

### Supplementary information

Data “% of area equipped for irrigation by surface water” provided by AQUASTAT in given year were calculated as:

$$[\% \text{ of area equipped for irrigation by surface water}] = 100 * [\text{Area equipped for irrigation by surface water}] / [\text{Area equipped for irrigation: total}]$$

Data “% sprinkler irrigation intensity out of total irrigation” were calculated as:

$$[\% \text{ sprinkler irrigation intensity out of total irrigation}] = 100 * [\text{Area equipped for full control irrigation: sprinkler irrigation}] / [\text{total agricultural water managed area}]$$

These two groups of data in the same period from 2013 to 2017 were collected for EU28. However, few abnormal results were higher than 100%. This is because of the incoherence of data collection time between two parameters in the calculation as shown in Table S. Solutions and corresponding values are also shown in Table S1.

Table S1. Discrepancies and solutions of irrigation related parameters

National data information		reference	solution and reference	value
Poland	% of area equipped for irrigation by surface water			
	total area equipped for irrigation	area equipped for irrigation by surface water		
value (1000ha)	75.81	104.1	Irrigation area sheet-Poland, AQUASTAT	calculate surface water percentage out of the sum of total areas for all the irrigation water sources (surface water and groundwater)
data collection time	2013	2007	2019	102.7/(102.7+11.6)
Latvia	% of area equipped for irrigation by surface water			
	total area equipped for irrigation	area equipped for irrigation by surface water		
value (1000ha)	0.63	0.77	Irrigation area sheet-Latvia, AQUASTAT	use surface water% data in 2007 provided by Irrigation report Latvia-AQUASTAT 2016
data collection time	2013	2007	2019	93%
Estonia	% of area equipped for irrigation by surface water			
	total area equipped for irrigation	area equipped for irrigation by surface water		
value (1000ha)	0.458	1.17	Irrigation area sheet-Estonia, AQUASTAT	use surface water% data in 2005 provided by Irrigation report Estonia-AQUASTAT 2016
data collection time	2010	2005	2019	86%
Estonia	% sprinkler irrigation intensity			
	Area equipped for full control irrigation: sprinkler irrigation	total agricultural water managed area		
value (1000ha)	3.68	0.43	Irrigation area sheet-Estonia, AQUASTAT	no coherent data provided by Irrigation report Estonia-AQUASTAT 2016
data collection time	1995	2010	2019	N/A
Lithuania	% sprinkler irrigation intensity			
	Area equipped for full control irrigation: sprinkler irrigation	total agricultural water managed area		
value (1000ha)	9.247	4.49	Irrigation area sheet-Estonia, AQUASTAT	no coherent data provided by Irrigation report Lithuania - AQUASTAT 2016
data collection time	1995	2010	2019	N/A

Table S2. AgNP concentrations measured in surface water in EU scope (from<sup>1</sup>)

Location	Measured	Distribution	Reference
----------	----------	--------------	-----------

	time		
River, Vltava, Czech Republic Surface water, 1.5 km downstream, Germany	2017	Uniform(100,3200)	2
River Isar, next to WWTP effluent discharge areas, Germany	2013-2014	Uniform(0.9,2.3)	3
River, next to WWTP effluent discharge areas, Germany	2013-2014	Uniform(2.0,8.6)	3
River, Germany	2016	Uniform(2.42,69.18)	4
IJssel, Holland	2016	Uniform(1,2)	4
Meuse, Holland	2018	Uniform(0.3,2.5)	5
Pre-alpine lakes, Germany	2018	Uniform(0.3,6.6)	5
Wastewater influent, Germany	-	Normal(2.35,0.08)	4
	2013-2014	Uniform(10.1,357)	3

Table S3. Country-specific sludge application and irrigation related parameters

reference	6,7	8	9 10		
data form	legislation limits	database provided calculated data	real data	real data	real data calculation
EU28	maximum sludge application rate (t DW/ha)	% of area equipped for irrigation by surface water (%)	total agricultural water managed area (1000 ha)	Area equipped for full control irrigation: sprinkler irrigation (1000 ha)	sprinkler percentage (Ir_sp)
Austria	2.5-10/2 yr	0.22052927	99.76	0	0.00%
Belgium	4/2 yr or 12/3 yr	0.509384776	19.18	0	0.00%
Bulgaria	0		115.5	23.18833652	20.08%
Croatia	0		29.68	1.473024523	4.96%
Cyprus	0	0.580592147	38.943	1.927305027	4.95%
Czechia	0		45.86	13.09265507	28.55%
Denmark	10/yr		299	0	0.00%
Estonia	-	0.86	0.43	3.68	
Finland	digestion (<5%) (assume negligible)	0.638981391	102.1	58.78	57.57%
France	-		2691	2464.416272	91.58%
Germany	5/3yr	0.136457717	676.4	655.8076401	96.96%
Greece	-		1517	0	0.00%
Hungary	0		230	192.6188786	83.75%
Ireland	2/yr	0.8	1.1	0	0.00%
Italy	5 (Collivignarelli et al. 2019; EC, 2011 PART1)	0.631910766	4124	1591	38.58%
Latvia	-	0.93	0.672	0	0.00%
Lithuania	0		4.49	9.247	
Luxembourg	3/yr		0	0	
Malta	0	0.003047619	4.2	0.825688073	19.66%
Netherlands	2-4/yr		522.6	0	0.00%
Poland	-	0.898512686	75.81	3.276145203	4.32%
Portugal	6/yr	0.650237313	547.8	168.6	30.78%
Romania	0		334.7	99.23600265	29.65%
Slovakia	0		99.64	0	0.00%

Slovenia	0		7.604	5.267	69.27%
Spain	-		3681	0	0.00%
Sweden	-	0.880025526	156.7	0	0.00%
United Kingdom	-		207.6	95.60526316	46.05%
<b>Fit distribution</b>	Uniform (0,10)	Triangular (0,0.89851,1)			Triangular (0,0,1)

Table S4. Calculation of crop-specific irrigation needs out of total water needs

Area	Crop	Year	Area harvested (ha) <sup>11</sup>	Harvested irrigated area (1000 ha) <sup>12</sup>	Percentage of irrigation need out of total water needs
Ireland	Wheat	2017	67047	0	0.00
Ireland	Root vegetables	2017	0	0	0.00
Ireland	leafy vegetables	2017	5722	0.5	0.09
Europe	Wheat	2017	61879579	478.95	0.01
Europe	Root vegetables	2017	11604	7.377	0.64
Europe	leafy vegetables	2017	3619763	956.172	0.26

Table S5. Calculations of bioaccumulation factors for crops to AgNPs through root or leafy exposure

symbol	crop (root exposure)	exposed AgNP concentration	unit	Ag concentration in edible parts	unit	reference	BF
	Mustard	108	mg/L (Hoagland solution)	30	ug/ml w.w.	13	0.28
	silverbeet (chard)	324	mg/L (soil colloid)	42			0.13
	spinach	70	mg/kg	7		14	0.10
	rocket	70	mg/kg	3.65		14	0.05
	lettuce	70	mg/kg	0.25	mg/kg d.w.	14	0.00
	lettuce	0.1				15	0.55
		0.5					0.64
		1					0.61
BF_lv_s							Triangular(0,0.0035714,0.64)
	Radish Sprouts	125	mg/L	114	mg/kg d.w.	16	0.91
		250	mg/L	204			0.82
		500	mg/L	900			
BF_rv3_s							Uniform(0.82,1.80)
BF_rv2_s	leek	70	mg/kg	3.65	mg/kg d.w.	14	0.05
	carrot	70	mg/kg	7	mg/kg d.w.	14	0.10
	beetroot	70	mg/kg	3.65		14	0.05
BF_rv1_s							Uniform(0.05,0.1)
BF_w_s	wheat	20	mg/kg	0.02	mg/kg d.w.	17	0.001
symbol	foliar exposure	exposed AgNP concentration	unit	Ag concentration in edible parts	unit	reference	BF

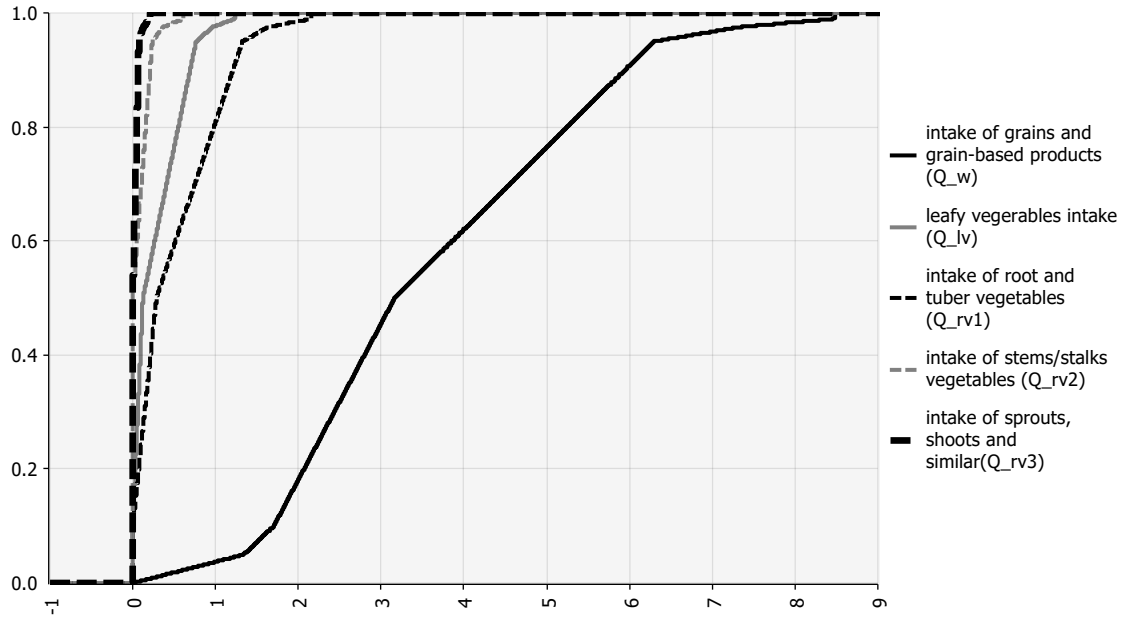
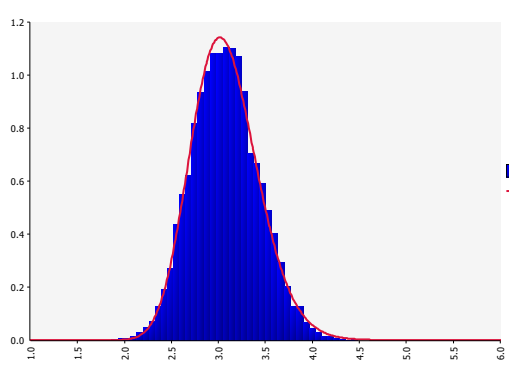
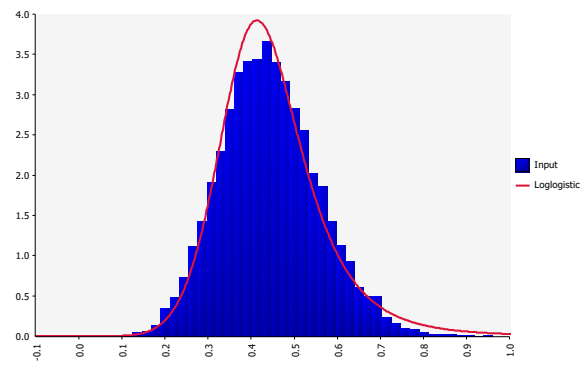


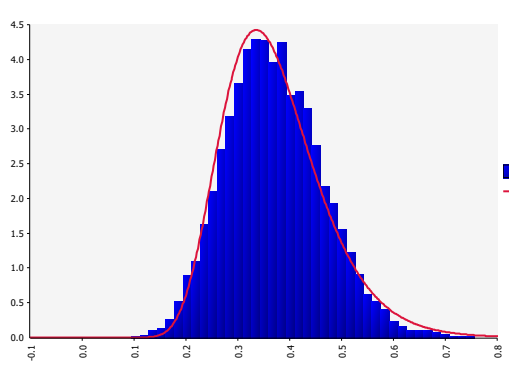
Fig. S1. Cumulative distributions fitted for crops' intake data in Ireland



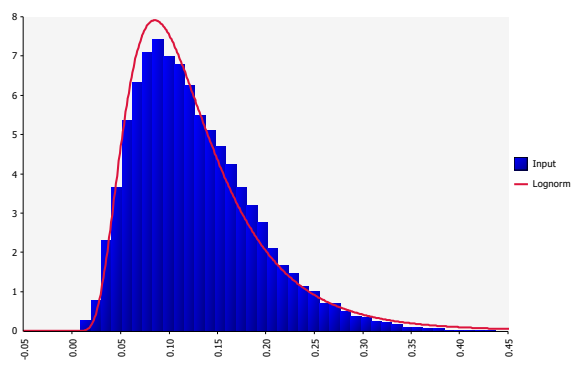
(a)



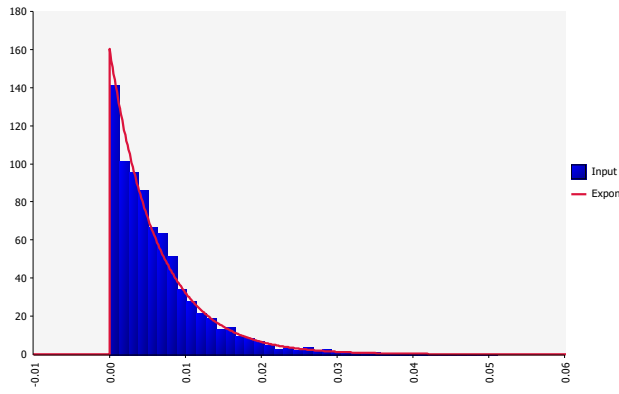
(b)



(c)

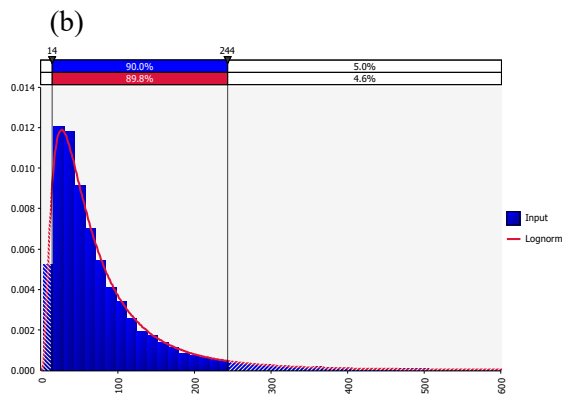
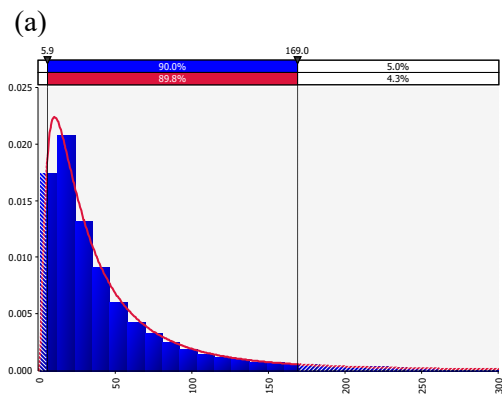
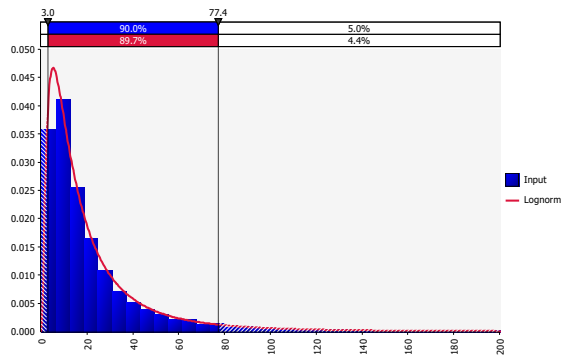
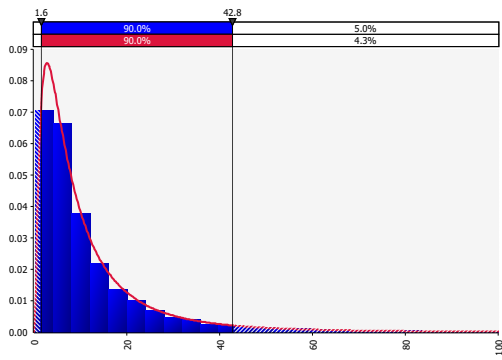


(d)



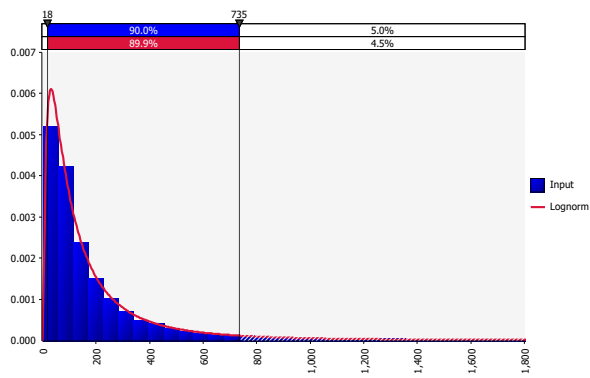
(e)

Fig. S2. Simulations and the best fit distributions of crops' intake data in EU average for (a) wheat and products, (b) leafy vegetables, (c) root and tuber vegetables, (d) stems/stalks vegetables, and (e) sprouts, shoots and similar

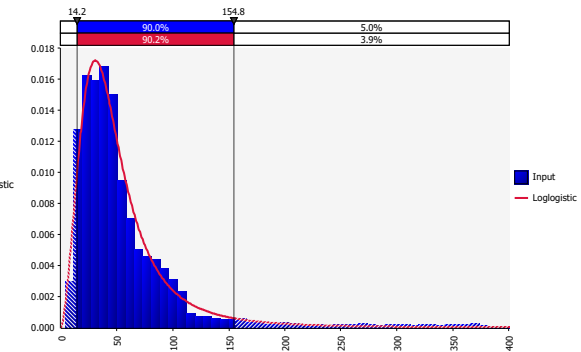
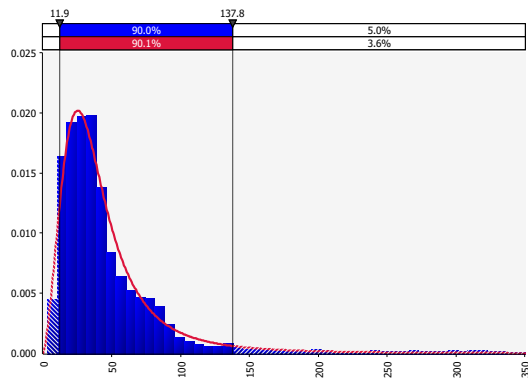


(c)

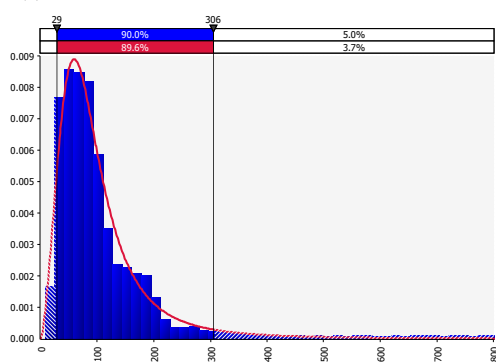
(d)



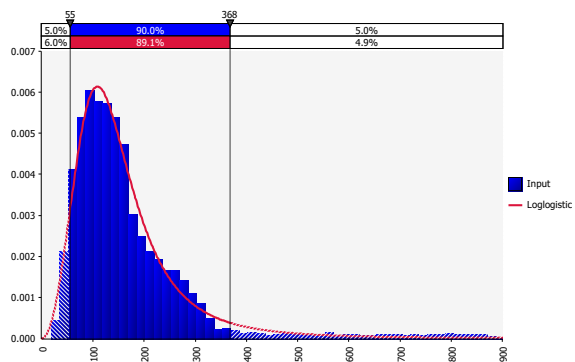
(c)



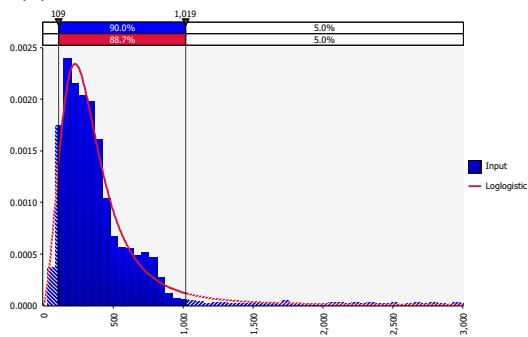
(f)



(g)



(h)



(i)

(j)

Fig. S3. The best-fit distributions for annual AgNP concentrations ( $C_{sl}$ ) in Europe in (a) 2005, (b) 2007, (c) 2010, (d) 2012, (e) 2020; and in Ireland in (f) 2005, (g) 2007, (h) 2010, (i) 2012, (j) 2020

## References

- 1 J. Zhao, M. Lin, Z. Wang, X. Cao and B. Xing, Engineered nanomaterials in the environment: Are they safe?, *Crit. Rev. Environ. Sci. Technol.*, 2020, 1–36.
- 2 M. Loula, A. Kaňa, R. Koplík, J. Hanuš, M. Vosmanská and O. Mestek, Analysis of Silver Nanoparticles Using Single-Particle Inductively Coupled Plasma–Mass Spectrometry (ICP-MS): Parameters Affecting the Quality of Results, *Anal. Lett.*, 2019, **52**, 288–307.
- 3 L. Li, M. Stoiber, A. Wimmer, Z. Xu, C. Lindenblatt, B. Helmreich and M. Schuster, To What Extent Can Full-Scale Wastewater Treatment Plant Effluent Influence the Occurrence of Silver-Based Nanoparticles in Surface Waters?, *Environ. Sci. Technol.*, 2016, **50**, 6327–6333.
- 4 A. Wimmer, A. A. Markus and M. Schuster, Silver Nanoparticle Levels in River Water: Real Environmental Measurements and Modeling Approaches - A Comparative Study, *Environ. Sci. Technol. Lett.*, 2019, **6**, 353–358.
- 5 R. J. B. Peters, G. van Bommel, N. B. L. Milani, G. C. T. den Hertog, A. K. Undas, M. van der Lee and H. Bouwmeester, Detection of nanoparticles in Dutch surface waters, *Sci. Total Environ.*, 2018, **621**, 210–218.
- 6 European Commission, Disposal and Recycling Routes for Sewage Sludge Part 1—Sludge Use Acceptance Report, 2001.
- 7 M. C. Collivignarelli, A. Abbà, A. Frattarola, M. C. Miino, S. Padovani, I. Katsoyiannis and V. Torretta, Legislation for the Reuse of Biosolids on Agricultural Land in Europe: Overview, *Sustainability*, 2019, **11**, 1–22.
- 8 FAO, % of area equipped for irrigation by surface water- AQUASTAT database, % of area equipped for irrigation by surface water- AQUASTAT database, <http://www.fao.org/aquastat/statistics/query/index.html?lang=en>, (accessed 12 March 2021).
- 9 FAO, total agricultural water managed area (1000ha)- AQUASTAT database, total agricultural water managed area (1000ha)- AQUASTAT database, <http://www.fao.org/aquastat/statistics/query/index.html?lang=en>, (accessed 12 March 2021).
- 10 FAO, Area equipped for full control irrigation: sprinkler irrigation (1000 ha)- AQUASTAT database, Area equipped for full control irrigation: sprinkler irrigation (1000 ha)- AQUASTAT database, <http://www.fao.org/aquastat/statistics/query/index.html?lang=en>, (accessed 1 March 2021).
- 11 FAO, FAOSTAT\_Crop statistics, FAOSTAT\_Crop statistics, <http://www.fao.org/faostat/en/#data/QC>, (accessed 8 June 2021).
- 12 FAO, Harvested irrigated area (1000 ha)- AQUASTAT database, Harvested irrigated area (1000 ha)- AQUASTAT database, <http://www.fao.org/aquastat/statistics/query/index.html?lang=en>, (accessed 16 March 2021).
- 13 K. Vishwakarma, Shweta, N. Upadhyay, J. Singh, S. Liu, V. P. Singh, S. M. Prasad, D. K. Chauhan, D. K. Tripathi and S. Sharma, Differential phytotoxic impact of plant mediated silver nanoparticles (AgNPs) and silver nitrate (AgNO<sub>3</sub>) on Brassica sp., *Front. Plant Sci.*, , DOI:10.3389/fpls.2017.01501.
- 14 N. Saleeb, R. Gooneratne, J. Cavanagh, C. Bunt, A. K. M. M. Hossain, S. Gaw and B. Robinson, The Mobility of Silver Nanoparticles and Silver Ions in the Soil-Plant System, *J. Environ. Qual.*, 2019, **48**, 1835–1841.
- 15 J. Wu, G. Wang, M. G. Vijver, T. Bosker and W. J. G. M. Peijnenburg, Foliar versus root exposure of AgNPs to lettuce: Phytotoxicity, antioxidant responses and internal translocation, *Environ. Pollut.*, 2020, **261**, 114117.

- 16 N. Zuverza-Mena, R. Armendariz, J. R. Peralta-Videa and J. L. Gardea-Torresdey, Effects of Silver Nanoparticles on Radish Sprouts: Root Growth Reduction and Modifications in the Nutritional Value, *Front. Plant Sci.*, 2016, **7**, 90.
- 17 J. Yang, F. Jiang, C. Ma, Y. Rui, M. Rui, M. Adeel, W. Cao and B. Xing, Alteration of Crop Yield and Quality of Wheat upon Exposure to Silver Nanoparticles in a Life Cycle Study, *J. Agric. Food Chem.*, 2018, **66**, 2589–2597.
- 18 M. Li, H. L. Liu, F. Dang, H. Hintelmann, B. Yin and D. Zhou, Alteration of Crop Yield and Quality of Three Vegetables upon Exposure to Silver Nanoparticles in Sludge-Amended Soil, *ACS Sustain. Chem. Eng.*, 2020, **8**, 2472–2480.