) in a flame into nanoparticles (*nanoAg-TCP*, *nanosilver*). The organic part of the precursors is burnt off and leaves behind the metal oxides, phosphates and the silver metal. Due to the fastness of the process, particles condense to nano-aerosols (*nanoAg-TCP*, *nanosilver*) which are subsequently filtered on a Teflon- or glass-fiber filter (*e.g.* [67, 68, 75]). The nanosilver particles have a size of 1-2 *nm* and in the case of *nanoAg-TCP*, they decorate the surface of 20-50 *nm* phosphate-based, ceramic carrier particles (*TCP*) [65]. Stoichiometry of the complete combustion for *nanoAg-TCP* is [65]:



The production of 1 *kg nanoAg-TCP* emits 2184 *mol* atomic *O*, 1860 *mol* atomic *H* and 998 *mol* atomic *C*. During combustion, excess amount of air is driving a complete combustion (using ambient nitrogen), described by following reaction [52]:



Reaction temperature of about 2000 °C results in non significant amounts of carbon monoxide [79] but ambient nitrogen is partly converted into thermal *NO*. The formation reactions are described by the ZELDOWICH mechanisms [119]):



An additional elementary reaction with hydrogenoxide is applicable in some cases when *NO* formation is under-estimated [48]. The above mentioned reactions happen in high-temperature air and *NO* production rate is calculated with Equation (4) [47, 62]:

$$\frac{d[\text{NO}_x]}{dt} = \frac{6,0 \cdot 10^{16}}{T^{0,5}} e^{\frac{-69090}{T}} [\text{N}_2][\text{O}_2]^{0,5} \quad (4)$$

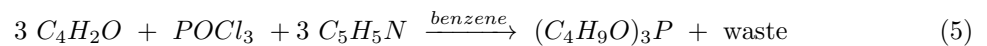
where *T* is the absolute temperature in Kelvin, $[\text{N}_2]$ and $[\text{O}_2]$ are ambient concentrations [*mol* · *cm*⁻³] and $d[\text{NO}_x]/dt$ is the rate of formation [*mol* · *cm*⁻³ · *s*⁻¹]. 78,08% of dry air is *N*₂ and 20,95% is *O*₂. Under standard conditions (ideal gas state), 1 *cm*³ of air contains $4,46 \cdot 10^{-5}$ *mol* of gas and hence $3,49 \cdot 10^{-5}$ *mol N*₂ and $9,35 \cdot 10^{-6}$ *mol O*₂. For the analyzed FSP process, the result of the reaction

above emits $8,56 \cdot 10^{-6} \text{ mol} \cdot \text{cm}^{-3} \cdot \text{s}^{-1} \text{ NO}$ (2000 °C). The production time of 1 kg nanoAg-TCP is 37'662 s. Concentration of formed NO (unlimited N_2 and O_2 supply) is $9,67 \text{ g} \cdot \text{cm}^{-3}$. The effective volume of NO formation is estimated as 40 cm^3 (at mean temperatures below 1600-1800 °C, thermal NO formation is significantly reduced [48]) which emits a total amount of $387 \text{ g NO} \cdot \text{kg}^{-1} \text{ nanoAg-TCP}$. Further air emissions are carbon dioxide ($43,92 \text{ kg} \cdot \text{kg}^{-1} \text{ nanoAg-TCP/nanosilver}$) and wastewater ($16,75 \text{ kg} \cdot \text{kg}^{-1} \text{ nanoAg-TCP/nanosilver}$). Table S.1 presents the FSP requirements for 1 kg nanosilver and nanoAg-TCP. For 1 kg pure nanosilver, no tributylphosphate but 2,35 kg silver-octanoate are required.

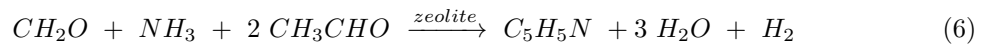
Table S.1: Precursors for 1 kg nanoAg-TCP and 1 kg nanosilver.

compound	amount [kg] (nanoAg-TCP)	amount [kg] (nanosilver)
oxygen (O_2)	33,39	33,39
methane (CH_4)	1,526	1,526
tap water (H_2O)	62,77	62,77
silver-octanoate ($AgC_8H_{15}O_2$)	0,047	2,35
tributylphosphate ($(C_4H_9O)_3P$)	1,735	0,0
calcium hydroxide ($CaOH$)	0,702	0,0
2-ethylhexanoic acid ($C_8H_{16}O_2$)	6,290	6,290
xylene (C_8H_{10})	6,290	6,290

- Synthesis of **tributylphosphate** (TBP) [27] is shown in reaction (5). Waste of synthesis of TBP is a mixture of amyl/butyl/propylester (waste products of benzene) and pyridine [27]. This mixture is incinerated with energy recovery. 50% of pyridine is recovered [98]). Environmental impacts of these residues are calculated with ECOSOLVENT[19]. For amyl/butyl/propylester, butyl acetate ($C_6H_{12}O_2$) is chosen as the reference solvent for all three of the compounds because of its similar chemical and physical properties. The modeled mixture of pyridine and butyl acetate is distilled batch-wise to recover the pyridine (boiling point: 115 °C). Environmental impacts of the pyridine recovery (modelled with ECOSOLVENT[19]) are allocated to the TBP production process.



- Large-scale synthesis of **pyridine** (6 [99]):

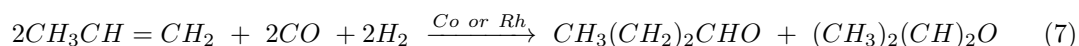


For synthesis of pyridine, a yield of 60% (dependent on the efficiency of the zeolite) is reached under optimized process conditions [22, 98, 99]. Emissions are hydrogen, water, used catalyst and solvent mixture. The solvent mixture is incinerated. Environmental impacts are calculated based on the ALM tool results [97].

- **Silver octanoate** is formed by octanoic acid and silver-ions. Octanoic acid, also known as caprylic acid, is an eight-carbon saturated fatty acid and is naturally found in coconut (5-10 %) [111]. For the manufacture of fatty acids on a commercial scale, only fats available in large quantities are used as raw materials – coconut oil is one of them [111]. Synthesis of the silver salt of a fatty acid which contains at least 8 carbon atoms is described in [2, 102] and is adapted from behenic acid to octanoic acid. An aqueous solution of silvernitrate is added to the solution with the octanoic acid.

The precipitate containing the silver salt of the octanoic acid is then separated from the reaction mixture. To obtain 20 g of pure silver (bound as silver octanoate), 54,17 g octanoic acid is needed, together with 51,5 g of silver nitrate, 7,57 g of sodium hydroxide, and 18,02 g of deionised water. Reaction takes place at 85 °C and takes 5 min.

- **N-butyraldehyde** (C_4H_8O) is commercially produced foremost by the oxo synthesis route of propylene (hydroformylation of propylene and CO/H_2 synthesis gas). It is an exothermic gas/organic liquid phase reaction (-118 – -147 kJ/mol olefin) using a homogeneous cobalt catalyst at 130-150 °C and 100-300 bar [30, 88, 112].



It gives a ratio of *n*- to *iso*-butyraldehyde of up to 4:1 whilst propylene is used to up to 98% [32, 55, 72, 116]. 80 % are butyraldehydes, and 10-14% butanols and butyl formates (co-products). Environmental impacts of butyraldehyde production process are economically allocated to n-butyraldehyde and isobutyraldehyde: n-butyraldehyde price goes 90% into the production of 2-ethylhexanol (1285 EUR/ton) and <10% into n-butanol (1165 EUR/ton) which gives an average price of 1273 EUR/ton [55, 88]. Isobutyraldehyde is cheaper with 564 EUR/ton. Hence, the allocation factor of environmental burdens for n-butyraldehyde is 90%. N-butyraldehyde is obtained after distillation from the crude oxo product. Carbon monoxide and hydrogen are converted up to 90% to the aldehydes and alcohols, the waste is combusted [109]. 4,6 mg copper catalyst ($7,2 \cdot 10^{-5}$ mol, 63,5 g/mol) is added to the life cycle inventory, and it is assumed that this catalyst is incinerated after use. This amount of catalyst is a standard value in ECOINVENT (no other information was available). N-butyraldehyde is then converted into butyraldol via an alkali (aqueous sodium hydroxide, always recovered) catalyzed reaction and then crotonized¹ to 2-ethylhexanal [28, 55]. The ratio of aldehyde to aqueous sodium hydroxide solution is in the range 1:10 to 1:20. Conversion rates are > 98% at a temperature close to 150 °C. The process running at 150 °C produces up to 120 kg steam/ton *n*-butyraldehyde (exothermic process). On large scale, the process temperature is lower [30] and therefore, steam production is neglected in the calculations. Copper-chromite and nickel are the most common commercially used catalysts [86] for the reaction. **2-ethylhexanoic acid** is then produced by selective hydrogenation of 2-ethylhexanal with *Pd* catalysts [42, 112] which is again an exothermic reaction (-178 kJ/mol) [30] and enables energy recovery through steam generation. Catalyst's life is over 2500 ton product/ton catalyst and therefore, the *Pd* catalyst is not taken into account. Waste gas (0,05 m³/ton ethyl hexenal) as well as other wastes (wastewater, solid waste) become insignificant when best available techniques are used [30]. Over 95% of the current 2-ethylhexanoic-acid production is based on propylene [111]. Generally, no or few waste for disposal is produced during these steps (< 50 kg/ton product) and if so, they are mostly combusted to recover their inherent energy [30].

¹Specific aldehyde condensation with ketones, releasing water [6]

Table S.2: Distillation processes of solvent mixtures

products	unit	butylacetate	butyl acetate in pyridine	amyl/butyl/propylester	pyridine, 32% recovery				
recovered products									
	<i>kg</i>	butyl acetate, at plant/RER U	0.675	-	-	butyl acetate, at plant/RER U	0.675	pyridine	0.322
resources									
water, process and cooling, unspecified natural origin	<i>m</i> ³		0.0270		0.00270		0.0270		0.00870
air	<i>kg</i>		0.107		0.00910		0.107		0.0348
materials/fuels									
natural gas, high pressure, at consumer / CH (RER for pyridine) U	<i>MJ</i>		0.0384		0.0115		0.0384		0.148
nitrogen, liquid, at plant/RER U	<i>kg</i>								0.001209
hydrochloric acid, from the reaction of hydrogen with chlorine, at plant / RER U	<i>kg</i>		0.005		0.0009		0.005		0.00161
ALM waste flow 2	<i>kg</i>								0.678
electricity/heat									
steam, for chemical processes, at plant / RER U	<i>kg</i>		1.49		0.152		1.49		0.482
electricity, medium voltage, production UCTE, at grid / UCTE U	<i>kWh</i>		0.219		0.0358		0.219		0.0753
emissions to air									
carbon dioxide, fossil	<i>kg</i>		1.45		0.254		1.45		0.580
chlorine	<i>kg</i>		0.005		0.0009		0.005		0.00161
nitrogen oxides	<i>kg</i>								0.000644
final waste flows									
chemical waste, regulated	<i>kg</i>		0.325				0.325		

Table S.3: Production of 1 kg nanoAg-TCP and nanosilver

products, [kg] or [kWh]	nanoAg-TCP	nanosilver
oxygen (O_2)	33.4	33.4
methane (CH_4)	1.53	1.53
tap water (H_2O)	62.8	62.8
silver-octanoate ($AgC_8H_{15}O_2$)	0.047	2.35
tributylphosphate ($(C_4H_9O)_3P$)	1.74	-
calcium hydroxide ($CaOH$)	0.702	-
2-ethylhexanoic acid ($C_8H_{16}O_2$)	6.29	6.29
xylene (C_8H_{10})	6.29	6.29
electricity, medium voltage, at grid, UCTE	25.1	25.1
emissions		
nitric oxide	0.387	0.387
carbon dioxide, fossil	43.9	43.9
water	16.8	16.8
wastewater treat. (Cl 3, m^3)	0.063	0.063

Table S.4: Production of 1 kg 2-ethylhexanoic acid

products	amount	unit
coproduct		
steam, for chemical processes, at plant/RER U	0.000716	kg
materials/fuels		
n-butyraldehyde	1.02	kg
transport, lorry 16-32 t, EURO5/RER U	10.8	tkm
emissions to air		
carbon dioxide, fossil	0.0499	kg

Table S.5: Production of 1 kg coating (5 μm), nanoAg-TCP not included

products	amount	unit
recovered products		
ethanol from ethylene, at plant/RER U	0.167	kg
resources		
air	6.39	kg
water, cooling, surface	7	kg
materials/fuels		
polyethylene terephthalate, granulate, amorphous, at plant/RER U	0.996	kg
ethanol from ethylene, at plant/RER U	0.25	kg
nitrogen, liquid, at plant/RER U	0.00125	kg
hydrochloric acid, from the reaction of hydrogen with chlorine, at plant/RER U	0.001	kg
ALM waste flow 3	0.084	kg
electricity/heat		
electricity, medium voltage, production UCTE, at grid/UCTE U	0.319	kWh
steam, for chemical processes, at plant/RER U	0.373	kg
emissions to air		
carbon dioxide, fossil	0.332	kg
nitrogen	0.00125	kg

Table S.6: Production of 1 kg pyridine

products	amount	unit
materials/fuels		
acetaldehyde, at plant/RER U	0.788	kg
zeolite, powder, at plant/RER U	2.02E-08	kg
ammonia, liquid, at regional storehouse/RER U	0.610	kg
formaldehyde, production mix, at plant/RER U	0.269	kg
ALM waste flow 7	0.667	kg
electricity/heat		
steam, for chemical processes, at plant/RER U	2.14	kg
emissions to air		
hydrogen	0.0252	kg
final waste flows		
water	0.683	kg
waste to treatment		
disposal, cation exchange resin f. water, 50% water, to municipal incineration/CH U	2.02E-08	kg

S.1.2 Plasma Polymerization with Silver Co-Sputtering (PlaSpu)

If the reader is interested in more information about the plasma technology with silver co-sputtering than provided below, we recommend the studies of Giessmann et al. (2002) [37], Koerner et al (2008, 2009) [57, 58], and Balazs et al. (2007) [9]. The energy and material requirements for the production of the precursors is presented below. The material and energy requirements of most of the chemicals which are used for the production of nanosilver are found in the ecoinvent database v2.2 [20]. For the precursors not found in the ecoinvent database, the inventory has been established, which is presented below in more details.

The investigated functional textile coating is produced with silver-particle sputtering during plasma polymerization: A thin amorphous hydrogenated carbon (*a-C:H*) film is deposited on polyester textile by plasma polymerization with simultaneous incorporation of silver nanoparticles (sputtered from a target) into a nanoporous polymeric matrix [43–45]. The plasma polymer is deposited using ethylene (C_2H_4) as precursor gas. Upon the addition of the additional reactive gas CO_2 , carboxylic functionalities are added to the (*a-C:H*) network. An excess of argon is used in the gas mixture for co-sputtering of silver which is carried out in vacuum (about 1 Pa). The sputtering process is initiated by the impact of energetic particles on the silver plate, causing a multi-atom kinetic collision process. As a consequence the dislodged silver atoms are emitted from the target (cathode). This process is controlled by the energy and the number of bombarding particles [44]. Process and production facilities are shown in Figure S.1. The process gas mixture composes $C_2H_4/CO_2/Ar$ (1:6:1). The degree of functionality of the (*a-C:H:O*) coatings strongly depends on the CO_2/C_2H_4 gas ratio. Typical CO_2/C_2H_4 ratios vary between 2:1 and 8:1. For the laboratory scale plant and for the pilot plant, a 6:1 ratio is used whilst the estimated commercialized process will run with a 2:1 ratio. The plasma-polymer nanosilver composite-film contains the silver in form of small particles which are encapsulated in a nearly homogeneous plasma polymer matrix. The particle diameter varies between 2 and 10 nm and the particles show elliptical shape. Nanosilver sticks partly out of the coating surface. The Ag particle distribution and the amount of Ag in the coatings is influenced by power input, process velocity, gas pressure and argon feed ratio [8]. With an increasing monomer proportion, the sputtering rate on the metal target decreases, and the deposition rate of the plasma polymer increases [45]. Nanosilver concentration in the analyzed coating is about $4,4 \mu m \text{ silver} \cdot cm^{-2}$, which is incorporated into the 50 nm thick polymer layer on top of polyester textiles.

The pilot scale process requires cleaning nitrogen gas to float the process chamber after processing in order to prevent pump system corrosion. Compared to other nanoparticle manufacturing processes, lower vacuum is required. The resulting nanosilver-polymer coating on polyester textiles is stable and durable – aging effects occur only due to internal chemical reactions and external adsorptions [21]. The laboratory scale batch reactor is a similar setting as the pilot plant web coater but is less efficient in material and energy input per coated area, despite for silver. A larger silver plate is less efficiently used (more silver waste) than in the laboratory scale setting. The nanosilver coating process for one T-shirt needs 120 hrs on the laboratory scale, 6 hrs on the pilot scale, and is estimated to 6 min when commercialized.

LCI data for the production of all precursors are available from the ECOINVENT v2.2 database. The contribution of the most important material and energy inputs to the climate footprint are shown in Figure S.6. Middle voltage UCTE electricity is taken. Infrastructure is neglected. Density of the *a-C:H:O* layer is $1,4 g \cdot cm^{-3}$, which results in a total mass of $70 mg \cdot m^{-2}$ polyester coating (50 nm,

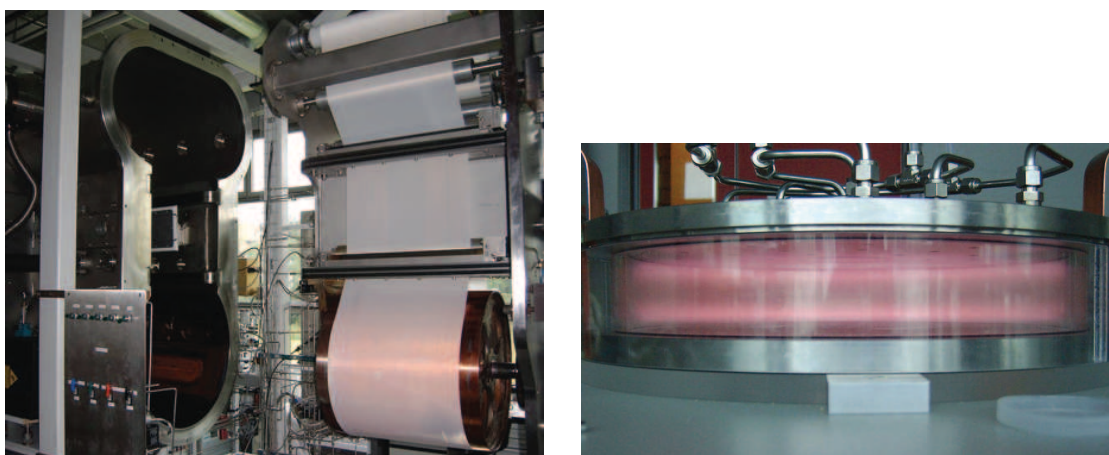


Figure S.1: Plasma polymerization with silver-co sputtering. Pilot scale plant (left side, apparatus open) and laboratory scale plant (right side, in action). *Source:* KOERNER (2008) [58].

without silver). Based on this estimation, following waste emissions are calculated (for the pilot plant): $C-H-O$ layer has an atomic ratio of $C_{0,48}O_{0,36}H_{0,16}$ [58, 100] when formed in hydrocarbon atmospheres like it is the case here. It results in a molecular weight of $9,16 \text{ g} \cdot \text{mol}^{-1}$. 6 mmol of the polymeric compound is needed for a total mass of 85 mg polymer. Exact values for input and emissions are found in Table S.8. It is assumed that all carbon will react to carbon dioxide in the excess of effluent ambient air. Hydrogen will react with oxygen to water. Emissions are $138 \text{ g } CO_2$ and $28 \text{ g } H_2O$ per m^2 coating.

Table S.7: Material and energy requirements of the laboratory and the pilot scale coating plant, normalized on 1 m^2 coated polyester textiles (50 nm 6:1 (laboratory, pilot), and 50 nm 2:1 (commercialized))^a. The higher amount of required silver in the pilot and in the estimated commercialized plant in comparison to the laboratory plant are due to a change in the production setting. Despite other efficiency gains, the amount of waste silver is higher in the upscaled plants. Silver concentration for all coatings: 4,4 μg nanosilver $\cdot cm^{-2}$.

	laboratory plant	scale pilot scale plant	commercialized plant (estimated)[58]
electricity [kWh]	183,6	10,6	0,13
coating time [h]	19,15	1,06	0,04
process time [h]	76,59	3,97	0,05
ethylene [g]	175,9	10,97	0,06
carbon dioxide [g]	1055	206,6	$1,9 \cdot 10^{-3}$
argon [g]	4398	31,25	$0,50 \cdot 10^{-3}$
nitrogen [g]	–	0,218	–
silver [g]	29,7	38,4	31,9

^a6:1 (2:1): 6 (2) mol carbon dioxide are injected per mol ethylene

Table S.8: Material requirements for the polymeric nanosilver layer and emissions

precursor	input [moles m^{-2}]	input [moles m^{-2}]	input for layer [mmoles m^{-2}]	emissions [moles m^{-2}]
ethylene	0,39	C: 3,13	C: 2,88	C: 3,13
carbon dioxide	2,35	O: 4,69	O: 2,16	O: 4,69
argon	0,39	H: 1,56	H: 0,96	H: 1,56
nitrogen	0,02			

Table S.9: Plasma polymerization with silver co-sputtering, 1 m^2 , 50 nm , 4%-w/w silver (coating). "realistic" is a currently planned commercial plant. The "optimistic" case is a best case estimation of an optimized future commercial production plant [58]. For the "recycling", a standard ecoinvent silver recycling process was chosen for the silver waste.

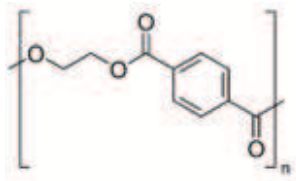
	unit	laboratory plant	pilot plant	commercial plant, realistic	commercial plant, optimistic	commercial plant, with silver recycling
recovered products						
silver, secondary, at precious metal refinery/SE U						0.031852
Materials/fuels						
nitrogen, liquid, at plant/RER U	<i>kg</i>		2.19E-4			
argon, liquid, at plant/RER U	<i>kg</i>	4.398	0.016	4.96E-7	4.96E-7	4.96E-7
carbon dioxide liquid, at plant/RER U	<i>kg</i>	1.055	0.103	1.09E-6	1.09E-6	1.09E-6
ethylene, average, at plant/RER U	<i>kg</i>	0.176	0.011	3.48E-7	3.48E-7	3.48E-7
silver, at regional storage/RER U	<i>kg</i>	0.030	0.038	3.19E-2	1.60E-2	3.19E-2
transport, lorry 3,5-20 t, fleet average/CH U	<i>tkm</i>	0.470	0.010	4.59E-7	4.59E-7	4.59E-7
Electricity/heat						
electricity, medium voltage, production UCTE, at grid/UCTE U	<i>kWh</i>	183.594	10.582	0.133	0.133	0.133
Emissions to air						
carbon dioxide, fossil	<i>kg</i>	1.607	0.138			
Emissions to water						
water	<i>kg</i>	0.452	0.028			
Outputs to technosphere						
water treatment, class 3	<i>m³</i>	0.000452	0.000028			

S.2 LCI of Polyester Textiles

A non-dyed, knitted polyester T-shirt is assessed in order to compare it to the nanosilver polyester T-shirt. An LCI is built up based on [20, 50, 71, 89, 95, 117] and own calculations as discussed below. The biocide coating of the polyester fibers is the same for *nanoAg-TCP* and for *nanosilver* and hence is not distinguished below.

PET is the condensation product of the reaction between terephthalic acid (TPA) and ethylene glycol [63] and becoming increasingly common to recycle after use by re-melting and extruding it as fibers [15]. Polyester properties are presented in Table S.10.

Table S.10: Properties of polyester [10, 15, 63].

	terephthalic acid	
educts for polyester synthesis	$C_6H_4(COOH)_2$	
	dimethylterephthalate	
	$C_6H_4(COOCH_3)_2$	
	mono ethylene glycol	
	$C_2H_6O_2$	
melting temperature [°C]	260-265	
molar enthalpy of formation	140	
ΔH_f [$kJ \cdot kg^{-1}$]		
mean filament diameter [μm]	25	
mean fiber diameter [μm]	250	
specific gravity [$g \cdot cm^{-3}$]	1,38	

For 1 *kg* PET, 0,87 *kg* TPA and 0,50 *kg* EG are required. PET is converted to PET-textile with a material loss of 3-5% [33]. In total, 1,05 *kg* PET resins are required to produce 1 *kg* fiber coating with particles from FSP. Ready-to-use PET resins (ECOINVENT) are melted. PET resins are dissolved in ethanol and *nanoAg-TCP* is added to this solution. PET is nearly infinitely soluble in ethanol [63]. On commercial scale, 0,25 *kg* ethanol is used for 1 *kg* of PET resins [33].

PET spinning temperature is typically 285 °C and a stirred tank-reactor model with heat transfer is used to calculate the process with similar properties to a general PET-fiber reactor [33]: 0,12 *kWh* · *kg*⁻¹ to keep the process running during 30 *min* (incl. stirring). A melt heat of $c_p \Delta T = 0,15$ *kWh* · *kg*⁻¹ is added [33]. This net energy input (0,26 *kWh* · *kg*⁻¹) is used for the *nanoAg-TCP*-polyester-ethanol mixture. The mixture is then applied to common polyester fibers [37]. Coating velocity is estimated as 10 *m*² · *min*⁻¹ with a power requirement for the apparatus of 1 *kW* [37]. Cleansing is not taken into consideration. The applied polymer (here: *nanoAg-TCP*-polyester) is integrated by keying-in and adhesion. *NanoAg-TCP* is functionalized with chemical groups of terephthalic acid in a similar way as shown with polyacrylic acid (PAA) in [4] to serve as building blocks which can be adsorbed by polyester. This is done directly on the fibers, in contrast to PLASPU where the polymer is applied to the knitted polyester textile. The mixture has to be cooled rapidly with quench air in order to prevent processes like phase-transformations [25]. For PET, 5,3 *Nm*³ *air* · *kg*⁻¹ PET (6,36 *kg* *air* · *kg*⁻¹ PET) cools the coating mixture down to 20 °C. Ethanol is recovered (67,6%). Emissions during coating and flue gas cleaning are not included. The 5% polyester loss during fiber production is incinerated, calculated with the ALM model [97].

Production of polyester fibers: A continuous polycondensation of TPA and EG to PET is done

in a cascade of autoclaves (mixed and esterificated to *Di-EG*, then prepolymerization, finishing, and extruding with pumps) [33]. A reactor is modeled with following properties:

reactor volume	4100 <i>litres</i> (spherical)
reactor surface	12,39 m^2
simple disintegration for discontinous processes	0,3 $kWh \cdot kg^{-1}$ <i>content</i>
heat transfer area	0,1 $m^2 \cdot dm^{-3}$ (isothermic temperature conduction)
heat transfer coefficients	250 $W \cdot m^{-2} \cdot K^{-1}$
stirrer drives	0,005 $kWh \cdot kg^{-1}$

This stirred tank reactor model [33] with heat transfer was used for all synthesis processes when process data (but no ECOINVENT data) were available. The level of detail was kept low because a generic reactor was the only way to cope with the numerous processes in an efficient way. Uncertainties include amongst others scale-up options, different tank shapes/volumes, time of reaction needed and the heat transfer coefficients. An important aspect of modeling is to stick to the thermodynamic laws, i.e. conservation of energy in the reactor. A general form for the energy balance equation is equation 8 [84]:

$$\begin{aligned}
 & [Amount\ of\ energy\ added\ per\ unit\ time] - [Amount\ of\ energy\ removed\ per\ unit\ time] \\
 & + [Amount\ of\ energy\ generated\ per\ unit\ time] = [Accumulation\ of\ energy\ per\ unit\ time]
 \end{aligned} \tag{8}$$

The heat of reactions is considered not to be significant, therefore an adiabatic reactor is used. Hence, *energy generated by unit time* is neglected and only the amount of *energy added* is calculated in order to keep the temperature of the reaction constant. Heating of the reactants results in an energy flux per unit area within the control volume of the vessel. An estimated steady-state heat conduction per unit area of $250\ W\ m^{-2} \cdot K^{-1}$ [33] is used for the whole reaction time. Heat flows through a single-layer spherical wall of homogeneous material and uniform inner and outer surface temperatures. ΔT is always the difference between reaction temperature and room temperature (25 °C). Pressure variations are neglected. Reactions to produce the precursors are calculated based on literature (pressure, temperature, time, pre-precursors) and own calculations (heat of reaction, heat capacities, reactor properties).

The amount of energy which is required for this reactor is defined as heat and power added to the system to start (instantaneously) and to keep the reaction running. No energy flows are included and it is estimated that internal energy is the dominant part (disregard of kinetic and potential energies). Released heat of reaction is neglected because overall enthalpy of the modelled reactions did not show highly negative enthalpies. Stirring is included for the *nanoAg-TCP-PET* process. The reactor is assumed to be well-mixed and in adiabatic condition. A constant volume reactor (for fluids and gases) is modeled, but pressure changes are not included. Heat capacities for constant-pressure are taken (the two total heat capacities are related as $C_v = C_p - nR$). Heat flows and heat capacity of the mixture are considered to be constant over the composition and temperature range. With the partial molar heat capacities the total heat capacity is calculated (Equation 9, with the previously stated estimations):

$$f \cdot V_R \cdot \rho_{mix} \cdot C_p = \sum_{j=1}^n C_{pj} \cdot n_j \tag{9}$$

Process power input is calculated the following:

$$P_{in} = \frac{A_r \cdot k_H \cdot \Delta T \cdot t}{f \cdot \rho_{mix} \cdot V_r} \tag{10}$$

- C_p = heat capacity of mixture [$J \cdot kg^{-1} \cdot K^{-1}$]
 C_{pj} = heat capacity of single compound [$J \cdot kg^{-1} \cdot K^{-1}$]
 n_j = mass of single compound [kg]
 A_r = surface area of reactor [dm^3]
 k_H = heat transfer coefficient [$W \cdot m^{-2} \cdot K^{-1}$]
 ΔT = temperature difference between media [K]
 t = reaction time [s]
 f = filling factor [$-$]
 ρ_{mix} = density of mixture [$kg \cdot dm^{-3}$]
 V_r = volume of reactor [dm^3]
 P_{in} = power input [$(W \cdot s \cdot kg^{-1})$]

In the modelled process, total amount of carbon dioxide emissions are $7,02 g \cdot kg^{-1}$ polyester textile. Heat, fuel, and electricity requirements of the sale stores are considered once the T-shirts are delivered to the sale stores (for transportation, see chapter S.5). The energy use is attributed to the functional unit with economic allocation based on data in ([46], see Table S.13). They correspond well to other LCA dealing with sales and distribution inventory data (e.g. [56]).

Table S.11: Production chain of one polyester T-shirt

	amount	unit
process		
PET spinning, knitting, making up	1	kg
materials/fuels		
water, deionised, at plant/CH U	24.8	kg
electricity/heat		
electricity, medium voltage, production UCTE, at grid/UCTE U	5.62	kWh
waste to treatment		
treatment, sewage, to wastewater treatment, class 3/CH U	0.0248	m ³
product		
polyester fabric (own, incl making up)	1	kg
materials/fuels		
polyethylene terephthalate, granulate, bottle grade, at plant/RER U	1.05	kg
PET spinning, knitting, making up	1.05	kg
transport, lorry >32 t, EURO5/RER U	0.1	tkm
heat, natural gas, at boiler condensing modulating < 100 kW/RER U	9.7	MJ
transport, freight, rail/RER U	0.2	tkm
waste to treatment		
disposal, polyethylene terephthalate, 0.2% water, to municipal incineration/CH U	0.05	kg
product		
polyester T-Shirt	1	p
materials/fuels		
polyester fabric (own, incl making up)	0.13	kg
distribution and Sale	1	p
electricity/heat		
electricity, medium voltage, production UCTE, at grid/UCTE U	1.93	kWh

Table S.12: Energy and water requirements for 1 kg polyester textile [20, 50, 95, 117].

process stage	water use [kg]	electricity use [kWh]
spinning		3
knitting		1,5
finishing	24,8	0,82
making up		0,3
total	24,8	5,62

Table S.13: Energy requirements for sale stores and allocated to one T-shirt (turnover 2004: $7,06 \cdot 10^9$ EUR; average T-shirt price: 17 EUR)

	company total [46]	T-shirt
heat for distribution centers [MJ]	8.62E7	0,20
fuels for heating sale stores [MJ]	3.00E7	0,07
electricity (UCTE) [kWh]	3.62E8	0,85

S.3 Use Phase

LCI data are collected for washing detergents [56] and for washing machines (material and energy requirements). Additional energy and material requirements for tumblers are included.

Washing machines have improved in energy and water efficiency in recent years (Figures S.3 and S.4), and the trend is expected to continue. Introduced energy labeling causes additional incentives to the distributors to sell only *best in class* machines. The low energy requirements, correlated with lower environmental impacts and lower maintenance costs, are good selling arguments. However, market penetration of new washing and tumbling machines is slow because the life span of them is around 15 years [96]. It is expected that all tumblers will have heat pumps in future. Semidry cloths require 1,68 kWh per tumbling ($\sigma=0,16$) and the tumbling program "completely dry" requires 2,22 kWh per run ($\sigma=0,19$)[85]. It is assumed in the study that the semidry program is sufficient to get the polyester T-shirts completely dry, i.e. that the functionality of the semidry and the dry program for polyester textiles is the same. However, for the environmental awareness scenarios, different values for energy and water requirements for tumbling (and washing machines) are considered, because consumers do not necessarily environmentally optimize washing and tumbling behavior. The differences in electricity requirements for tumbling are caused by the semidry or alternatively dry program:

- *low environmental awareness*: arithm. mean of new washing machines and 2,22 kWh for tumbling, per cycle
- *mean environmental awareness*: arithmetic mean + 2 Stdev and 1,68 kWh for tumbling, per cycle
- *high environmental awareness*: projected optimistic value (estimation) for 2020 (see Figure S.3; 1,68 kWh for tumbling, per cycle)

Only few LCA studies (*e.g.* [74, 83, 118]) on washing machines are published, indicating that the greatest environmental impacts occur during use stage (electricity for water heating) [61]. The electricity mix and availability of water are important factors influencing the environmental impacts. In addition to the sources stated above, a good overview of electricity requirements of washing machines is given under <http://db.eae-geraete.ch>. For long living washing machines, 85,5% of primary energy is required during use whilst maintenance (2,8%), production/distribution (11,7%) and disposal (0,01%) have small shares [29]. It was assumed for the disposal phase that metals are recycled whilst plastics are incinerated. However, due to the cut-off rule of 5%, only the production and use phase are considered in the impact assessment.

Polyester T-shirts have a low water absorption capacity which reduces the water volume needed. Water is changed 2 to 4 times during one washing, depending on the program. Concerning absolute energy and water demand, a lower loading of the washing machine results in a decreased demand, but the environmental impacts per washed T-shirt decrease when the machine is fully loaded (see also sensitivity analysis. For this study, average numbers on energy and material use for washing are gathered and standard deviations calculated (Table S.14). One load is considered equal to 15 polyester T-shirts. Polyester T-shirts are lightweight textiles, therefore the 5-6 kg washing load which is mostly defined as one full washing machine load [105] is not suitable (it would result in about 40 T-shirts per washing). The amount of water does not change substantially by changing the washing temperature [105] but the amount of required washing detergents depends on the physical properties: whether it is in the form of powder, tabs or perls. For this study, powder is taken [56] with an estimated dosage of 67,5 g per washing cycle [29]. Average washing habits of Swiss inhabitants set the basis (Figure S.2).

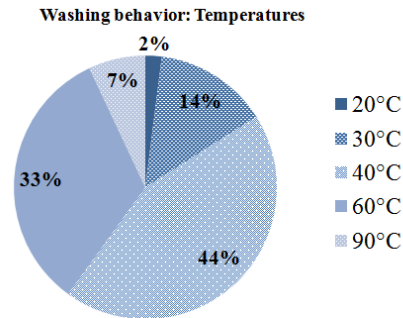


Figure S.2: Share of washing temperatures in Switzerland [93].

Table S.14: Energy and water requirements for of washing machines. Sources: [31, 93, 94, 103–105].

washing indicator	arithm. mean	σ	n
20 °C [$kWh \cdot cycle^{-1}$]	0,25	-	1
30 °C [$kWh \cdot cycle^{-1}$]	0,36	0,057	2
40 °C [$kWh \cdot cycle^{-1}$]	0,60	0,078	30
60 °C [$kWh \cdot cycle^{-1}$]	1,00	0,179	5
90 °C [$kWh \cdot cycle^{-1}$]	1,74	0,233	2
water [$litres \cdot cycle^{-1}$]	49,37	6,96	30
washing loads [$year^{-1} \cdot household^{-1}$]	241,5	57	4
life span [$years$]	10	-	

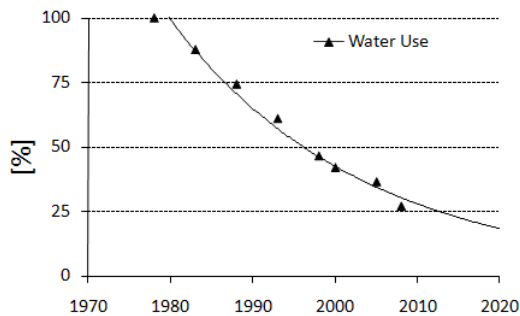


Figure S.3: Water use of washing machines over the last years and estimated for 2020. Base year is 1978 (100% = 180 litres, logistic regression curve $R^2=.98$, std error of the estimate: .069), values are normalized and based on absolute values in [41, 91].

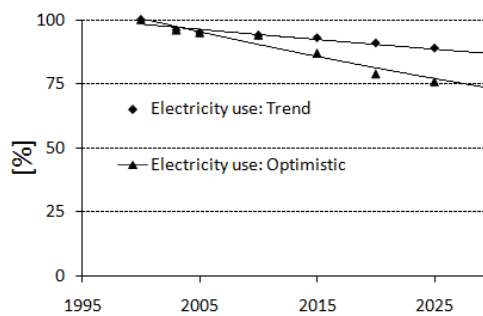


Figure S.4: Electricity use of washing machines over the last years and estimated possible future pathways. "Optimistic" is an optimistic development (information from producers) and trend is the extrapolation of past time series. Base year is 2000 (100% = 1,2 kWh, logistic trend curves, potential: $R^2 = .97$, std error of the estimate: .023; trend: $R^2 = .95$, std error of the estimate: .011), values are normalized, based on numbers in [16, 50] and Table S.14.

Table S.15: Material requirements for a washing machine (49,4 kg) [74, 83].

component	weight [kg]	material
washing tube	3,5	PP
cover	2,4	ABS
balance	2,3	PP
others	5,5	ABS
frame	11,6	steel
counter weights	17,4	cement
door hole, small parts	5,0	aluminium
electric components	0,7	copper
pipes	1,0	rubber

S.4 Disposal Phase

Disposal of nanomaterials is not regulated, mainly because of the lack of experimental data [64]. Therefore, nanosilver T-shirts are disposed like conventional T-shirts. About one third of the disposed textiles go to clothing drives before entering the final waste stream [108]. Recycling (5%) and disposal (incineration; 95%) [17] are modeled with the ECOINVENT v2.2 recycling and waste treatment unit processes for PET [34]. There may be potential environmental gains from keeping the polyester material separate from other materials because different recycling options exist for polymer material. Reuse, mechanical recycling of polyester, de-polymerization, and pyrolysis (which recovers a mixture of lower molecular weight compounds) are economically feasible [11, 78] and increasingly applied.

Table S.16: Fate of silver [24, 36, 40, 49, 59, 64]. Values as fractions and as mass per T-shirt. Emissions are valid for the use phase after wastewater treatment. Silver in the wastewater treatment sludge is incinerated and end up in landfill as solid incineration residue. In brackets: silver in associated form [106], which was not assessed for aquatic toxicity.

	silver [% of applied]	silver [mg/T – shirt]
biocidal content	100	30,89
freshwater emission, from WWTP	6,25 (4,69)	1,93 (1,45)
from incineration of T-shirts and sludge, to air	0,19	0,058
from incineration of T-shirts, to landfill		3,14
from incineration of sludge, to landfill		25,76
from incineration, to landfill (total)	93,56	28,9
longterm leaching, from landfill, to groundwater		0,19

S.5 Transportation

Transportation of the precursors and the products from supplier to producer within Switzerland were included for the nanosilver production processes. For all other processes, standard distances were applied to the LCI: For most materials, 100 km with lorry (3,5-20 tons) and 200 km with railway were considered to be typical for Switzerland. Average levels of emissions per ton kilometre for various modes of transportation are given in ECOINVENT v2.2 [5]. It was assumed that 50% of the polyester cloths are imported by truck from Eastern Europe (1000 km transport distance, lorry EURO 5, > 32 tons) [18, 46]. The remaining textiles are imported from Asia, either by flight (9%) or ship (91%) with further transportation by train (600 km) and truck (100 km) [5, 18, 46].

Table S.17: Distribution and Sale of one T-shirt.

name	amount	unit	comment
transport, aircraft, freight, intercontinental/RER U	0,117	tkm	9% China-Europe
transport, freight, rail/RER U	0,026	tkm	ecoinvent standard distance Europe
transport, lorry 16-32 t, EURO5/RER U	0,0117	tkm	ecoinvent standard distance Europe(90%)
transport, lorry 16-32 t, EURO4/RER U	0,0013	tkm	ecoinvent standard distance Europe (10%)
transport, transocenanic freight ship/OCE U	1,183	tkm	91% China-Europe
electricity, medium voltage, UCTE, at grid	0,854	kWh	distribution and sale
heat, light fuel oil, at boiler 100 kW, non-modulating CH/U	0,071	MJ	heat for distribution and sale
natural gas, burned in boiler modulating < 100 kW/RER U	0,204	MJ	fuel for distribution and sale

S.6 Background Data (Including Steam and Electricity)

Energy outputs from exothermic reactions are not credited to the reactions because of lacking information. Therefore, a quantitative assessment is not conducted. However, for exothermic reactions, recovered heat is estimated to serve as energy input to start the reactions (activation energy). For LCA modelling, steam ($3,228 \text{ MJ} \cdot \text{kg}^{-1}$) is used as process heat input if no other energy input is explicitly stated in the information sources (ECOINVENT 2.2). Unless otherwise noted, thermodynamic data for reactions (heats of reaction, equilibrium constants, specific heat capacities) are calculated from the tables of the NIST database [1] and the yield is derived from literature. Processes are assumed to take place in non-ideal gaseous state. Material and energy losses as well as waste streams are considered. For production processes, medium voltage UCTE electricity (1-24 kV) and for end user consumption (*e.g.* washing), low voltage UCTE electricity (<1000 V) at European grid is taken. If data availability for precursor production processes was poor, stoichiometric balances are used to determine the raw materials and energy demand. If no specific information about production processes was available, a 95% yield is estimated following the ECOINVENT v2.2 methodology [5].

S.7 Allocation

Environmental impacts from multi-output processes are allocated according to physical relationships (mass basis). Recovered solvent is subtracted from primary solvent demand. Information about distillation for recovery and disposal of solvents is obtained with ECOSOLVENT tool [19]. ECOSOLVENT is designed to identify environmentally preferable waste solvent treatment options [19] and is used in this study to calculate the absolute environmental impacts of waste solvent treatment in different processes.

S.8 Uncertainty Analyses

Unit process data are neither vertically nor horizontally aggregated in this study. Unit processes are described with mean values and underlying uncertainty distributions (already modeled for ECOINVENT 2.2 unit processes). Especially nanotechnology applications which are moving forward rapidly entail large levels of uncertainty. Quantitative uncertainties of the analyzed processes as such cannot be derived from the available information. Therefore, a simplified standard procedure based on a Pedigree matrix is used [5]. These uncertainty factors are set for each gathered data point and then combined to a system process uncertainty ([35], Table S.18). Lognormal distributions are used for system processes and for nearly all unit processes. Data sources for technosphere inputs and outputs and for elementary flows are assessed according to the six characteristics *reliability*, *completeness*, *temporal correlation*, *geographic correlation*, *further technological correlation* and *sample size*. Then, an uncertainty factor is attributed to each of these characteristics and the square of the geometric standard deviation is calculated (variance, 95%-interval) according to the ecoinvent methodology [35]. Finally, a Monte Carlo Simulation (1000 iterations) was conducted to calculate the uncertainty of the accumulated LCI data.

Table S.18: Pedigree approach values for the new unit processes.

subprocess	process	variance σ^2
2-ethylhexanoic acid	nanoparticle production process FSP	1,53
nanoparticle production process FSP	coating ($5 \mu m$)	1,07
distillation pyridine (recovery rate: 32.2%)	tributylphosphate	1,57
n-butyraldehyde	2-ethylhexanoic acid	1,55
pyridine	tributylphosphate	1,53
silver nitrate	silver octanoate	1,53
silver octanoate	nanosilver production process FSP	1,56
treatment, sewage, unpolluted, to wastewater treatment class 3 / CH U	nanoparticle production process FSP	1,11
tributylphosphate	nanosilver production process FSP	1,53
distillation water with salt impurities (NO_3^- , NH_4^+)	silver nitrate	1,57
materials (total)	washing & tumbling machine	1,14
washing detergents	washing cycle	1,00

S.9 Substance Properties, Characterization Factors, and Concentrations of Silver and Triclosan in T-shirts

Nanosilver

Nanosilver is toxic to bacteria [13, 14, 59] and applied to textiles in order to reduce or inhibit bacteria growth. Antibacterial efficacy was proved for 1% and 2% evenly dispersed silver in polymers whilst 0,5% was revealed as bacteriostatic (tested genus: *Staphylococcus*) [12]. A general conclusion about the efficacy of specific nanosilver concentrations in textiles is difficult because it heavily depends on (i) the durability of the applied silver and (ii) the activity (= release of Ag-ions and silver nanoparticles) of the nanosilver coating. A reasonable nanosilver and triclosan concentration for commercialized T-shirts has been calculated based on data on nanosilver and triclosan textiles [13, 36, 38, 76, 77, 87, 110]. In addition, biocidal T-shirts with a high content of biocides was also taken into account, based on the mean concentration of products in early development stage (laboratory products) [23, 23, 38, 39, 53, 59, 60, 70, 73, 81, 82, 90, 101]. Due to the different toxicity of nanoAg-TCP compared to pure nanosilver [65] we used the commonly applied concentration in the master batch reported by the supplier and translated this concentration into the required mass for a T-shirt (see equations below and Table S.19) [66]. Table S.20 shows the data calculation procedure with the underlying data sources for the nanosilver T-shirt concentrations. Biocidal T-shirts with nanoparticles from FSP have a layer of *nanoAg-TCP* or pure nanosilver in a polyester matrix which is applied onto the polyester fibers before knitting. The nanosilver T-shirts with the plasma-polymer nanosilver layer have a knitted polyester textile as basis. The representative non-dyed polyester T-shirts contain 0,031 g pure nanosilver, 0,047 g *nanoAg-TCP* (0,93 mg pure nanosilver), or 0,022 g triclosan.

$$\varnothing = \sqrt{\frac{4 \cdot 10^{-5} \cdot tex}{\pi \cdot \rho}} \quad (11)$$

$$\rho_{\text{layer}} = \frac{0.02 \cdot \rho_{\text{Ag}} + 1.98 \cdot \rho_{\text{TCP}} + 8 \cdot \rho_{\text{PET}}}{10} \quad (12)$$

$$C_{\text{Ag}} = \frac{M_{\text{Ag}}}{M_{\text{shirt}} + M_{\text{nanoAg layer}}} \quad (13)$$

$$M_{\text{nanoAg layer}} = \frac{\pi \cdot (r_{\text{PET fibre}} + r_{\text{nanoAg layer}})^2 - r_{\text{PET fibre}}^2 \cdot M_{\text{PET T-shirt}}}{\rho_{\text{nanoAg layer}} \cdot tex_{\text{PET T-shirt}}} \quad (14)$$

\varnothing	= fibre diameter [μm]
ρ	= density [g cm^{-3}]
tex	= mass density of fibres [$\text{g } 1000\text{m}^{-1}$]
C_{Ag}	= silver concentration in textile [mg g^{-1}]
M	= mass [g]
L	= length [m]
r	= radius [μm]
A	= cross chapter surface [μm^2]
V	= Volume [cm^3]

In LCA, sewage treatment belongs to the technosphere and only the compounds that reach the eco-sphere are assessed (in addition to the ancillaries and energy requirements of the wastewater treatment plant). Out of the complete amount of nanosilver contained in the T-shirt, 67% were assumed to be released to the wastewater during the use phase (washing of T-shirt)[36]. In the wastewater treatment

Table S.19: Results of the equations 2.1 – 2.4, specifically for the *nanoAg-TCP* application

properties	value
diameter PET fibre [μm]	249
diameter PET fibre with <i>nanoAg</i> layer [μm]	259
cross chapter surface PET fibre [μm^2]	48662
cross chapter surface PET fibre and <i>nanoAg</i> layer [μm^2]	52650
cross chapter surface <i>nanoAg</i> layer [μm^2]	3989
volume <i>nanoAg</i> layer [cm^3]	31,11
tex [$g\ 1000\ m^{-1}$]	16,67
length polyester yarn [m]	7800
density polyester (ρ) [$g\ cm^{-3}$]	1,38
weight polyester T-shirt [g]	130
density <i>nanoAg-TCP</i> layer (ρ , 20% of Ag(2%)-TCP, 80% polyester) [$g\ cm^{-3}$]	1,75
<i>nano nanoAg-TCP</i> concentration in coating [$mg\ Ag-TCP\ g^{-1}\ coating$]	1,5
<i>nanoAg</i> (from <i>nanoAg-TCP</i> mass in T-shirt [$mg\ Ag\ T - shirt^{-1}$])	0,93
<i>nano nanoAg-TCP</i> mass in T-shirt [$mg\ Ag-TCP\ T - shirt^{-1}$]	46,7

Table S.20: Nanosilver, nanoAg-TCP, and Triclosan concentrations, normalized on one T-shirt (130 g). Commercial products: [13, 36, 38, 76, 77, 87, 110]; products in development stage: [23, 23, 38, 39, 53, 59, 60, 70, 73, 81, 82, 90, 101] .

	mean	interquartile range	standard deviation
triclosan ($g/T - shirt$), commercial products ($n = 20$)	0,0217	0,0280	0,0293
nanosilver ($g/T - shirt$), commercial products ($n = 45$)	0,0309	0,0172	0,0648
triclosan ($g/T - shirt$), products in development stage ($n = 10$)	3,72	4,81	2,54
nanosilver ($g/T - shirt$), products in development stage ($n = 14$)	2,56	5,201	3,29

plant, about 91% of the silver is removed (Table S.21). This value is calculated according to the USES-LCA tool which also includes a wastewater treatment plant (WWTP) tool, developed by Struijs et al. [106]. Even though this tool does not explicitly address emissions in the form of nanoparticles, the results are in the same range as reported for nanosilver specifically (e.g. [40]). In the scenario options of the WWTP tool, we chose freshwater as the emission compartment and steady state as temporal horizon. We considered the dissolved fraction as bioavailable (see Table S.16). The associated form is seen as much less toxic (e.g. associated with particulate and colloidal fractions, and/or silver in the form of silver sulfide in the effluent [3, 80]) and was not assessed for its aquatic toxicity in our study. Therefore, characterization factors for the aquatic environment were only applied to the released bioavailable silver fraction from the wastewater treatment plant. Characterization factors were calculated with the USES-LCA model. The predicted final environmental partitioning of the silver is modeled in a simplified way by the USES-LCA, neglecting for chemical transformations of the silver downstream of the wastewater treatment plant. This is a limitation of the study which should be investigated further in future research, because silver is expected to form strong sulfide complexes with comparatively low toxicity in natural waters [3, 80]. Note that the toxicity studies used for calculating the predicted no effect concentration - PNEC: this is one component of the characterization factor - are based on various chemical forms of commercially available silver compounds. Hence, the freshwater effect factor which was used in the simulation applies for silver in various speciations. This includes colloidal silver which often is in the nanosize range. However, the effect factor (based on 50 tested species) does not specifically address the chemical binding of silver to

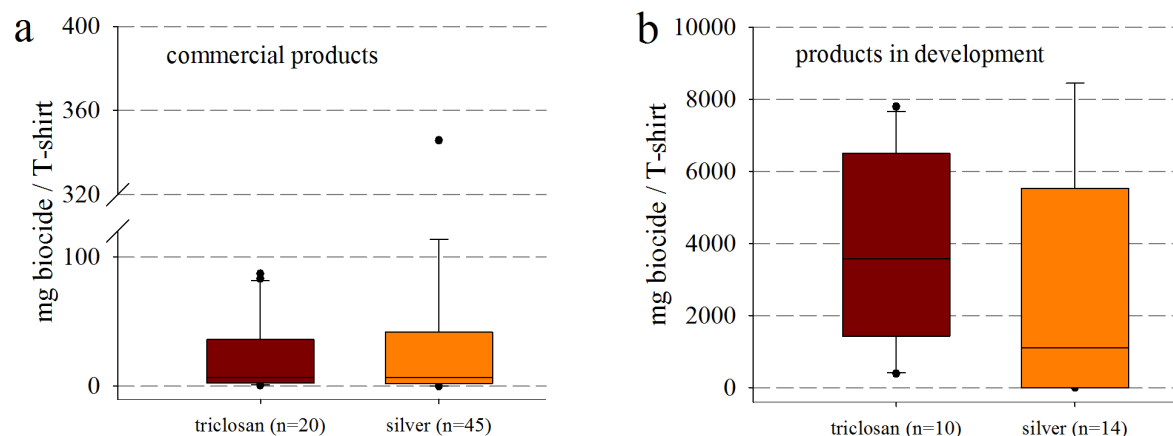


Figure S.5: Biocidal concentrations for commercial textiles (a) and for textiles in development (b) [13, 23, 23, 36, 38, 38, 39, 53, 59, 60, 70, 73, 76, 77, 81, 82, 87, 90, 101, 110]. *Boxes*: The central mark is the median, the edges of the boxes are the 25th and 75th percentiles, the whiskers embrace the 95% confidence interval of the applied biocide concentrations.

other substances.

The silver in the wastewater treatment sludge is incinerated (use in agriculture is not allowed in Switzerland). During incineration, the major part of the silver will be transferred to the slag and then put to landfill (no silver recovery assumed). Silver may leach over time into the groundwater (0.0064% of the applied silver, assuming a long-term transfer coefficient (theecoinvent landfill methodology includes long-term emissions with a time horizon of 60'000 years after present [24])). The emissions of (nano)silver into the environmental compartments are also discussed in S.4.

Potential human health impacts due to exposure to nanosilver were not calculated because the established characterization factors in USES-LCA could only be backed with values from Kim et al. and Sung et al. [54, 107]. Due to this limited number of human toxicity studies for airborne silver, only the results for the aquatic environment are interpreted, where most of the silver ends up.

Table S.21: WWTP efficiency, silver in wastewater = 100% ([106] and own calculations)

	WWTP elimination	freshwater emission	
	efficiency [%]	dissolved [%]	associated [%]
silver	90,7	2,3	7,0
triclosan	86,8	12,0	1,2

Table S.22: Selected characterization factors for the toxicity of silver and triclosan, according to the USES-LCA model ([49, 92])

	characterization factors [$kg\ 1,4 - DCB - eq\ kg^{-1}$]	
	freshwater	seawater
silver	81.45	14760
triclosan	17.89	0.3296

Triclosan

The estimated triclosan release during use of the T-shirt of 1.5% [77] is similar to the 0.55% assumed by the US EPA [81]. The average concentration of triclosan in T-shirts is estimated as $21.77 \text{ mg}/T - \text{shirt}$ with a freshwater emission (after elimination in WWTP) of 0.036 mg dissolved triclosan per T-shirt. We used the same input parameters for the USES-LCA tool as mentioned above with the only difference that the freshwater effect factor was based on 4 species (USES-LCA database). The remaining triclosan in the T-shirts is destroyed completely during incineration of the textiles. The same is the case for triclosan in the sludge from the WWTP. New characterization factors for the ecotoxicity of triclosan are established, applying the following chronic toxicity values: *Ceriodaphnia dubia* 7 days, NOEC: $1,46 \mu\text{g l}^{-1}$, rats NOAEL: $25 \text{ mg kg}^{-1} \text{ day}^{-1}$ [7].

S.10 Results from Selected Impact Assessment Methods

The production process in this study, without considering the supply chain, emits $44 \text{ kg CO}_2 - \text{eq}/\text{kg}$ for nanosilver and nanoAg-TCP. A further selection of impact assessment results are presented below and on the following page.

- Please note that the impact assessment results are heavily influenced by the choice of the electricity mix. Here we chose the european electricity mix (UCTE).
- For the investigated nanoparticle production processes, the main contributor to the Cumulative Exergy Demand (CExD) is the (renewable) water requirement for electricity production. In contrast to the non-renewable CED which was used, the indicator CExD considers renewables and assesses the quality of energy demand and includes the chemical, kinetic, hydro-potential, nuclear, solar-radiative and thermal exergies. The exergy of water contributes more than 90% to the total CExD via the hydropower electricity generation.
- The current impact assessment methods do not consider indoor (and outdoor) nanoparticle exposure.

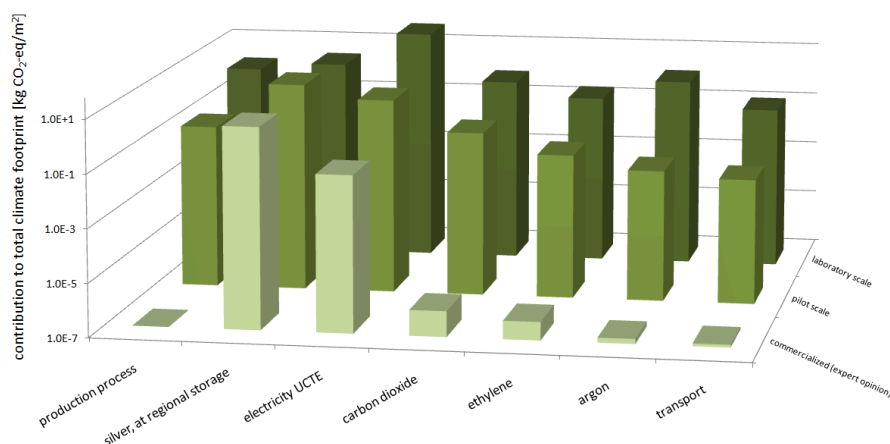


Figure S.6: Contribution of the most important material and energy inputs to the total climate footprint of 1 m^2 nanosilver textile, produced with PLASPU technology.

Table S.23: Impact assessment, selected results of the cradle-to-gate assessment of the production.

	1 kg nanosilver FSP	1 kg nanoAg-TCP FSP	1 m ² nanosilver coat- ing PlaSpu (pilot)	1 kg triclosan ^a	1 polyester T-shirt (production)
Non-renewable Cumulative Energy Demand (MJ - eq)	3230	1710	180	377	55.9
Cumulative Exergy Demand (MJ - eq)	64500	17800	4040	-	803
ReCiPe Midpoint (Europe, E/A) (kg 1, 4-DB - eq)					
human toxicity	73100	2570	2920	-	56.8
terrestrial ecotoxicity	1.35	0.0737	0.0438	-	0.00146
freshwater toxicity	19.9	0.984	0.821	-	0.0295
seawater toxicity	35900	1590	1470	-	46
ReCiPe Endpoint (Europe, E/A) (pts)	553	29.3	21	-	0.655

^aassessed with FineChem [113]

S.11 Scenario Construction

The selection of the impact variables, including their impact on each other (impact matrix) and analysis of their activity/passivity (system grid and system graph) was entirely done based on [114, 115]. The projections of the impact variables are combined into a pool of scenarios with different consistencies using a bottom-up approach (with software support SystAim (Tietje, 2005)). The estimated consistencies of the impact variable combinations are shown in Figures S.7 and S.8. A top-down approach based on [114] was used to search for three diverse scenarios in this pool named 'slow development', 'estimated development' and 'fast development'. The developed storylines behind these scenario titles are further explained in the next subsection. For instance, people's behavior is shaped by their (i) awareness of, (ii) interest in, and (iii) knowledge about nanotechnology. This will then influence the demand for nano-enabled T-shirts. A baseline scenario 'no nano' is used in addition to the three nanotechnology development scenarios to show the difference to a future without nanosilver clothes.

Consistency Matrix Nanotechnology (Formative Scenario Analysis)			d ₁ ¹	d ₁ ²	d ₂ ¹	d ₂ ²	d ₂ ³	d ₃ ¹	d ₃ ²	d ₄ ¹	d ₄ ²	d ₄ ³	d ₅ ¹	d ₅ ²
nanoparticle production and incorporation process	d ₁ ¹	flame spray pyrolysis (FSP)	-	-	-	-	-	-	-	-	-	-	-	-
	d ₁ ²	commercialized plasma process and FSP process (20%/80%)	-	-	-	-	-	-	-	-	-	-	-	-
washing pattern [ratio °C (60°C/40°C/30°C)] with corresponding water/energy use; incl Tumbler ¹⁾	d ₂ ¹	10/44/25	2	2	-	-	-	-	-	-	-	-	-	-
	d ₂ ²	20/44/13	2	2	-	-	-	-	-	-	-	-	-	-
	d ₂ ³	0/44/35	2	2	-	-	-	-	-	-	-	-	-	-
laws and regulations	d ₃ ¹	liberal: no additional regulation in addition to the silver effluent threshold and airborne exposure limits, no risk assessment and certification needed	2	2	2	2	2	-	-	-	-	-	-	-
	d ₃ ²	strict: ban of nanoparticle or lowered threshold value (e.g. 0.1 mg/l), risk assessment and certification needed	0	0	2	2	2	-	-	-	-	-	-	-
demand	d ₄ ¹	high = 3 T-shirts/cap/year (3 per cap/year)	2	0	2	2	3	2	1	-	-	-	-	-
	d ₄ ²	middle = 2 T-shirts/cap/year (5 per cap/year)	2	0	2	2	3	2	1	-	-	-	-	-
	d ₄ ³	low = 0.5 T-shirts/cap/year (5 per cap/year)	1	1	2	2	2	1	2	-	-	-	-	-
washing frequency nanosilver T-shirt	d ₅ ¹	high: same as polyester T-shirts	2	2	1	1	2	2	2	1	2	3	-	-
	d ₅ ²	low: half as polyester T-shirts	2	2	2	1	1	2	2	3	2	1	-	-

¹⁾ possible linkage to environmental awareness

Figure S.7: Consistency values for the combinations of impact variable characteristics; 0 = inconsistent, 1 = partial or weak inconsistency, 2 = consistent, 3 = complete consistency, the characteristics of the impact variables are coherent and support each other.

Consistency Matrix Environmental Behavior (Formative Scenario Analysis)			d ₁ ¹	d ₁ ²	d ₁ ³	d ₂ ¹	d ₂ ²	d ₂ ³	d ₃ ¹	d ₃ ²	d ₃ ³	d ₄ ¹	d ₄ ²	d ₄ ³
washing pattern [ratio °C (60°C/40°C/30°C)] with corresponding water/energy use; incl Tumbler ¹⁾	d ₁ ¹	10/44/25	-	-	-	-	-	-	-	-	-	-	-	-
	d ₁ ²	20/44/13	-	-	-	-	-	-	-	-	-	-	-	-
	d ₁ ³	0/44/35	-	-	-	-	-	-	-	-	-	-	-	-
washing amount over the lifetime of a T-shirt	d ₂ ¹	high = 100 times	1	2	3	-	-	-	-	-	-	-	-	-
	d ₂ ²	medium = 50 times	2	2	2	-	-	-	-	-	-	-	-	-
	d ₂ ³	low = 20 times	3	2	1	-	-	-	-	-	-	-	-	-
washing and tumbling load	d ₃ ¹	high: 20 T-shirts	1	2	3	3	2	1	-	-	-	-	-	-
	d ₃ ²	middle: 15 T-shirts	2	2	2	2	2	2	-	-	-	-	-	-
	d ₃ ³	low: 10 T-shirts	3	2	1	1	2	3	-	-	-	-	-	-
tumbling frequency T-shirts	d ₄ ¹	every second wash, completely dry	3	1	0	1	2	2	1	2	3	-	-	-
	d ₄ ²	every second wash, medium dry	2	2	2	2	2	2	2	2	2	-	-	-
	d ₄ ³	every fifth wash, medium dry	1	2	3	2	2	1	3	2	1	-	-	-

¹⁾ possible linkage to nanotechnology development

Figure S.8: Consistency values for the combinations of impact variable characteristics; 0 = inconsistent, 1 = partial or weak inconsistency, 2 = consistent, 3 = complete consistency, the characteristics of the impact variables are coherent and support each other.

S.12 Scenario Storylines

The scenario titles represent larger storylines taking into account the alternative combinations of impact variable characteristics.

In the scenario **slow development**, people are concerned about nanotechnology in general and have a rather negative opinion on the issue. Their risk perception of nanotechnologies is high and hence sensitive on the frequent nano-critical media reports. Although laws and regulations permit the development of nanosilver products, industry is careful in advertising their nanosilver functionalized textiles with naming nano explicitly. Even if the efficacy of the textiles is given and perceived by the public, nanosilver T-shirts are rarely bought because citizens worry about getting in direct contact with nanosilver. Nanosilver T-shirt production technologies develop slowly: The already commercialized FSP technology is used, but also the less efficient PLASPU technology is applied to a minor degree because the (although more expensive) coating is thinner than produced with the FSP technology.

The scenario **estimated development** displays a market situation where nanosilver in textile applications is widely available for private and public usage. People are interested in the new technology and its enhanced functionality. Some critical voices question the sustainability of these nano-enabled products. However, the market potential is not decreased with these opinions and is still high. The media covers nanotechnology with comprehensive, well documented reports, reflecting benefits and drawbacks of nanosilver applications. Laws and regulations set basic rules which are not restricting nanosilver innovations and economic development. FSP technology is applied for the production of commercially available nanosilver textiles.

The frequent positive reporting about findings of silver nanoparticles risk research contributes to a very high profit potential of nanosilver T-shirts in the scenario **fast development**. Citizens are risk tolerant even if a few concerned scientists do not support the fast development of nano-enabled textiles. These textiles constitute a high benefit and improve the quality in comparison to conventional polyester products. The consumer's demand is economically oriented and in parallel they are confident in supporting reduction of environmental impacts by buying the nanosilver T-shirts. With a supportive nanoregulation, there are no hurdles towards reaching a high market demand if the nanosilver textiles are sold with reasonable prices. Nanosilver textiles are produced with FSP technology.

S.13 Scenario Characteristics

In addition to the main text, some impact variable characteristics are hereby explained in more detail.

- **Laws and Regulations:** Potential and time frame are difficult to estimate: based on risk assessments, the level of threshold values is defined. Governmental R&D funds will influence technological development of nanosilver products. Even though not applied to textiles so far, an amendment to the directive "Restriction of Hazardous Substances (RoHS)" by the European Commission recommends a labelling or ban of nanosilver in electronic and electrical equipment [69]. Hence, a ban of nanosilver in textiles is not unimaginable. Triclosan is not regulated so far. For nanosilver, the two impact variable versions are either
 - *Liberal:* no regulation in addition the silver effluent threshold ($0,1 \text{ mg l}^{-1}$), no risk assessment and certification needed

– *Strict*: ban of nanoparticles or lower threshold value than for bulk silver ($<0,01 \text{ mg l}^{-1}$), risk assessment and certification needed

- **Demand for nanosilver T-shirts**: The total amount in stock was estimated 5 polyester T-shirts per person (whether with nanosilver/triclosan or not). The amount of nanosilver T-shirts is determined by the scenario. In the no-nano scenarios, the nanosilver T-shirts are replaced by triclosan treated T-shirts.
- **Use**: The washing loads and washing/tumbling frequency depend on the environmental awareness rather than on nanosilver (or other biocidal) T-shirts. The impact variables *amount of washings until disposal* and *washing and tumbling load* are chosen because of their immediate influence on the environmental impacts.

S.14 Scenario Results

Table S.24: Climate footprint (GWP IPCC 100 yrs 1.02) and characteristics of the no-nano scenarios.

BASE CASE without NANO			
low environmental awareness	mean environmental awareness	high environmental awareness	
26.00	17.33	13.00	washing cycles per year
26.00	8.67	1.30	tumbling cycles per year
72.66	24.52	9.65	use phase [$kWh \cdot yr^{-1}$]
26877	7167	2688	production phase [$tons CO_2 \cdot yr^{-1}$]
402811	149166	67274	use phase [$tons CO_2 \cdot yr^{-1}$]
2502	667	250	disposal phase [$tons CO_2 \cdot yr^{-1}$]
20	50	100	assumed life time [<i>washing cycles</i>]

Table S.25: Climate footprint (GWP IPCC 100 yrs 1.02) and characteristics of the nano scenarios.

production [$tons CO_2 \cdot yr^{-1}$]				
0.5	2	3	<i>nanosilver T-shirts per cap</i>	
1	0.8	0.5	<i>washing ratio compared to conventional T-shirts</i>	
slow development	estimated development	fast development		
28089	24941	19115	<i>low env awareness</i>	
7490	6651	5097	<i>mean env awareness</i>	
2809	2494	1912	<i>high env awareness</i>	
use [$tons CO_2 \cdot yr^{-1}$]				
0.5	2	3	<i>nanosilver T-shirts per cap</i>	
1	0.8	0.5	<i>washing ratio compared to conventional T-shirts</i>	
slow development	estimated development	fast development		
402811	370586	281968	<i>low env awareness</i>	
149166	137233	104416	<i>mean env awareness</i>	
67274	61892	47092	<i>high env awareness</i>	
disposal [$tons CO_2 \cdot yr^{-1}$]				
0.5	2	3	<i>nanosilver T-shirts per cap</i>	
1	0.8	0.5	<i>washing ratio compared to conventional T-shirts</i>	
slow development	estimated development	fast development		
2502	2302	1752	<i>low env awareness</i>	
667	614	467	<i>mean env awareness</i>	
250	230	175	<i>high env awareness</i>	

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