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Probabilistic modelling of engineered nanomaterial emissions to the environment: A spatio-temporal approach

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Supporting Information

System parameters

A brief introduction of SA

South Australia is a state located in southern central Australia with a land area of 975,612 km^2 [1] and is divided into 12 distinctive government identified regions, encompassing 4 metropolitan, 3 greater metropolitan and 5 country regions [2]. Each region comprises a set of local councils and is spatially zoned to assist in planning and management according to a range of demographic, economic and geographic features. Approximately three quarters of the population live in metropolitan and greater Adelaide, the state capital. Metropolitan Adelaide is home to a number of important manufacturing industries such as defense and provides the gateway to a number of regional tourist attractions and economies. Greater Adelaide extends from McLaren Vale and the Fleurieu Peninsula in the south around the Adelaide Hills to the Barossa in the north, providing world renowned vineyards. The Murray-Mallee and Limestone Coast regions of the south and south-east provide temperate geographies amenable to agriculture and orchards, whereas the temperate and semi-arid landscape of the Yorke and Mid-North produces cereals and wool. In contrast to crop-centred economies, the Far North and Eyre and Western regions are characterised by a greater dependence on mining and aquaculture, respectively [2]. Figure S1 shows the administrative geographical regions and the locations of the regions studied.

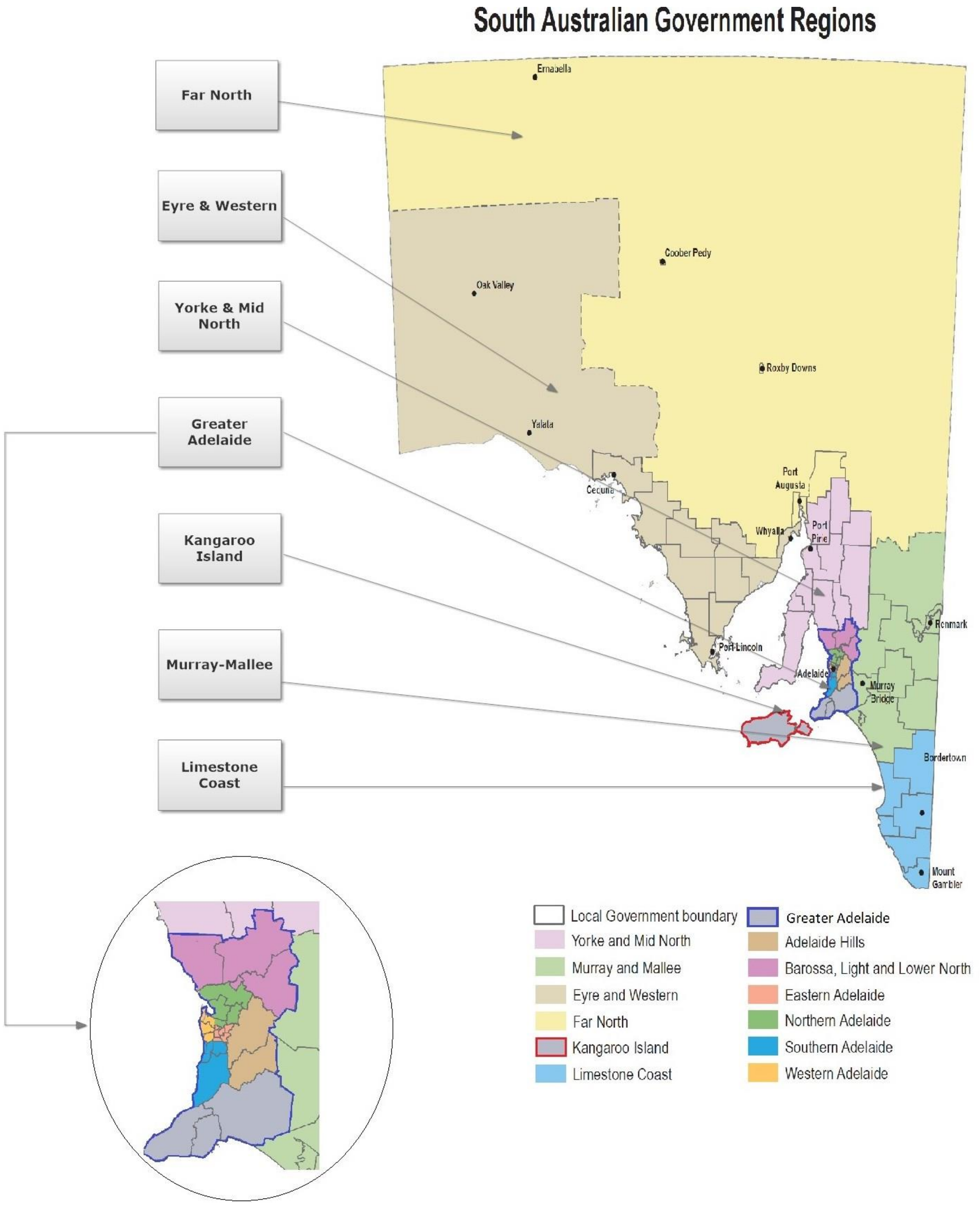


Figure S1.The administrative geographical regions and the locations of the regions studied in South Asutralia. Map was taken from website of South Australian Government[1].

Definition of the seven regions:

The 12 South Australian government regions provided the basis by which to define the flow and fate of engineered nanomaterials throughout the state. This approach was significant because it enabled the use of existing demographic and geographic data when defining each region. Metropolitan and greater Adelaide regions were coalesced into a single region with the exception of Kangaroo Island. Kangaroo Island was defined as a separate region in recognition of differences in the use and recycling of biosolids and treated wastewater on the island in contrast to the mainland. The 5 remaining regions corresponded to the country South Australian government regions resulting in a total of 7 regions(see figure S1), each with a distinctive population[3] and size[1]. The flow and fate of engineered nanomaterials for South Australia were then characterized by studying (i) the flow of materials out of the state for recycling, (ii) the fate of materials such as wastewater and biosolids within regions, and (iii) the flow and fate of biosolids between regions.

The areas of the agricultural soils receiving biosolids covered by each region were defined by the the areas of soil in each region aiming for crop farming. This is given in PIRSA report[4].

Table S1: Summary of volumes of different technical and environmental compartments used for South Australia

Compartments			Formula	Mass/Volumes	Unit	Comments
Agricultural soil	Far North		4278*10^{6}*0.10*1333	5.70E+11	kg	10^{6} is the conversion factor from km^2 to m^2 0.10 m is the depth of agricultural soil considered to be affected by pollutant. [5] 1333 kg/m^3 is the density of agricultural soil (same as above) [5] 4278 km^2, 10781 km^2, 14087 km^2, 214 km^2 , 182 km^2, 7101 km^2, 3214 km^2 are the area of agricultural soil in region of Far North, Yorke & Mid North, Eyre & Western, Greater Adelaide, Kangaroo Island, Murray-Mallee and Limestone Coast[6]
	Yorke & Mid North		10781*10^{6}*0.10*1333	1.44E+12	kg	
	Eyre & Western		14087*10^{6}*0.10*1333	1.88E+12	kg	
	Greater Adelaide		214*10^{6}*0.10*1333	2.85E+10	kg	
	Kangaroo Island		182*10^{6}*0.10*1333	2.43E+10	kg	
	Murray-Mallee		7101*10^{6}*0.10*1333	9.47E+11	kg	
	Limestone Coast		3214*10^{6}*0.10*1333	4.28E+11	kg	
Ocean gulfs	St. Vincent		75000*50000*30*1000	1.13E+14	litre	75000 m is the length of the gulf[7] ; 1000 is conversion factor from m^3 to litres 50000 m is the width of the gulf[7] 30 m is the depth of the gulf suggested [8]
	Spencer		200000*40000*21*1000	1.68E+14	litre	200000 m is the length of the gulf[7] ; 40000 m is the width of the gulf[7] 30 m is the depth of the gulf suggested. [8]
Ocean sediments	St. Vincent	Bolivar	1761.3*10^{6}*0.03*1300*(1-0.8)	1.37E+10	kg	1761.3 km^2 is the area of sediments that can be affected by WWTPs of Bolivar, Glenelg and Christies beach. 628.3 km^2 is the area of sediments of Whyalla and Port Pirie 104.7 km^2 is the area of sediments of Port Augusta 10^{6} is the conversion factor from km^2 to m^2. 0.03 m is the depth of sediments for calculation suggested by ECHA[9] 1300 kg/m^3 is the density of wet sediments[9] 0.8 is the porosity of sediments[9]
		Glenelg				
		Christies				
	Spencer	Whyalla	628.3*10^{6}*0.03*1300*(1-0.8)	4.90E+09	kg	
		Pirie	628.3*10^{6}*0.03*1300*(1-0.8)	4.90E+09		
		Augusta	104.7*10^{6}*0.03*1300*(1-0.8)	8.17E+08		

WWTP biosolids stockpiles and transfer

Regional biosolids production was determined on a per capita basis using SA Water figures for the average total biosolids produced in metropolitan Adelaide over the study period using SA Water Activity Reports for 2005–2012[3, 10]. It was assumed that the per person rate of biosolids production would be stable between metropolitan and regional WWTPs.[11]. Due to the complexity of the system we also assumed that regionally produced biosolids would be applied to soils in the year of production, but we acknowledge that this may not be the case. Quantities transported between regions were determined by identifying and charting post-2005 biosolids movements as part of the SA Water Biosolids Re-use Scheme[10]. This approach provided the rationale for calculating the total quantities of biosolids applied to agricultural soils for soil amendment within each region.

Table S2: Temporal production and distribution of WWTP biosolids between 2005 and 2012 from SA water (unit: tons)

Year	2005	2006	2007	2008	2009	2010	2011	2012	Total
Far North and Kangaroo Island biosolids produced and applied									
Far North	633	688	697	706	712	717	721	723	5597
Kangaroo Island	116	116	117	118	118	119	120	120	944
Eyre & Western									
	Biosolids Applied								**Total**
	2005	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**	
Biosolids Produced									
2005	1,534	55	592	563	26	0	0	0	2,770
2006	0	1,606	0	0	14	14	0	0	1,634
2007	0	0	1,615	0	158	297	139	0	2,209
2008	0	0	0	1,581	25	0	77	77	1,760
2009	0	0	0	0	1,590	27	55	55	1,727
2010	0	0	0	0	0	1,572	30	30	1,632
2011	0	0	0	0	0	0	1,869	293	2,162
2012	0	0	0	0	0	0	0	1,587	1,587
Total	1,534	1,661	2,207	2,144	1,813	1,910	2,170	2,042	15,481
Yorke & Mid North									
	Biosolids Applied								**Total**
	2005	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**	
Biosolids Produced									
2005	3,783	2,375	5,401	15,358	10,682	210	0	0	37,809
2006	0	2,165	0	8,736	9,452	1,602	886	0	22,841
2007	0	0	2,716	522	11,983	19,291	7,693	385	42,590
2008	0	0	0	6,030	4,444	363	1,418	1,418	13,673
2009	0	0	0	0	4,999	3,041	6,749	6,749	21,538
2010	0	0	0	0	0	5,150	2,663	13	7,826
2011	0	0	0	0	0	0	7,149	4,629	11,778
2012	0	0	0	0	0	0	0	1,975	1,975
Total	3,783	4,540	8,117	30,646	41,560	29,657	26,558	15,169	160,030

Greater Adelaide

	Biosolids Applied								Total
	2005	2006	2007	2008	2009	2010	2011	2012	
Biosolids Produced									
2005	4,396	237	577	1,176	796	56	0	0	7,238
2006	0	4,397	0	2,859	2,979	120	0	0	10,355
2007	0	0	4,805	51	2,639	2,809	237	67	10,608
2008	0	0	0	5,882	1,080	1,228	1,437	209	9,836
2009	0	0	0	0	4,891	289	1,304	1,304	7,788
2010	0	0	0	0	0	5,119	301	0	5,420
2011	0	0	0	0	0	0	6,526	0	6,526
2012	0	0	0	0	0	0	0	4,781	4,781
Total	4,396	4,634	5,382	9,968	12,385	9,621	9,805	6,361	62,552

Murray-Mallee

	Biosolids Applied								Total
	2005	2006	2007	2008	2009	2010	2011	2012	
Biosolids Produced									
2005	1,952	429	627	764	497	79	0	0	4,348
2006	0	1,823	0	780	847	67	0	0	3,517
2007	0	0	2,306	87	1,190	1,310	206	86	5,185
2008	0	0	0	2,901	832	1,348	1,194	104	6,379
2009	0	0	0	0	2,067	247	0	0	2,314
2010	0	0	0	0	0	2,368	1,379	924	4,671
2011	0	0	0	0	0	0	2,521	608	3,129
2012	0	0	0	0	0	0	0	1,824	1,824
Total	1,952	2,252	2,933	4,532	5,433	5,419	5,300	3,546	31,367

Limestone Coast

	Biosolids Applied								Total
	2005	2006	2007	2008	2009	2010	2011	2012	
Biosolids Produced									
2005	1,679	131	252	245	124	0	0	0	2,431
2006	0	2,617	0	262	316	54	0	0	3,249
2007	0	0	5,090	2,453	196	196	0	0	7,935
2008	0	0	0	1,705	138	308	324	15	2,490
2009	0	0	0	0	1,778	79	47	47	1,951
2010	0	0	0	0	0	1,784	79	0	1,863
2011	0	0	0	0	0	0	1,701	0	1,701
2012	0	0	0	0	0	0	0	1,706	1,706
Total	1,679	2,748	5,342	4,665	2,552	2,421	2,151	1,768	23,326

Table S2 shows the temporal year of production and distribution of biosolids into seven defined regional farms between 2005 and 2012. Due to the mechanism of biosolids stockpile and biosolids transport between regions, biosolids produced in one region were not applied in the same regional farms in which they were produced; similarly biosolids were not applied in their totality to the soils in the same year when they were produced. The matrix of table S2 shows in rows how much of each year biosolid is distributed over time; in column it shows one year's application of biosolid into soils consisted of different years product input. In the farms of Far North and Kangaroo Island there were no stockpiled biosolids applied; only “fresh” biobolids, which means biosolids were applied in the same year of production. Determination and differentiation of the biosolids produced and applied in different years is important; because ENM concentrations in biosolids produced in different year vary considerably. This is crucial for the calculation of annual as well as total ENM load into a regional farm.

Distribution of treated waste water

Regional ocean wastewater discharge figures for South Australia were determined on a per capita basis using wastewater produced in metropolitan Adelaide. The average rate of wastewater produced in metropolitan Adelaide was calculated by integrating population and wastewater production figures[12, 13]. Regional wastewater production was correlated to metropolitan wastewater production on the basis of population. This approach provided the rationale for calculating regional discharge by multiplying the population serviced by WWTP in respective regions by the quantity of wastewater not recycled[14, 15]. In table S3 the summary of wastewater production and distribution in SA over the period 2005-2012 is given.

Table S3: Summary of wastewater production and distribution in SA over the period 2005-2012

Year/units/sources				2005	2006	2007	2008	2009	2010	2011	2012	Units	References/Comments
Waste water	Produced	WWTP	Gt. Adelaide	104,916	103,475	95,549	91,266	91,520	92,706	98,676	96,801	10^3*m^3	SA Water report [13]
			Regions	10,890	11,002	11,090	11,080	11,237	11,312	11,342	11,412	10^3*m^3	Extrapolation based on SA Water information [13]
			Sum	115,806	114,477	106,639	102,346	102,757	104,018	110,018	108,213		
		CWMS		16,712	16,973	17,141	17,359	17,486	17,640	17,721	17,858		
		Total		**132,518**	**131,450**	**123,780**	**119,705**	**120,243**	**121,658**	**127,739**	**126,071**		
	Recycled			27,084	29,934	34,061	35,769	37,131	35,368	28,392	24,041	10^3*m^3	SA Water [13]
	Discharge to Ocean	St.Vincent	Bolivar	49,100	49,978	40,704	37,948	37,450	39,215	45,884	42,816	10^3*m^3	[14, 15]
			Glenelg	19,476	19,825	16,146	15,053	14,855	15,555	18,201	16,984		
			Christies	13,257	13,494	10,990	10,246	10,112	10,588	12,389	11,560		
		Spencer	Whyalla	976	980	976	990	994	1,000	1,003	1,010		
			Pirie	1,114	1,118	1,125	1,129	1,135	1,138	1,139	1,145		
			Augusta	583	634	641	649	655	660	661	666		
		Sum of discharge		84,506	84,506	84,506	84,506	84,506	84,506	84,506	84,506		

Table S4: Summary of the application rate of treated waste water in grass, gardens and pastures (GGP), viticulture (VC), horticulture (HC) from 2005 to2012

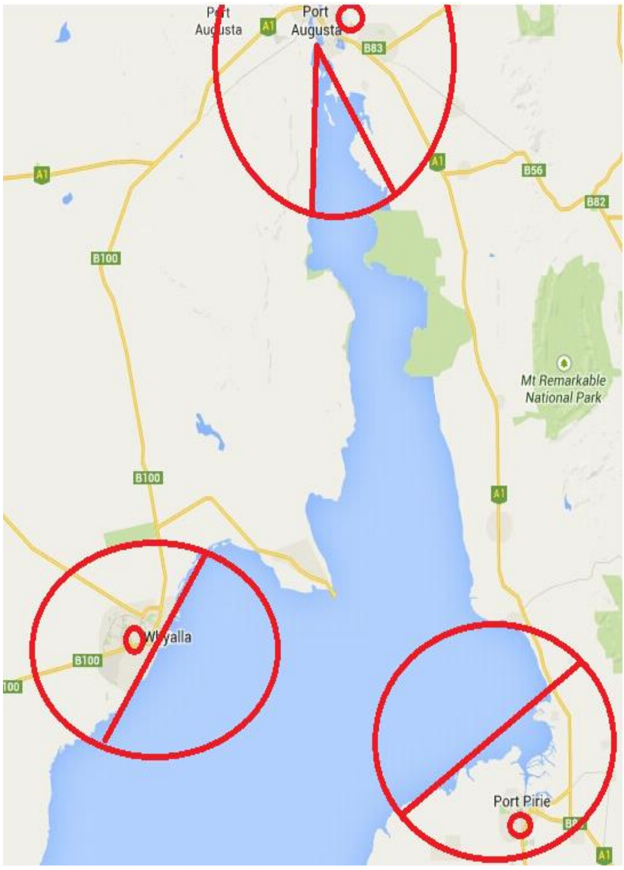
Soil types	Application rate	Source
Grass, gardens and pastures (GGP)	**4.48** million litres per hectare per year	Laurenson, S., et al. 2010[16]
Viticulture (VC)	**2.10** million litres per hectare per year	
Horticulture (HC)	**3-5** million litres per hectare per year	

Gulf Sedimentation Area Estimates & WWTP Discharges for 2005 – 2012

Two separate approaches were taken to calculating the likely ENM exposure areas in the Spencer Gulf and the Gulf of St Vincent due to differences in the availability of suitable data.

Spencer Gulf

The strategy used for WWTPs in the Upper Spencer Gulf were based on Kämpf et al's[17] observation that slow flushing times in this region indicated that the accumulation of materials at 0.3% concentration in the far field would reach 20km. As such, it was reasoned an area of sediment exposed to ENM from discharge could be estimated based on a 20km radius around each of the three WWTPs discharging into the Upper Spencer Gulf. Figure S2.a shows how the geography of the region influenced the calculation of sediment areas. Since the coast surrounding both Whyalla and Port Pirie WWTPs is relatively open (ie. 180^{o}), it was assumed that half the area of a circle with a radius of 20 km would produce a fair estimate. In contrast, the coast adjacent to Port Augusta provides a relatively narrow body of water in which wastewaters are discharged. It was assumed that this would equate to approximately 30^{o}.



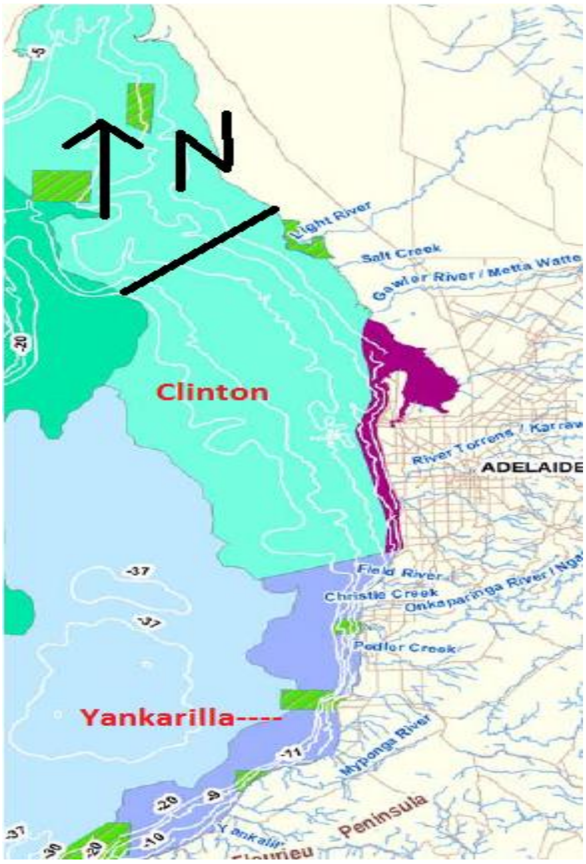


Figure S2: a. Upper Spencer Gulf ***b. Gulf of St Vincent***

Whyalla

It was assumed that approximately half of a hypothetical circle of 20km radius would cover ocean sediment.

The area of sediment affected by discharge from Whyalla WWTP was calculated as:

Area = $(1/2)\ \pi r^2 = 0.5\pi \times (20)^2 = 628.3\ km^2$

Port Pirie

It was assumed that approximately half of a hypothetical circle of 20km radius would cover ocean sediment.

The area of sediment affected by discharge from Port Pirie WWTP was calculated as:

Area = $(1/2)\ \pi r^2 = 0.5\pi \times (20)^2 = 628.3\ km^2$

Port Augusta

It was assumed that approximately one eight of a hypothetical circle of 20km radius would cover ocean sediment.

The area of sediment affected by discharge from Port Augusta WWTP was calculated as:

Area = $(1/12)\ \pi r^2 = 0.083\pi \times (20)^2 = 104.7\ km^2$

Discharge

Table S3 provides a summary of the volumes of wastewater discharge by each of the WWTPs into the receiving environment.

Table S5: Upper Spencer Gulf Wastewater Discharge (ML) for 2005 – 2012

Year	2005	2006	2007	2008	2009	2010	2011	2012
Whyalla 628.3 km^2	976	980	976	990	994	1,000	1,003	1,010
Port Pirie 628.3 km^2	1,114	1,118	1,125	1,129	1,135	1,138	1,139	1,145
Port Augusta 104.7 km^2	583	634	641	649	655	660	661	666

Gulf of St. Vincent Strategy

This was estimated using data contained in a South Australian EPA report which detailed the state of the marine/coastal zone environment of the Gulf of St Vincent according to the concept of a bioregion. A bioregion is a part of the environment identified by conservation ecologists for its distinctive and unique properties with respect to flora and fauna etc. The EPA report listed a range of activities that threated each of the bioregions of the Gulf of St Vincent which listed the Clinton (2,491.4 km^2) and Yankalilla (515.6 km^2) (Figure S2-b) bioregions as those under significant threat from WWTP and Community Wastewater Management Systems (CWMS, formerly called Septic Tank Effluent Drainage Scheme or STED; http://www.lga.sa.gov.au/page.aspx?u=253) contamination[18]. Since the constituents of wastewater are widely acknowledged as a direct threat to the health of Gulf St Vincent seagrass [19, 20], it was reasoned that the relationship between seagrass die back and wastewater could provide the basis for estimating sediment areas from bioregion data. Given that the prevailing currents of the Gulf of St Vincent move in a clockwise direction [1] and die back of seagrass north of Bolivar had not extended more than 6 km between 1949 and 1993 [20, 21], it was assumed that WWTP/CWMS threats further north in the Clinton bioregion could be attributed to CWMS alone. Accordingly, it was assumed that approximately 50% of the Clinton bioregion area was likely to be affected by WWTP wastewater. Therefore, in combination, the total area of sediment in the Gulf of St Vincent that is likely to be exposed to ENM from discharged wastewaters is 1,761.3 km^2. In addition, Wear and Tanner[22] describe distance gradients from the point source for pH and dissolved nutrients. While they do not characterise the gradient, this information could be used to consider an ENM gradient. These authors state: “From the present study, it appears that water quality improves out to a distance of 2 km after which it stabilizes. However, the spatial resolution of the data is insufficient to determine if there is a gradual change or abrupt transition” and then go on to say “Lara et al (1985) demonstrated that water quality rapidly declined within 900 m of a sewage outfall, reaching normal values at approximately 1700 m”.

Metropolitan WWTP

Bolivar is the largest of the three metropolitan Adelaide WWTPs, serving 60% of the population[23], which equated to 705,016 people in 2012. Christies Beach treats 45 ML/day[24] and using the per person rate of treatment (0.08631 ML/year) calculated by dividing the metropolitan total wastewater produced[13] by the average metropolitan population [3] over the study. It was inferred that this equated to a population of 190,433. In the absence of specific data for Glenelg, population and wastewater figures were estimated by subtracting Bolivar and Christies Beach WWTP populations from the Adelaide total population of 1,175,026 for 2012. Based on the information and specific wastewater recycling knowledge of the treated wastewater recycling rates for metropolitan Adelaide over the study period, the volumes of wastewater discharged was calculated, as shown in Table S6.

Table S6: Upper Spencer gulf wastewater discharge (ML) for 2005 – 2012

Year	2005	2006	2007	2008	2009	2010	2011	2012
Bolivar 1,761.3km^2	45,331	43,768	42,210	43,833	44,427	43,557	43,563	43,850
Glenelg 1,761.3km^2	22,432	22647	22,924	23,213	21,551	21,188	20,798	20,540
Christies Beach 1,761.3km^2	11,980	11,654	11,483	11,262	11,075	10,863	10,593	10,773

ENM parameters

Production volume:

Global and regional production and consumption data of ENM have been reported by quite a few studies.[25-29] However, to the best knowledge of the authors of this study, no specific report of ENM consumption especially for South Australia (SA) is available. Keller et al. [28] is the only study that reported ENM emission in Australia. Yet they did not report any explicit figures of how much ENM is consumed in Australia, but only ENM emissions to technical and environmental compartments. By summing up the total emission amount, we were able to estimate an approximate consumption volume for four ENM that is of interest to this study suggested by them. We followed a top-down approach to estimate the ENM consumption in SA on the basis of the known EU ENM production. This was done by scaling the known ENM consumption data in European Union(EU)[25] down to the number of SA in proportional to the ratio of their Gross Domestic Product(GDP), which is an index that we think is very correlated to its power of purchase and consumption of products. According to World Bank statistics in 2012[30], EU has a GDP of 16.67 trillion the U.S. dollar, the number for Australia is 1.532 trillion[30], and for SA is 86 billion.[31] Sun et al.[25] reported the EU nano-TiO_2, nano-ZnO, nano-Ag, CNT and fullerenes production (also taken as consumption) for 10,200 tons, 1,580 tons, 32 tons, 380 tons and 22 tons respectively. These values are the most probable values from the probability distribution. Combining the information of GDP in EU, Australia and South Australia and the predicted ENM consumption data in EU, we calculated the ENM consumption of Australia and SA. Table S7 shows the results of a comparison between our estimation and the estimation of Keller et al. in tons.

Table S7: The comparison of ENM consumption estimation in Australia and SA between Keller et al.2013 and this study, unit:ton.

ENM	Australia and Oceania (Keller et al. 2013)	Australia (2012)	South Australia (2012)
nano-TiO_2	867	961	54
nano-ZnO	334	178	10
nano-Ag	4	3.2	0.18
CNT	31	37	2.1

Because there is no data of fullerenes from Keller's study, we just compared the estimation of nano-TiO_2, nano-ZnO, nano-Ag and CNT. From table S6 we can see out of the four ENM compared, except nano-Zn, the estimated consumption from the two studies for all the other three ENM matches very well. For the case of nano-ZnO, Keller et al. has a higher estimation of 334 tons which is around twice of the estimation of this study. While this discrepancy between the two figures is just relatively bigger than the others, they still fall within the same range. The estimation of Australian data was not necessary for our modelling, but together with the comparison it was intended for cross validation. The result implies estimations of both methods are in very good agreement; this increases the reliability of the approach we used.

The extrapolation of the ENM consumption for the years before 2012 were made by proportionating the value of 2012 to other years according to ENM development trend analysis[26] and nanotechnology market projections [32, 33] together with a direct production trend estimation for the case of CNT by Piccinno et al.[26] The final development tendency estimation of ENM between 2005 and 2012 is an information combination of all these studies. Modelled estimations of the five ENM consumption developments between 2005 and 2012 are shown in Figure S3.

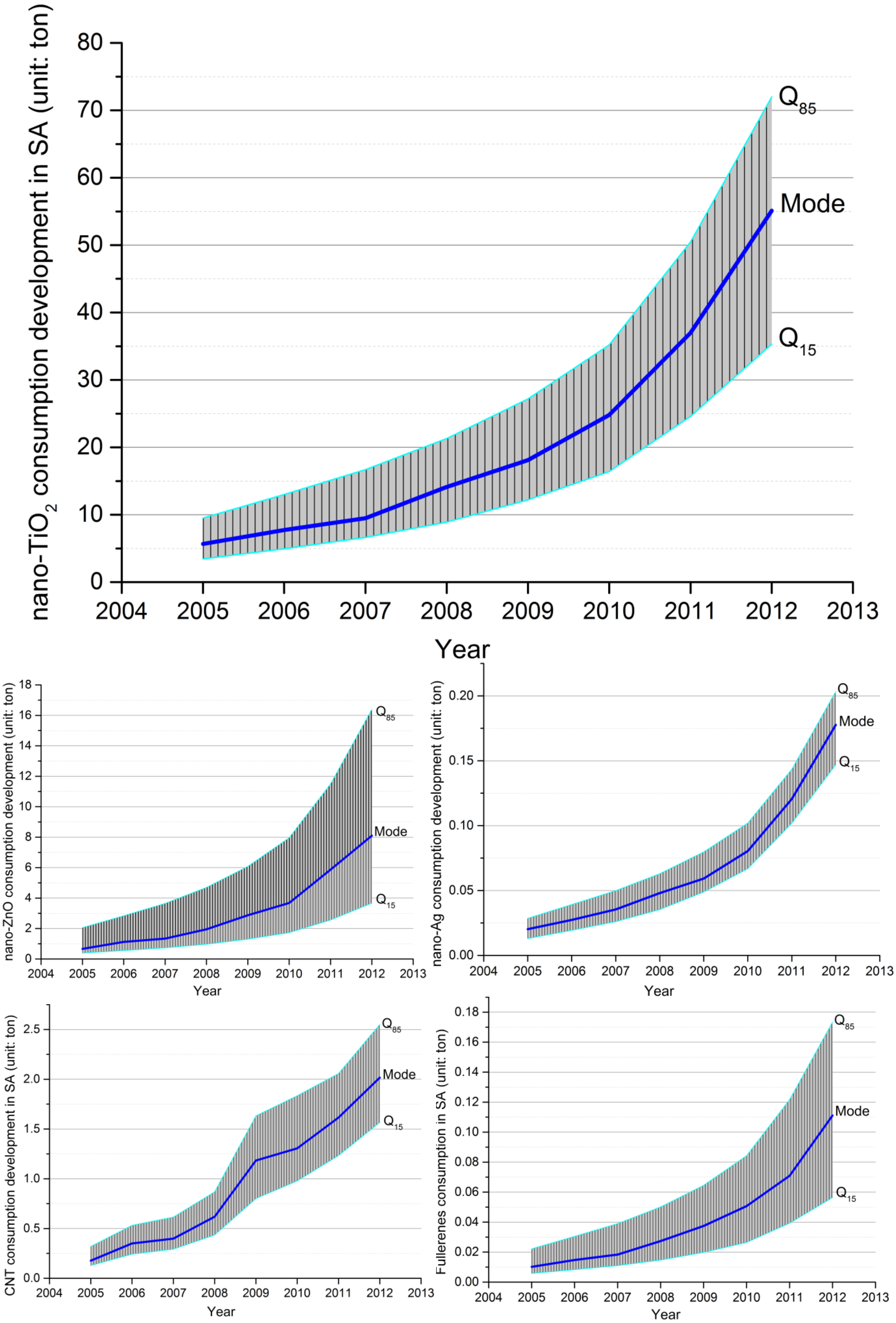


Figure S3: Estimation of five ENM production/consumption developments in South Australia between 2005 and 2012. Curves of mode values (most probable values) and ranges between quantile 15 and quantile 85 are shown.

Transfer coefficients of ENM from production to technical and environmental compartments

ENM release from products to wastewater, to air, and direct to soil, is normally dependent only on the way they are attached to products and the scenarios of how these products are used. In this case, release is assumed independent of where they are consumed but only dependent on the product categories. Therefore, it is reasonable to assume that the ENM release from products to these compartments in South Australia is the same as it is in Europe[25], which was summarised from literature reviews or if not on expert judgement. There is no incineration plant in South Australia; therefore all the solid wastes if not recycled or exported go to landfill.

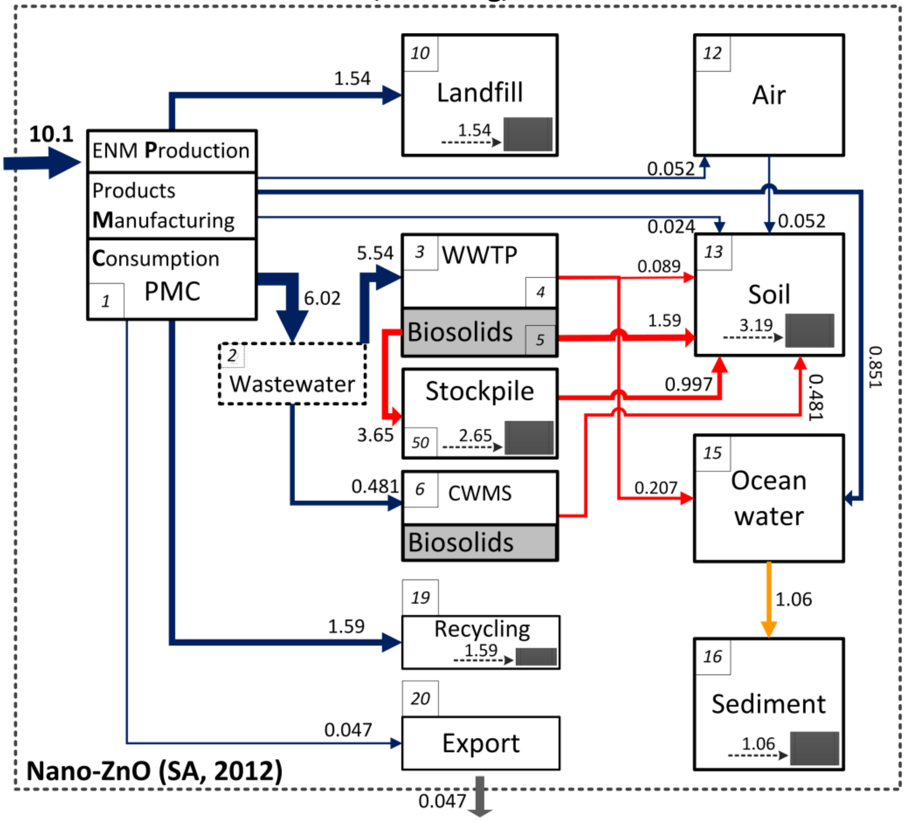
The distribution of ENM contained in cosmetics especially those in sunscreen to wastewater and to ocean water is very much dependent on where the applications of these products take place. An unpublished study by Conroy et al.[34] suggested a ratio of 6.78:1 for sunscreen applications inland (ending up in wastewater) and on beach. According to a local government report [35], around 10% of plastic products are recycled, 6% is exported out of South Australia; for glass 66% is recycled and 3% is exported; for household electronics 30% is recycled, 19% is exported; for textile 18% is exported; 60% of cement is recycled and reused; for paper 1% is recycled. 61% is recycled. Metal in South Australia according to a report prepared for government of South Australia[36], in average 23% is recycled and 67% is exported. Table S8 below shows the ENM release transfer into technical and environmental compartments. In probabilistic modelling, to cover the unknown uncertainty of these data, minus and plus 50% of these single numbers are deviated to get a range for a transfer coefficient, together with their original values they were used to build a triangular distribution for each ENM transfer coefficient.

Table S8: Transfer coefficients of ENM from production to technical and environmental compartments on the basis of Sun et al.2014[25]

	Wastewater	Landfill	Air	Soil	Ocean water	Recycling	Export
nano-TiO_2							
Production	0.45	0.30	0.25				
Manufacture	0.45	0.30	0.25				
Plastics	0.05	0.79				0.10	0.06
Cosmetics	0.83	0.05			0.12		
Coating	0.90	0.05	0.05				
Batteries& Capacitors		0.51				0.30	0.19
Metals	0.05	0.05				0.23	0.67
Paints	0.01	0.35	0.01	0.01		0.62	
Light Bulbs		0.80				0.20	
Glass & Ceramics	0.01	0.30				0.66	0.03
Filter	0.25	0.70	0.05				
Consumer Electronics		0.51				0.30	0.19
Textiles	0.01	0.80	0.01			0.00	0.18
Dietary Supplyment	0.90	0.10					
Ink	0.80	0.20					
Cement	0.01	0.37				0.62	
Cleaning agent	0.95	0.05					
Spray	0.85	0.05	0.10				
Paper		0.38				0.01	0.61
Sport goods	0.02	0.96	0.02				
WWTP	0.95	0.05					
nano-ZnO							
Production	0.45	0.30	0.25				
Manufacture	0.45	0.30	0.25				
Plastics		0.84				0.10	0.06
Cosmetics	0.83	0.05			0.12		
Cleaning agent	0.95	0.05					
Foods	1.00						
Textiles	0.11	0.70	0.01			0.00	0.18
Metals	0.05	0.05				0.23	0.67
Paints	0.06	0.30	0.01	0.01		0.62	
Glass &Ceramics	0.06	0.25				0.66	0.03
Consumer Electronics		0.51				0.30	0.19
Woods	0.06	0.94					
Filter	0.25	0.70	0.05				
Paper		0.38				0.01	0.61
nano-Ag							
Production	0.45	0.30	0.25				
Manufacture	0.45	0.30	0.25				
Plastics	0.14	0.70				0.10	0.06
Cosmetics	0.80	0.03			0.17		
Coatings&Cleaning agent	0.90	0.05	0.05				
Metals	0.05	0.05				0.23	0.67
Paints	0.06	0.92	0.01	0.01			
Glass & Ceramics	0.06	0.30				0.61	0.03
Filter Aggregates	0.35	0.60	0.05				
Consumer Electronics	0.01	0.50				0.30	0.19
Textiles	0.35	0.50	0.03				0.12
MedTech	0.05	0.46				0.30	0.19
Additive to Soil				1.00			
Sanitary	0.15	0.85					
Dietary Supplyment	0.90	0.10					
CNT							
Production	0.45	0.30	0.25				
Manufacture	0.45	0.30	0.25				
Composites(Polymer)		0.83	0.01			0.10	0.06
R&D	0.05	0.95					
ConsumerElectronics		0.51				0.30	0.19
Paint	0.01	0.35	0.01	0.01		0.62	
Textiles	0.01	0.80	0.01				0.18
Aerospace		0.39	0.01			0.60	
Automotive		0.39	0.01			0.60	
Energy		0.50				0.50	
Sensor		0.50				0.50	
Fullerenes							
Production	0.45	0.30	0.25				
Manufacture	0.45	0.30	0.25				
Composites		0.83	0.01			0.10	0.06
Cosmetics	0.77	0.03			0.20		
R&D	0.05	0.95					
Motor oil (Lubricant)	0.05	0.05				0.90	
Metals (coating)	0.05	0.05				0.23	0.67
Electronics and optics		0.51				0.30	0.19
Catalysts		0.50				0.50	
Aerospace		0.39	0.01			0.60	
Energy/environment		0.50				0.50	

Results

Mass flowchart of nano-ZnO, nano-Ag, CNT and fullerenes



Compartments codes: Q15 – **Mode** - Q85

1 : 3.75 - **7.58** - 16.5
1 → 10 : 0.538 - **1.24** - 2.54
1 → 2 : 2.17 - **4.93** - 9.84
1 → 19 : 0.470 - **0.816** - 2.77
1 → 20 : 0.014 - **0.022** - 0.081
1 → 12 : 0.018 - **0.037** - 0.086
1 → 13 : 0.006 - **0.008** - 0.042
1 → 15 : 0.306- **0.677** - 1.40

2 → 3 : 1.99 - **4.48** - 9.05
2 → 6 : 0.165 - **0.313** - 0.799

4 → 13 : 0.023 - **0.030** - 0.158
4 → 15 : 0.056 - **0.067** - 0.365

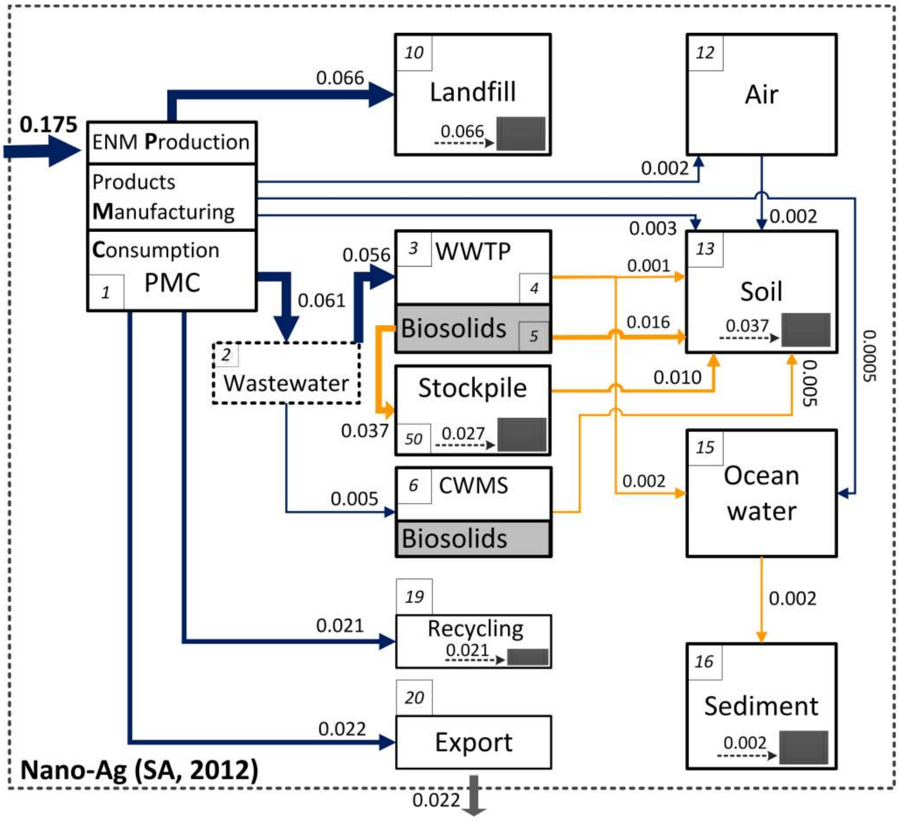
5 → 13 : 0.573 - **1.59** - 2.60
5 → 50 : 1.31 - **3.02** - 6.00

6 → 13 : 0.165 - **0.313** - 0.799

12 → 13 : 0.018 - **0.037** - 0.086

15 → 16 : 0.378- **0.870** - 1.74

50 → 13 : 0.000 - **1.22** - 2.07



Compartments codes: Q15 – **Mode** - Q85

1 : 0.147 - **0.179** - 0.203
1 → 10 : 0.053 - **0.065** - 0.079
1 → 2 : 0.047 - **0.058** - 0.075
1 → 19 : 0.015 - **0.020** - 0.027
1 → 20 : 0.016 - **0.020** - 0.027
1 → 12 : 0.002 - **0.002** - 0.003
1 → 13 : 0.001 - **0.002** - 0.004
1 → 15 : 0.0003 - **0.0004** - 0.0007

2 → 3 : 0.043 - **0.053** - 0.069
2 → 6 : 0.003 - **0.005** - 0.006

4 → 13 : 0.0002 - **0.0003** - 0.001
4 → 15 : 0.0006 - **0.0007** - 0.003

5 → 13 : 0.012 - **0.016** - 0.020
5 → 50 : 0.028 - **0.035** - 0.047

6 → 13 : 0.003 - **0.005** - 0.006

12 → 13 : 0.002 - **0.002** - 0.003

15 → 16 : 0.0007 - **0.001** - 0.002

50 → 13 : 0.006 - **0.011** - 0.014

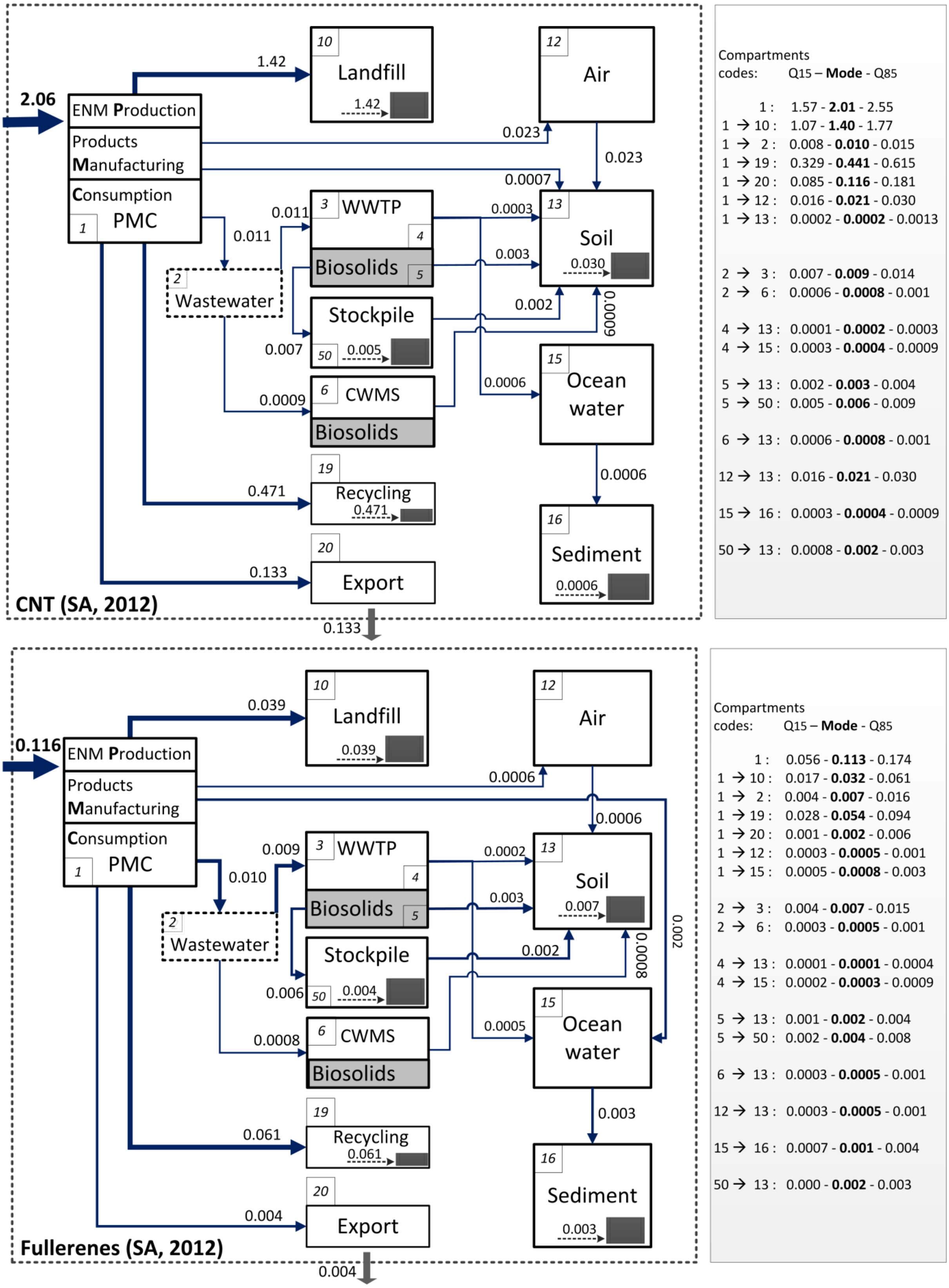


Figure S4: Annual mass flow of nano-ZnO, nano-Ag, CNT and fullerenes in South Australia in 2012, unit: ton. The values taken are MEAN values from each probability distribution; meanwhile mode values and quantile 15(Q_{15}) and quantile 85(Q_{85}) are also given in the right text field. A compartment code is given to each compartment in the corner of each compartment box. Arrows between two compartment codes in the right text field indicate flows from one compartment to the other. The volumes of flows are determined by transfer coefficients (TC) that describe the exchange of

ENM between and within these boxes. All the ENM mass-flows are computed regardless of their agglomerated and aggregated form; the balance between input and put flows from one compartment might not be 100% closed due to rounding. Blue arrows indicate all the nano flows which is in contrast to red arrows indicating ENM flows transformed into other chemical variants; yellow arrows is the mixture of nano flows and transformed flows, in other words the mixture of blue and red flows. In this concrete case, red flows means the transformed nano-ZnO flows after waste water treatment processes; yellow flows are the flows comprising both transformed nano-Ag after waste water treatment and the survived nano-ZnO flows from waste water treatment. Black squares indicate the ENM sinks such as landfills, soils and sediments.

In Figure S4 the Ag flow after waste water treatment is shown as a mixture of flows of nano-Ag and transformed nano Ag. The differentiation of them is not given. Therefore, a more explicit description of the nano-Ag flows, transformation and distributions from waste water treatment plant (WWTP) to ocean water, soils, stockpile and landfill is demonstrated by Figure S5 below.

Over the processes of WWTP, majority of ENM including nano-Ag end up in biosolids, there are many studies in this context.[37-41] In sewage effluent between 86% and100% of nano-Ag is transformed to Ag_2S and in biosolids only 1% to 2% of nano-Ag can survive and remains its original nano form, the rest is transformed into Ag_2S too due to the presence of abundant sulphate primarily from urine.[40] In South Australia around 70% of the sewage effluent is discharged into ocean, the remaining 30% is recycled for irrigation of grassland, garden, viticulture and horticulture etc. So the flows of surviving nano-Ag and transformed nano-Ag are proportional to the distribution of sewage effluent to soils and ocean. All of biosolids is used in soils, either directly applied in agricultural soils or first composed or stockpiled for later soil use. Again these values are also valid for the distribution of nano-Ag and transformed nano-Ag contained in biosolids to soils, stockpile and landfill.

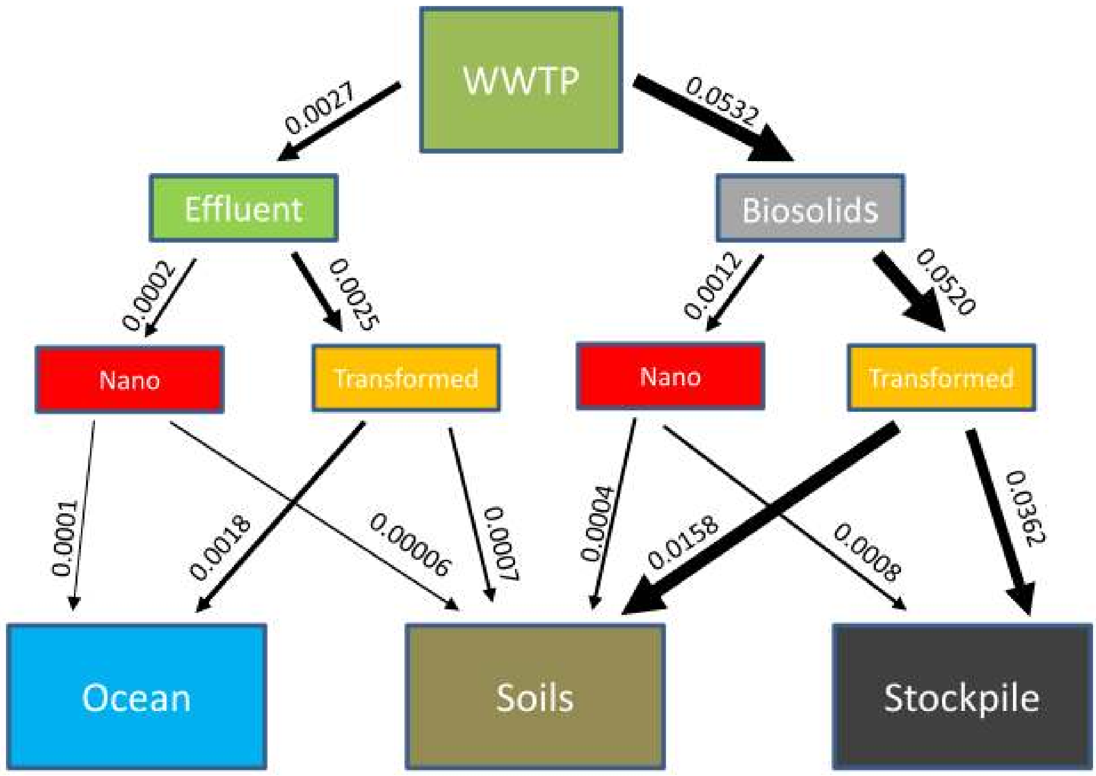


Figure S5: Mass flow, transformation and distributions of nano-Ag from Waste Water Treatment Plant (WWTP) to ocean gulf, soils and stockpile in South Australia in 2012, unit: tonnes. Mass balance due to individual rounding might not be 100% closed.

Table S9: Summary of accumulated concentration of five ENMs in 2012. Values are mode values (most probable values) taken from the probability distributions.

Compartments			nano-TiO_2	Zn from nano-ZnO	Ag from nano-Ag	nano-Ag	CNT	fullerenes	Units
Biosolids treated soils	Greater Adelaide		4.5E+02	9.2E+01	9.8E-01	2.3E-02	2.1E-01	1.5E-01	µg/kg
	Yorke & Mid North		1.7E+01	3.7E+00	3.8E-02	9.0E-04	8.1E-03	6.0E-03	
	Limestone Coast		1.0E+01	2.2E+00	2.3E-02	4.7E-04	4.7E-03	3.7E-03	
	Murray-Mallee		6.9E+00	1.5E+00	1.5E-02	3.2E-04	3.1E-03	2.4E-03	
	Kangaroo Island		9.5E+00	1.7E+00	2.0E-02	1.1E-04	3.9E-03	2.5E-03	
	Far North		2.3E+00	4.4E-01	5.0E-03	2.8E-05	1.0E-03	6.4E-04	
	Eyre & Western		1.9E+00	4.0E-01	5.0E-03	9.8E-05	8.4E-04	6.1E-04	
WWTP effluent treated soils	Grass/gardens/pastures		1.8E+01	4.1E+00	3.5E-02	1.2E-03	2.3E-02	1.4E-02	
	Horticulture		1.6E+01	4.1E+00	3.1E-02	1.0E-03	2.1E-02	1.4E-02	
	Viticulture		8.6E+00	2.0E+00	1.6E-02	4.5E-04	1.1E-02	7.0E-03	
Gulfs sediments	St. Vincent	Bolivar	3.8E+01	9.0E+00	7.4E-02	2.5E-03	4.8E-02	3.1E-02	
		Glenelg	1.5E+01	3.6E+00	2.9E-02	9.7E-04	1.9E-02	1.2E-02	
		Christies Beach	1.0E+01	2.4E+00	2.0E-02	6.6E-04	1.3E-02	8.4E-03	
	Spencer	Whyalla	2.5E+00	5.9E-01	4.9E-03	1.6E-04	3.2E-03	2.1E-03	
		Pirie	2.8E+00	6.8E-01	5.6E-03	1.8E-04	3.6E-03	2.4E-03	
		Augusta	9.8E+00	2.3E+00	2.0E-02	6.3E-04	1.3E-02	8.1E-03	

Table S10: Summary of the measured Zinc and Silver concentrations in SA biosolids and soils

Metals	Compartments	Values or Range	References
Zn	Biosolids	**1. 300-1,300 mg/kg** **2. 503-688 mg/kg**	**1. Personal communications with SA Water[42]** **2. Unpublished data from University of South Australia[43]**
	Soils	**1. 12-69 mg/kg** **2. 11-18 mg/kg** **3. 50 mg/kg** **4. 7-58 mg/kg** **5. 10-43 mg/kg** **6. 4-110 mg/kg** **7. 4-61 mg/kg** **8. 11-86 mg/kg**	**1. Unpublished data from University of South Australia[43]** **2. Betrand et al. [44]** **3. Donner et al.[45]** **4. Broos et al.[46]** **5. Donner et al.[47]** **6. Blackburn et al.[48]** **7. Tiller et al.[49]** **8. McKenzie et al.[50]**
Ag	Biosolids	**1. 6-10 mg/kg**	**1. Unpublished data from University of South Australia[43]**
	Soils	**1. 0-0.1 mg/kg** **2. 0-0.1 mg/kg** **3. 0-0.04 mg/kg** **4. 0.01-1.01 mg/kg**	**1. Cornelis et al. [51]** **2. Cornelis et al.[52]** **3. Settimio et al.[53]** **4. Unpublished work[54]**

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