Waste Not Want Not: Life Cycle Implications of Gold Recovery and Recycling from Nanowaste

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Supporting Information

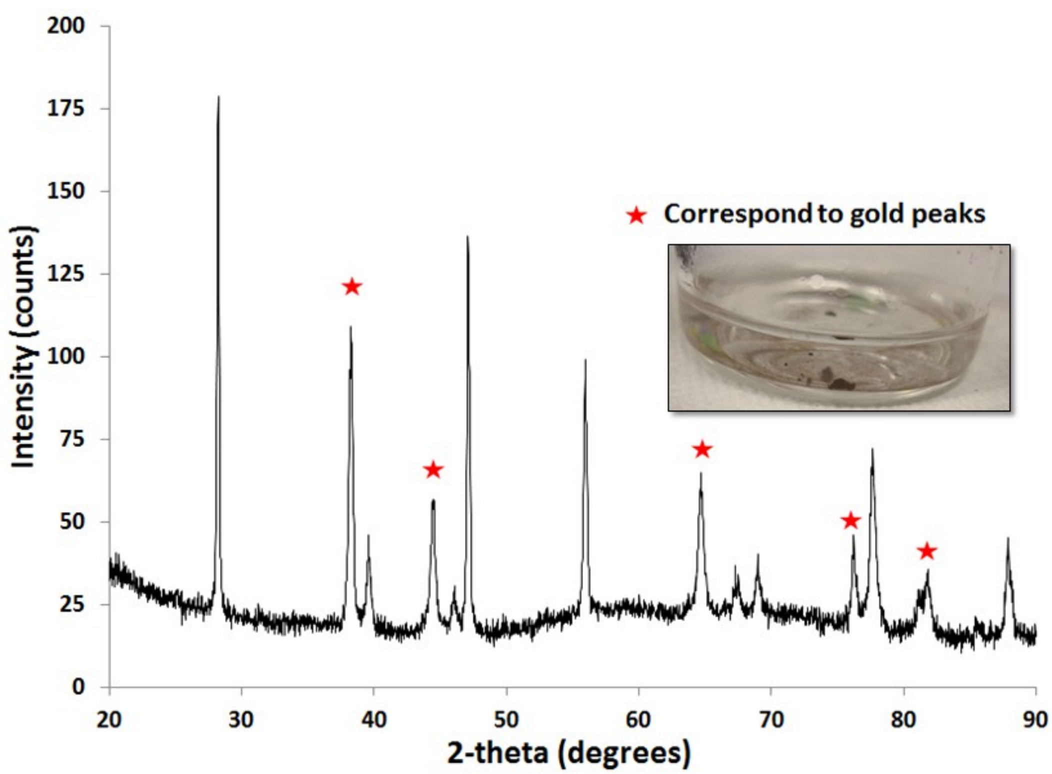


Figure S1: Powder X-ray diffraction of recovered gold. The highlighted peaks correspond to gold peaks. The unidentified peaks are presumably due to impurities. XRD measurements were performed on a Rigaku MiniFlex II instrument (Rigaku Americas, The Woodlands, TX, USA).

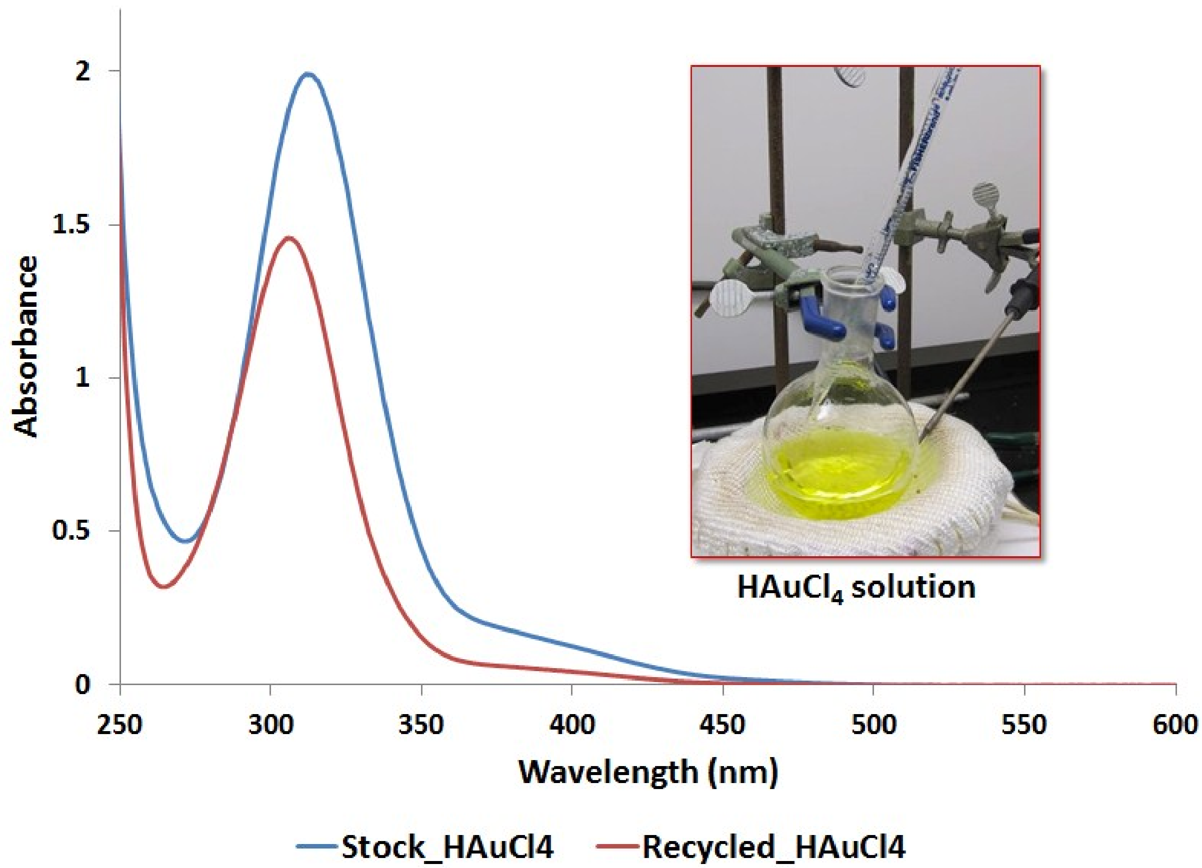
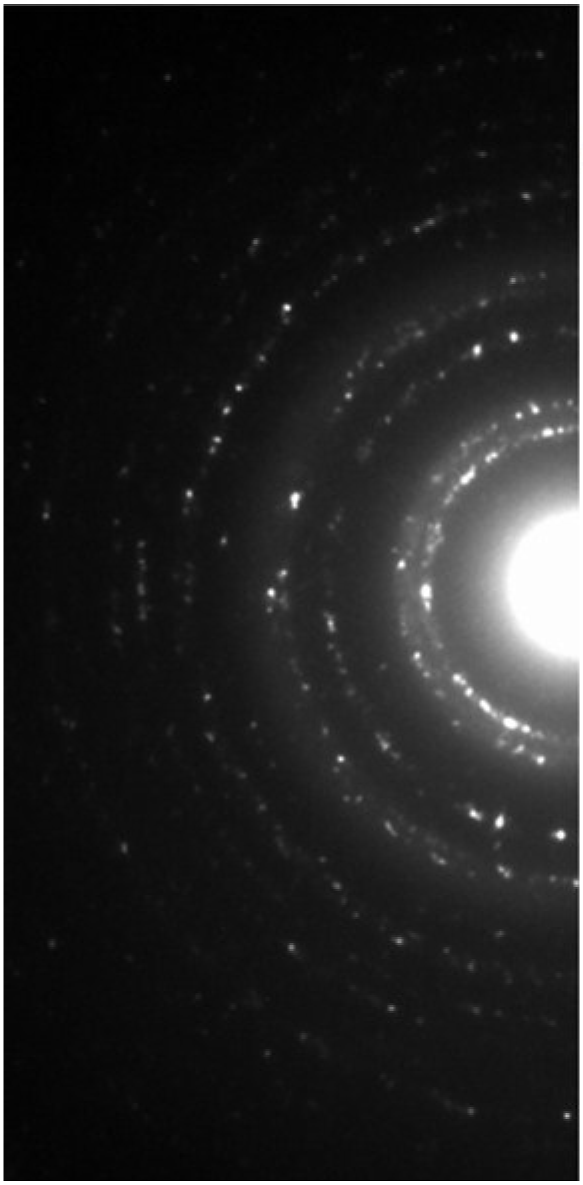


Figure S2: UV-vis spectra of recovered gold chloride and chloroauric acid standard. All measurements were using a Cary 5000 UV-Vis-NIR spectrophotometer (Agilent, Santa Clara, CA). All samples were scanned in quartz cuvettes (Starna, model# 1-Q-10) with 10 mm path length.



Calculated d-spacing (Å) from recovered gold	d-spacing for gold (Å) reported in the literature
1.25, 1.19	1.23, 1.18
1.46	1.44
2.06	2.04
2.39	2.36

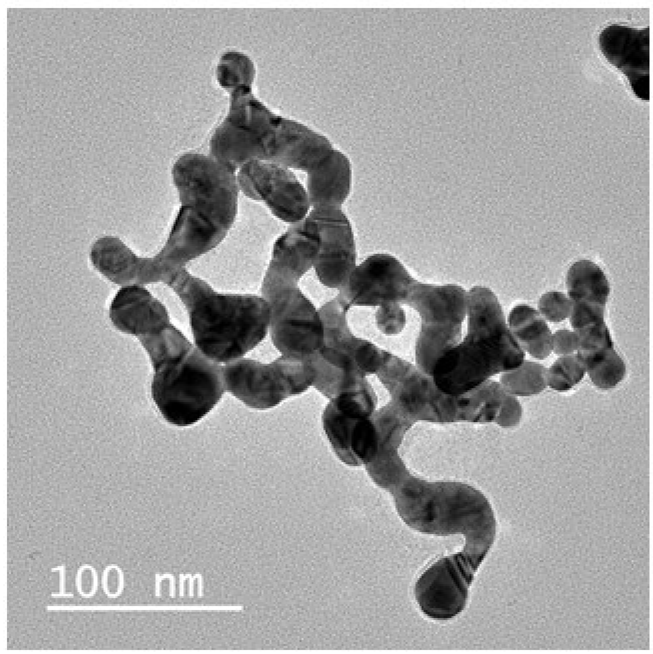


Figure S3: Crystal structure information from SAED measurements confirms that the recovered precipitate is gold. TEM image shows highly aggregated citrate-reduced AuNPs produced by this approach. The existence of 'throats' between individual AuNPs provides evidence of AuNP coalescence. All TEM and SAED measurements were performed on a JEOL 2100 (JEOL, Peabody, MA, USA)

Table S1: Life cycle inventories for custom defined chemicals AuNP synthesis and recovery steps.

[Custom defined] Chloroauric acid (1 mg)		
Gold {US}| production | Alloc Def, S	0.72	mg
Hydrochloric acid, without water, in 30% solution state {RER}| hydrochloric acid production, from the reaction of hydrogen with chlorine | Alloc Def, S	0.13	mg
Chlorine, gaseous {RER}| sodium chloride electrolysis | Alloc Def, S	0.39	mg
[Custom defined] Trisodium citrate (1 mg)		
Citric acid {GLO}| market for | Alloc Def, S	0.51	mg
Soda ash, light, crystalline, heptahydrate {GLO}| market for | Alloc Def, S	0.66	mg
[Custom defined] Hydrobromic acid (1 mg)		
Phosphorus, white, liquid {GLO}| market for | Alloc Def, S	0.13	mg
Bromine {GLO}| market for | Alloc Def, S	0.99	mg
Water, deionised, from tap water, at user {GLO}| market for | Alloc Def, S	0.22	mg
[Custom defined] α-cyclodextrin (1 mg)		
Potato starch {GLO}| market for | Alloc Def, S	1.67	mg
Water, deionised, from tap water, at user {GLO}| market for | Alloc Def, S	16.67	mg
[Stirring] Electricity, medium voltage {NPCC, US only}| market for | Alloc Def, S	0.02	MJ
[Heating] Electricity, medium voltage {NPCC, US only}| market for | Alloc Def, S	0.18	MJ

Table S2: Life cycle inventories for AuNP synthesis steps.

Citrate-reduced gold nanoparticles (1 mg)		
[Custom defined] Chloroauric acid	1.73	mg
[Custom defined] Trisodium citrate	5.08	mg
Water, deionised, from tap water, at user {CH}| production | Alloc Def, S	505.08	g
Tap water {CH}| market for | Alloc Def, S	30.00	g
Cleaning solvents		
Hydrochloric acid, without water, in 30% solution state {RER}| hydrochloric acid production, from the reaction of hydrogen with chlorine | Alloc Def, S	1.81	mg
Nitric acid, without water, in 50% solution state {RER}| nitric acid production, product in 50% solution state | Alloc Def, S	0.72	mg
[Stirring] Electricity, medium voltage {NPCC, US only}| market for | Alloc Def, S	0.01	MJ
[Heating] Electricity, medium voltage {NPCC, US only}| market for | Alloc Def, S	0.08	MJ

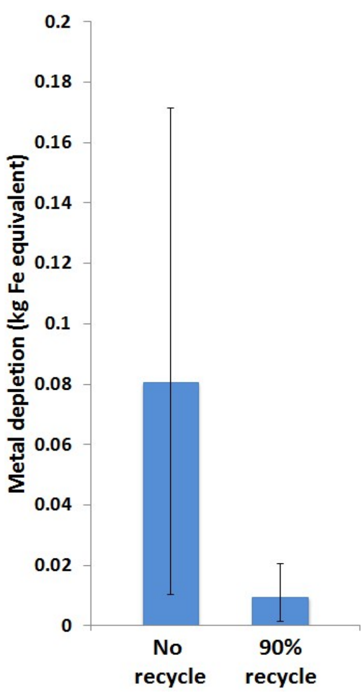
Table S3: Life cycle inventories for AuNP recovery steps to treat 1 mg of gold nanowaste

AuNP precipitation using NaCl		
Sodium chloride, powder {RER}| production | Alloc Def, S	7.42	mg
Dissolution of precipitate using HBr and HNO_3, followed by pH adjustment using KOH		
[Custom defined] Hydrobromic acid	680.81	mg
Nitric acid, without water, in 50% solution state {GLO}| market for | Alloc Def, S	216.28	mg
Potassium hydroxide {GLO}| market for | Alloc Def, S	148.45	mg
Water, deionised, from tap water, at user {GLO}| market for | Alloc Def, S	1015.38	mg
Gold : α-cyclodextrin complex formation		
[Custom defined] α-cyclodextrin	9.88	mg
Gold : α-cyclodextrin complex resuspension using sonication		
Water, deionised, from tap water, at user {GLO}| market for | Alloc Def, S	5076.92	mg
Electricity, medium voltage {NPCC, US only}| market for | Alloc Def, S	4.21	kJ
Gold precipitation from gold : α-cyclodextrin complex		
Sodium hydrogen sulfite {GLO}| market for | Alloc Def, S	138.95	mg
[**Note:** Sodium metabisulfite ($Na_2S_2O_5$) was not available in the EcoInvent inventory. Instead, we used sodium hydrogen sulfite ($NaHSO_3$) in the LCA models]		
Dissolution of recovered gold in aqua regia		
Hydrochloric acid, without water, in 30% solution state {RER}| hydrochloric acid production, from the reaction of hydrogen with chlorine | Alloc Def, S	452.99	mg
Nitric acid, without water, in 50% solution state {RER}| nitric acid production, product in 50% solution state | Alloc Def, S	180.35	mg
HNO_3 boil-off , HCl addition and pH adjustment using KOH to obtain chloroauric acid for AuNP synthesis from recovered gold		
Hydrochloric acid, without water, in 30% solution state {RER}| hydrochloric acid production, from the reaction of hydrogen with chlorine | Alloc Def, S	604.15	mg
Water, deionised, from tap water, at user {GLO}| market for | Alloc Def, S	10153.83	mg
Potassium hydroxide {GLO}| market for | Alloc Def, S	7.42	mg
Electricity, medium voltage {NPCC, US only}| market for | Alloc Def, S	70.0	kJ

Table S4: Effect of a 5-fold increase in different inputs on life cycle impacts in the 90%-recycle scenario. The percentages in the parenthesis show the increase in impacts relative to the baseline 90%-recycle scenario. The impacts increased substantially with increase in acid use and energy consumption. Similar trends were observed for the 10%- and 50%-recycle scenarios (data not shown).

Impact category	90%-recycle scenarios					
	Baseline 90%-recycle scenario	90%-recycle scenario with 5-fold in increase in cyclodextrin	90%-recycle scenario with 5-fold in increase in sodium metabisulfite	90%-recycle scenario with 5-fold in increase in DI water	90%-recycle scenario with 5-fold in increase in acid use	90%-recycle scenario with 5-fold in increase in energy use
Metal depletion (g Fe eq)	9.591	9.617 (+0.28%)	9.673 (+0.86%)	9.599 (+0.09%)	10.852 (+13.15%)	10.274 (+7.13%)
Freshwater ecotoxicity (g 1,4-DB eq)	4.244	4.258 (+0.32%)	4.261 (+0.38%)	4.249 (+0.10%)	4.562 (+7.48%)	4.712 (+11.01%)
Human toxicity (g 1,4-DB eq)	207.604	207.827 (+0.11%)	208.015 (+0.20)	207.658 (+0.03%)	218.061 (+5.04%)	214.964 (+3.55%)
Fossil fuel depletion (g oil eq)	12.222	12.542 (+2.62%)	12.555 (+2.73%)	12.249 (+0.22%)	20.050 (+64.05%)	23.660 (93.59%)

Uncertainty analysis of life cycle impact assessment. LCA results typically involve correlated uncertainties. For example, the 90%-recycle and no-recycle models use chemicals and processes from the life cycle inventories (such as gold, water, electricity, etc.) that are common to both scenarios. In such cases, the uncertainty in the LCA inventory for a chemical (say, gold) is common to all recycle scenarios, and is therefore correlated. In the case of correlated uncertainties, differences in results may be statistically significant, even if the error bars at the 95% confidence level overlap (Figure S4, left). Therefore, we have chosen to represent uncertainty by comparing the actual Monte Carlo simulations. As seen from the tabulated results in Figure S4 (right), of the 1000 runs performed during Monte Carlo simulation, the majority show that recycling has lower environmental burdens in the key impact categories (ecotoxicity, eutrophication, and metal depletion).



Impact category	Percent of 1000 Monte Carlo simulations showing: Impact of no-recycle scenario is less than that of 90%-recycle scenario	Impact of 90%-recycle scenario is less than that of no-recycle scenario
Metal depletion	0.0%	100.0%
Freshwater ecotoxicity	2.2%	97.8%
Marine ecotoxicity	1.8%	98.2%
Human toxicity	25.8%	74.2%
Freshwater eutrophication	0.0%	100.0%
Marine eutrophication	0.0%	100.0%

Figure S4 – (Left) The overlapping error bars for 95% confidence intervals should not be interpreted as statistically insignificant differences, because these LCA models involve correlated uncertainties. (Right) The majority of the Monte Carlo simulations showed that 90%-recycle scenario has lower impact than no-recycle scenario in terms of metal depletion, toxicity and eutrophication.

IIn figures S5, S6 and S7, we show the percentage of the Monte Carlo simulations for different recycle scenarios. For each of the impact categories, longer hatched bars indicate that for the majority of Monte Carlo simulations, recycling has lower impact than the no-recycle scenario. Longer solid bars, on the other hand, indicate that no-recycle scenarios have lower impact in those impact categories (as seen, for example, in the Climate Change category).

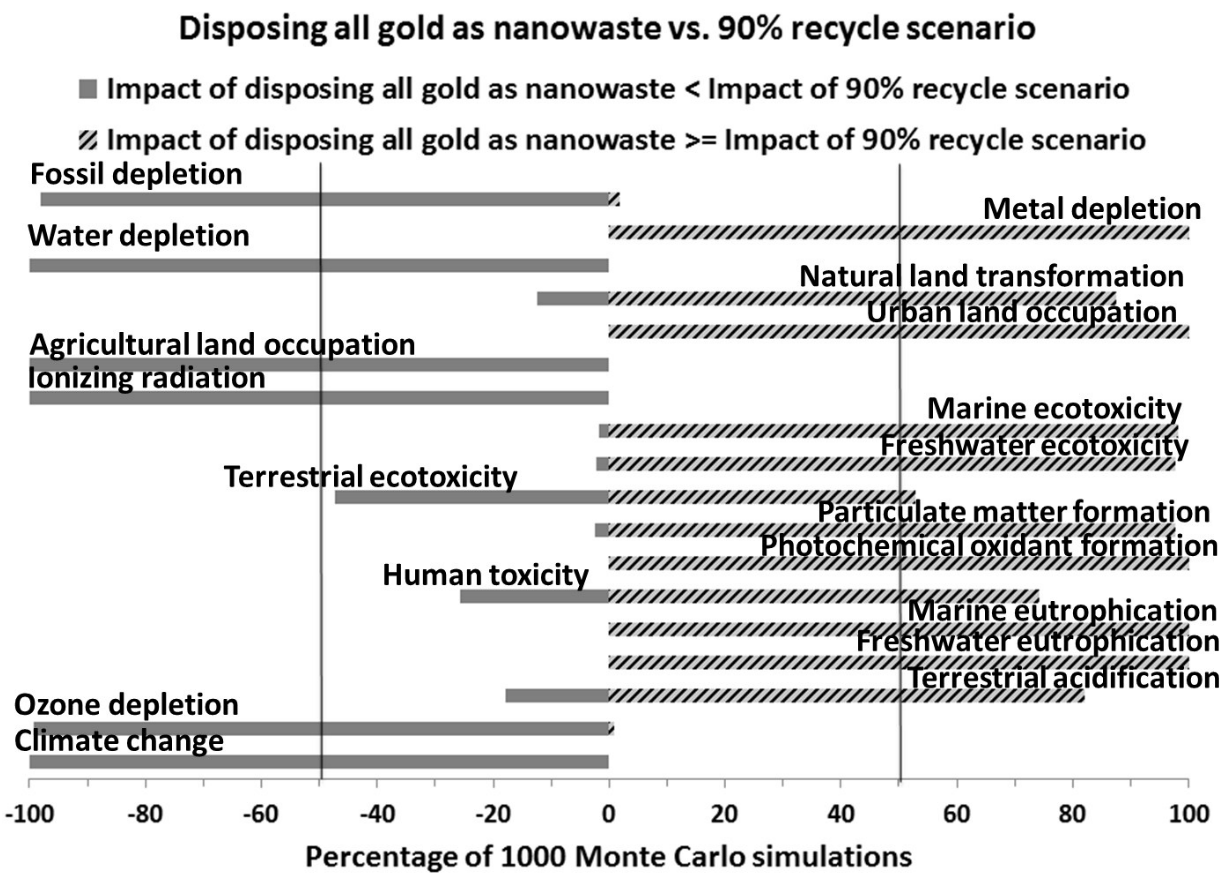


Figure S5 – Uncertainty analysis for 90% recycle scenario vs. no-recycle scenario.

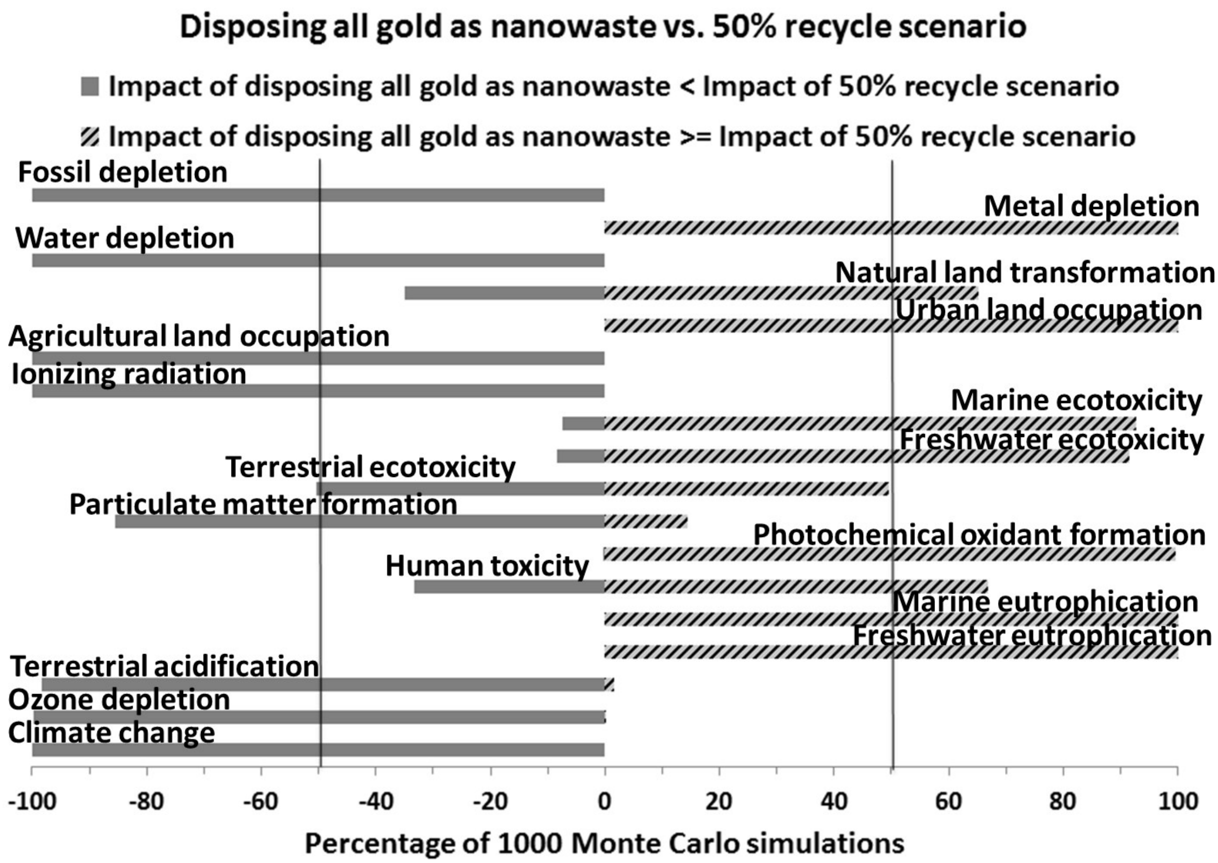


Figure S6 – Uncertainty analysis for 50% recycle scenario vs. no-recycle scenario.

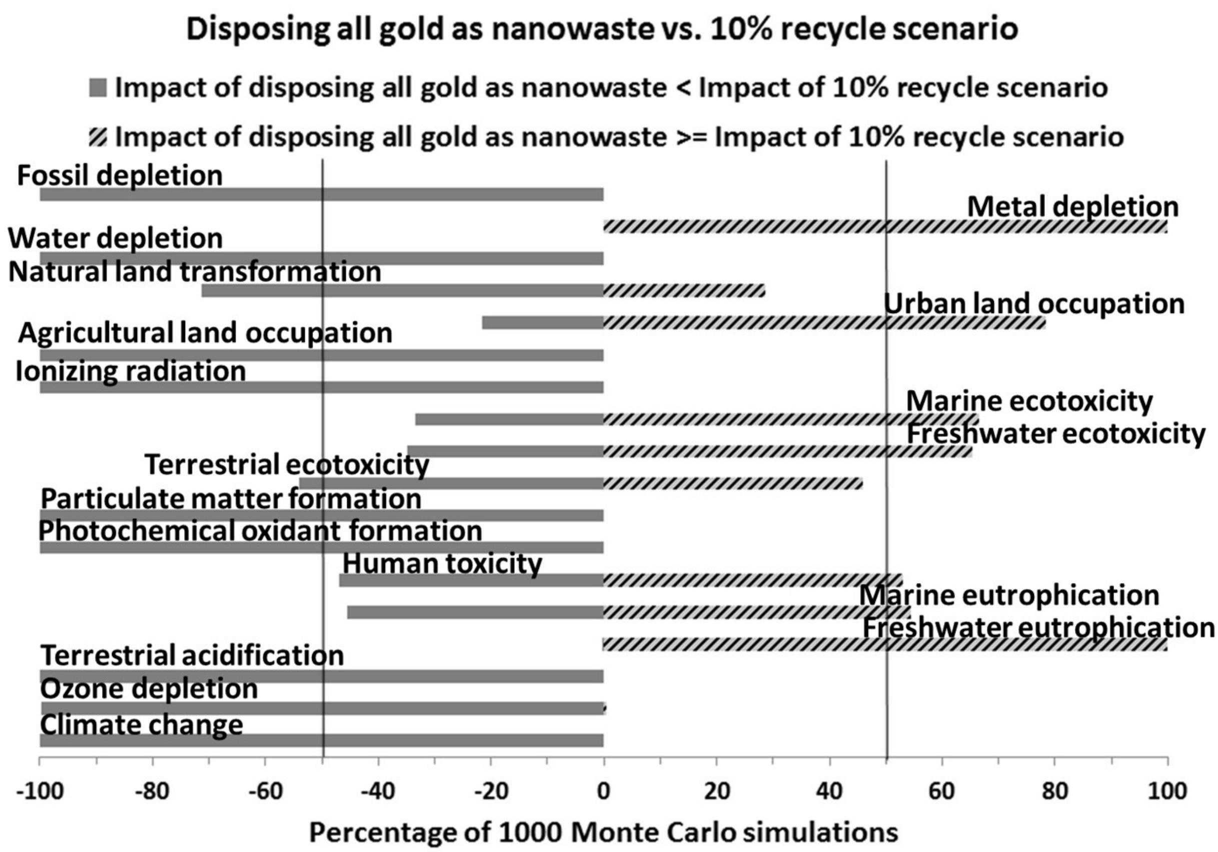


Figure S7 – Uncertainty analysis for 10% recycle scenario vs. no-recycle scenario.

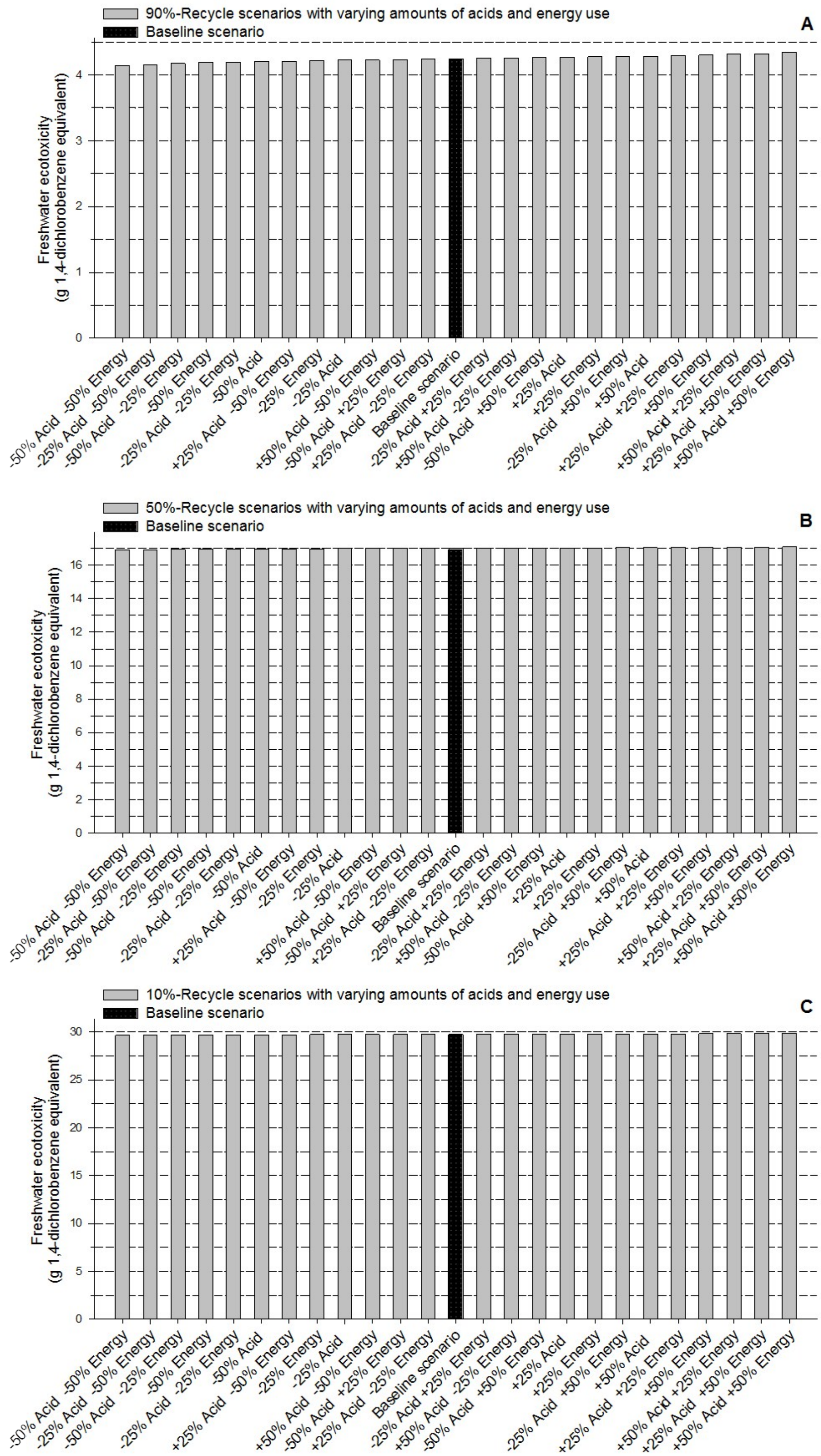


Figure S8 – Sensitivity analysis for freshwater ecotoxicity. The effects of acids (solvents) and energy consumption on freshwater ecotoxicity are modeled for different recycle scenarios. 'Baseline scenario' denotes recycle models where acid use and energy consumption were not varied. For comparison, the metal depletion for no-recycle scenario is 32.5 g dicholorobenzene equivalent, which is higher than all the recycle scenarios modeled.

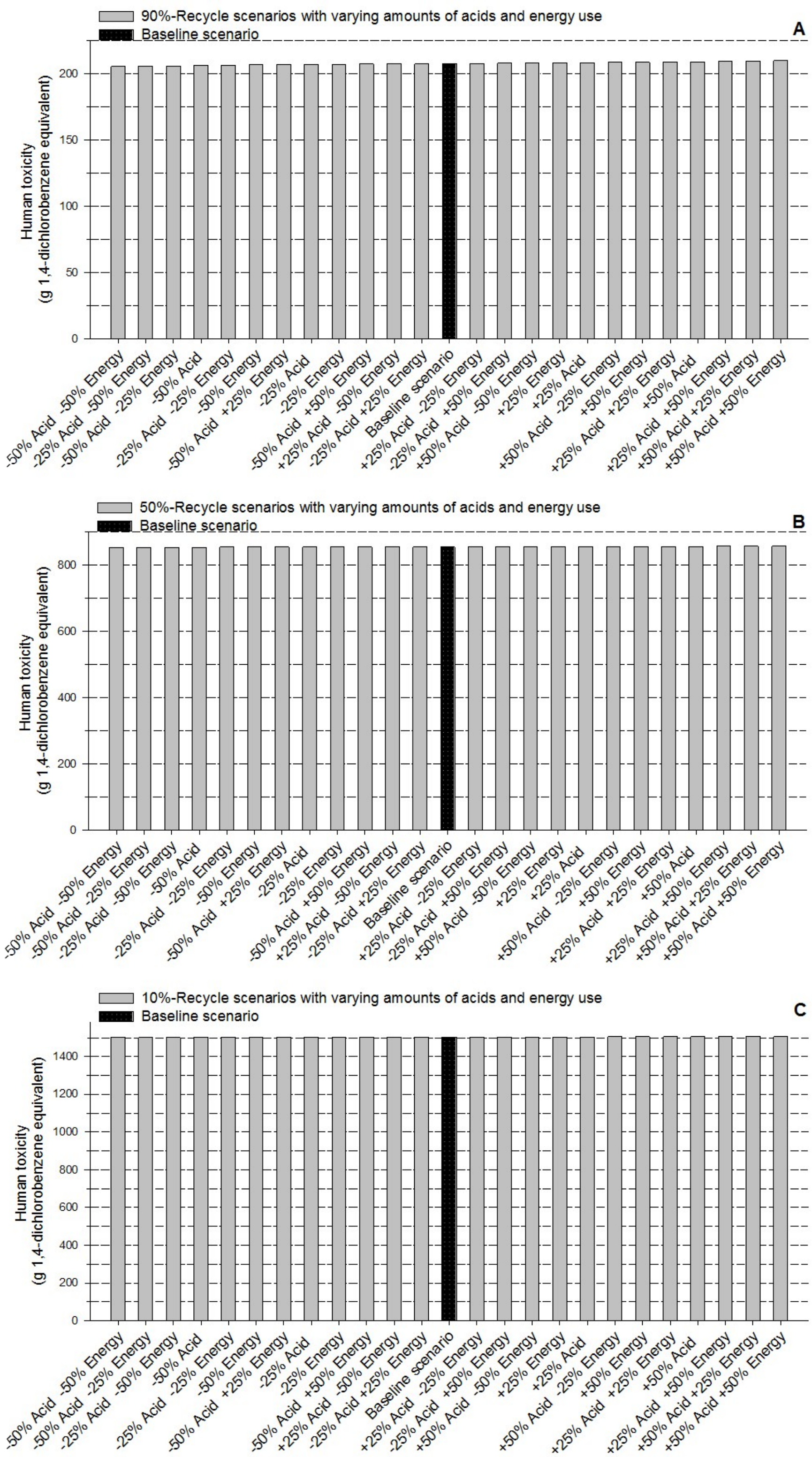


Figure S9 – Sensitivity analysis for human toxicity. The effects of acids (solvents) and energy consumption on human toxicity are modeled for different recycle scenarios. 'Baseline scenario' denotes recycle models where acid use and energy consumption were not varied. For comparison, the metal depletion for no-recycle scenario is 1660 g dicholorobenzene equivalent, which is higher than all the recycle scenarios modeled.