LCA Case Studies

The Effect of Compact Formulations on the Environmental Profile of Northern European Granular Laundry Detergents

Part I: Environmental Risk Assessment

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Abstract. Regular (1988) and compact granular (1992, 1998) laundry detergents were compared on the basis of two distinct, complementary approaches: Environmental Risk Assessment and Life-Cycle Assessment. The results are presented in this paper and an accompanying paper in this volume (Part II: Life-Cycle Assessment). Exposure data from The Netherlands and Sweden were used for this retrospective analysis. The time period studied (1988-1998) spans many innovations in laundry detergents, one of which was the introduction of compact detergents. The aquatic risk assessment resulted in risk quotients below 1 for all detergent ingredients in both countries over the period studied. Furthermore, it showed that risk quotients decrease two to five-fold between 1988 and 1998 in each country due to the introduction of compact detergents. Slightly lower risk quotients were observed in Sweden, when compared to The Netherlands, attributable to the lower water hardness resulting in lower detergent usage per wash cycle in that country. If water hardnesses were equal, the outcome of the product risk assessments would also be the same in the two countries.

Keywords: Compact detergent; environmental risk assessment; granular laundry detergents; LCA case studies; life cycle assessment; product risk assessment; regular detergent; sensitivity analysis

Introduction

The introduction of compact detergents in the early 1990's was the result of a major technological innovation in the detergent industry. In order to accomplish this, products with higher weight-efficiency had to be developed. Compact products required less packaging and were accompanied by a reduction in manufacturing waste and lower laundry detergent dosages per wash load, hence a reduction in environmental releases from households [1-7]. The new generation of compact products, promising the same number of wash loads in smaller packages, met with varying degrees of consumer acceptance in different European markets, ostensibly due to differences in laundering practices and cultural preferences. While Northern Europe had essentially converted to compact and super compact detergents by 1998, many Southern European consumers, such as the French and the Spanish, continued to prefer traditional, bulky granular products [1]. This may be due to a number of factors such as value perceptions (compacts may be seen as 'less product' for the money), high consumer loyalty to traditional brands, less socio-economic pressure attached to environmental benefits, just to name a few. The relatively high level of success of compact detergents in Northern Europe may be related to the environmental benefits they are perceived to bring in terms of resource-efficiency and safety. However, such perceived benefits have not been systematically analyzed nor communicated to consumers.

1 Objectives

Ariel regular (1988-regular), Ariel Ultra (1992-compact) and Ariel Futur (1998-super compact) granular detergents, as represented by formulations of The Procter & Gamble Company (P&G), were analyzed using two different, complementary approaches: Environmental Risk Assessment (ERA) and Life-Cycle Assessment (LCA) [8]. ERA is the evaluation of the probability that a specific, adverse effect will occur as the result of a certain exposure. Life-Cycle Assessment (LCA) is a methodology developed to evaluate the mass balance of inputs and outputs of systems and to organize and convert those inputs and outputs into environmental themes or categories relative to resource use, human health and ecological areas [9,10]. The Life-Cycle Inventory (LCI) refers to the mass balance itself. Each approach provides a unique environmental analysis by compiling different information from distinct perspectives. The most cost-effective way to conduct ERA and LCA is in sequential steps or 'tiers', with each step refining the previous assessment by bringing in new data, as a result of which the uncertainty in the assessment is reduced. A screening-level dataset was adequate for the purposes of this study, therefore the assessments presented here are at tier-1 level.

Our objectives were:

1. To characterize and evaluate trends in environmental parameters of granular laundry detergents in Europe from 1988 until 1998, the period during which the transition from traditional detergents to compacts took place.

2. To compare environmental parameters of granular laundry detergents in The Netherlands and Sweden. The compact and super compact products in Sweden have been developed to be compliant with the national Ecolabel scheme since 1996, while this has not been the case for The Netherlands[1]. If the assessments would demonstrate differences, we wanted to characterize those differences and understand what they were due to.

2 Detergent Marketing Trends, Laundering Habits and Conditions in The Netherlands and Sweden

In the course of the last decade, compact granular laundry detergents have become market leaders in Northern Europe [1] as illustrated by the A.C. Nielsen shares (Wavre, Belgium) presented in **Table 1**. Nielsen market share information is collected by a private company on a variety of products and is used by companies to assess market trends. In 1988, traditional granular laundry detergents accounted for 78 to 87% of the market in The Netherlands and Sweden. During the following 3 or 4 years, compact granular products were introduced and gained significant terrain. In 1992, more than 60% of the laundry detergent market in The Netherlands and 44% of the Swedish market had converted to compact granular products. Based on '98/'99 Nielsen shares, this increasing trend in the usage of compact detergents has continued: in The Netherlands and Sweden, market shares reached approximately 69% and 61%, respectively. P&G market distribution reveals a similar trend: in 1998, virtually all P&G laundry detergents sold in these two countries were compact granular powders.

To guide their research and development efforts, companies such as P&G collect data on detergent markets, laundering conditions and practices worldwide. Much of this information is obtained through consumer surveys designed to characterize laundering habits and consumer expectations for well-defined population groups. Because this information is often the main driver of the development of new products or product upgrades, it is normally considered confidential and rarely published. **Table 2** illustrates laundering conditions and practices in The Netherlands and Sweden, two countries very similar in terms of laundering habits, per-capita Gross Domestic Product (GDP) and average household size. In both countries, the bulk of the laundry is machine-washed (~90%) and four to five loads of laundry are washed per household per week. Peripheral laundry practices such as soaking, pre-treatment or pre-wash are relatively uncommon. Most loads are washed at 40°C or 60°C. Based on 1988 and 1998 data, the wash temperatures evolved during that decade. The frequency of 'boil wash' decreased by about 50% and has been replaced by wash loads at lower temperatures. In 1998 the majority of the loads was washed at low temperature (~40°C). Based on our estimation, total laundry detergent consumption in Sweden and The Netherlands dropped by almost 50% between 1988 and 1998. Per-capita detergent usage is about 1.5 times lower in Sweden than in The Netherlands, principally due to the lower hardness of Swedish water (Table 2). Harder wash water has a higher mineral content and it is primarily the Ca^{2+} and Mg^{2+} ions that form insoluble salts with surfactants, thus effectively removing the principal cleaning agents from the wash solution. Higher water hardness therefore requires more surfactant to achieve equal cleaning performance.

3 Environmental Risk Assessment

The objective of the environmental risk assessments is to evaluate whether and, if so, how the ingredient risk quotients of granular laundry detergent products changed between 1988 and 1998. In other words, what effect did the transition from traditional detergents to compacts have on risk quotients? To make a fair comparison, we assumed that each product accounted for 100% of the market. Because of that assumption, the assessments presented here are only useful for *comparative purposes between products*, in this case to *illustrate trends in risk quotients* in going from traditional laundry detergents used in 1988 to compacts in 1992 and super compacts in 1998, and to identify differences between countries. The product ERA presented in this article is therefore not absolute. Moreover, the ERA presented here

[1] A discussion about the relevance and benefit of the Nordic Swan Ecolabel or national ecolabelling schemes in general is outside the scope of this paper

Table 1: Market evolution in The Netherlands and Sweden between 1988 and 1996 for traditional granular, compact granular and liquid detergents (including regular and compact) based on the A.C. Nielsen fiscal market share 1987/88, 1991/92 and 1998/99)

	The Netherlands			Sweden		
Market share (%)	**1988**	**1992**	**1998**	**1988**	**1992**	**1998**
Regular Granular	77.8	25.9	8.7	87.4	31.2	20.8
Compact Granular	0.5	60.6	69	0	44.3	61.3
Liquids	21.7	13.5	19[a]	12.6	24.5	14.7
Tablets	-	-	3.3			3.1
P&G market distribution (%)						
Regular Granular	62	3	6	24	-	1
Compact Granular	1	86	81	-	52	58
Liquids	37	11	14	76	48	41

a) 5.2% regular liquid and 13.8% compact liquid
b) 0.5% regular liquid and 14.2% compact liquid

Table 2: Country statistics, laundering practices, wash temperatures, estimated detergent consumption and recommended dosages for selected P&G products in The Netherlands and Sweden

	The Netherlands			Sweden		
	1988	1992	1998	1988	1992	1998
Population (in million)	14.350	15.183	15.678	8.225	8.680	8.875
Per-capita GDP (US$ x 1000)		21	23		28	25
Household size	-	2.32	2.41	-	2.16	2.23
Wash loads/2 weeks/per household						
Machine wash	9.4	12.6	9.6	-	-	8.7
Hand wash	0.8	0.7	1.0	-	-	1.1
Peripheral treatments (% of loads)						
Soak[a]	4	3	2	-	-	2
Pre-treat	6	6	1	-	-	1
Pre-wash[b]	25	-	12	16	-	16
Wash Temperature (% of loads)						
30°C	7	-	18	3	-	4
40°C	32	-	40	47	-	49
60°C	41	-	33	37	-	42
>60°C	20	-	9	13	-	5
Water consumption (l/cap/day)	213	212	210	291	279	243
Water hardness (dH)[c]	-	-	9.6	-	-	2.9
Estimated laundry detergent consumption based on Nielsen shares (kg/cap·year)	10.4	7.6	5.56	7	5.8	3.4
Recommended dosage (g/wash)[d]	153	109	75	115	51	37.5
Product name and type	Ariel, regular	Ariel Ultra, compact	Euro Futur, super compact	Ariel, regular	Ariel Ultra, compact	Ariel Futur, super compact

a) Laundry + water + detergent are allowed to interact without agitation for 10-15 minutes
b) Laundry is treated with detergent (usually liquids) before wash in washing machine with additional detergent
c) Soft water = 0 - 4°dH; medium hardness = 4 - 12°dH; hard water >12°dH
d) for medium soiling, medium hardness for The Netherlands and soft water for Sweden

addresses the aquatic compartment only; this does not imply that the similar trends occurred for the other environmental compartments (sediment, soil, etc.). The overall environmental impact assessments of individual ingredients are published elsewhere [11-20].

ERA is defined as the process that evaluates the probability of adverse effects in the environment as a result of exposure. It is outside the scope of this paper to describe the ERA methodology in depth, as this information has been published elsewhere [21-23]. In brief, estimated or measured exposures are compared with adverse-effect concentrations that can also be either estimated or measured. If the highest anticipated exposure concentration (the predicted environmental concentration or PEC) is below what is considered to be the 'protective' concentration (the predicted no-effect concentration or PNEC), the risk is considered acceptable. In other words, the risk characterization (also risk quotient or RQ), defined as the ratio between the PEC and the PNEC, should be lower than 1. The choice of the PEC and the PNEC is a matter of convention and agreed statistical procedures. Such an assessment needs to be conducted for each environmental compartment where significant exposure is expected. To account for the uncertainty associated with the determination of the PNEC from laboratory toxicity testing, application factors (AF) - are applied. The AF allows for uncertainties such as species inter-sensitivity, acute vs. chronic effects and laboratory to field extrapolation. The magnitude of the application factor is also a matter of convention and depends on the toxicity database: acute ECs require larger AFs than chronic ECs, which in turn require a larger AF than the ecosystem data. As a result of laboratory toxicity testing, one or more Effect Concentrations (EC) are obtained, depending on how many different effects are observed. Effect concentrations will likely be different for different test species. In order to arrive at a Predicted No-Effect Concentration (PNEC) for each relevant compartment the lowest observed EC is divided by the AF.

ERA is a multi-step or tiered process, progressing from the use of simple screening tests and conservative assumptions to increasingly more realistic, refined experiments paired with more realistic assumptions, until a point is reached where one can show with reasonable certainty that predicted environmental concentrations are expected to remain below the predicted, no-effect concentration (PEC/PNEC <1). In the event that a favorable PEC/PNEC ratio is not reached at the highest possible tier, risk management is called for: this may mean a decision not to use the chemical, to limit its use or to use it within contained systems to control exposure.

3.1 Material and methods

Three P&G laundry products were selected for this analysis: Regular Ariel as formulated in 1988, representing the traditional granular detergent; compact granular Ariel Ultra as formulated in 1992, representing the first generation of compact granular detergents and Ariel Futur as formulated in 1998, representing the second generation of compacts or the 'super compact' granular detergents (Table 2). Compacts and super compacts were introduced prior to 1992 and 1998, respectively and slowly gained a significant market share. We selected 1992 and 1998 as representative for two periods during which both compact and super compact granular laundry products were well established in the Dutch and Swedish markets. Because of the high fraction of wastewater that receives biological treatment in these countries (95% in Sweden, 88% in NL [24]), the simplified assumption was made that 100% of the wastewater is treated.

Risk quotients (or PEC/PNEC ratio) for ingredients were calculated using the following algorithm and using country-specific data for 1988, 1992 and 1998 (ingredient consumption, population density, sewage flow):

$$RiskQuotient = \left(\frac{IngredientConsumption \times (1 - Removal)}{Sewageflow \times Dilution} \right) / \ PNEC$$

Ingredient Consumption = expressed in mg/capita·year
Removal = fraction eliminated by wastewater treatment
Sewage flow = per capita water consumption in l/year.

It was furthermore assumed that the sewage treatment plant effluent is diluted 10-fold by the receiving river, as recommended in the European Technical Guidance Document for Tier 1 ERA [25].

A last assumption was that, at the time of each analysis, each P&G brand represented 100% of the detergent present on the market.

This assumption was necessary because companies normally do not share compositional information on their products, but at the same time assessments should be based on total exposure, not just exposure to the products of one company. Ingredient consumption (mg/year per capita) was therefore calculated assuming a 100% market share for the selected P&G products. For P&G-proprietary ingredients (i.e. ingredients only used by P&G), consumption was calculated using real tonnage.

Some laundry detergent ingredients are also used in other applications (e.g. bar soaps, shampoos, dish washing detergents, etc.). Such contributions are typically small (<10%) relative to the amount of laundry detergents [26]. To account for uses of ingredients in products other than laundry detergents, ingredient consumption was increased by 10%.

Because the objective of the exercise was to monitor trends in risk quotients of ingredients over the years, we ensured that the same fate and effect data was used throughout, thereby eliminating any variability due to the use of different removal and toxicity data for different time periods.

The removal of ingredients by wastewater treatment was estimated from laboratory testing such as traditional biodegradation tests or bench-top wastewater treatment simulation tests. Toxicity data was derived from acute or chronic toxicity laboratory assays with representative aquatic species.

To the extent possible, the removal and toxicity data used for this analysis are identical to those previously published in the Detergent Ingredient Database or DID List of the European Union (EU) [27]. For ingredients not included in the DID List, internal P&G data or published data was used to support the risk assessments.

The DID List is the result of a consensus achieved among European detergent manufacturers, raw material suppliers, member states and the Commission in charge of the development of Ecolabel criteria for the EU. The so-called Long Term Effect concentrations (LTE) from the DID List were considered as an acceptable surrogate to the PNEC. While not developed for risk assessment purposes, the DID list contains an acceptable PNEC estimate for most of the ingredients needed for this comparative assessment.

4 Results

The Tier 1 risk assessment presented in this article is based on a number of assumptions that may lead to an over or underestimation of environmental exposure. The use of default removal values such as those listed in the DID List is important and this will be discussed in more detail later. National consumption data on detergent ingredients are difficult to obtain. The Swedish Society for Nature Conservation recently published its own estimates of the evolution of the detergent consumption in Sweden between 1988 and 1996 [3]. Another source of such information (our estimate) can be derived from the Nielsen shares. While it seems that we may have slightly underestimated the total LAS consumption in Sweden in 1988, we probably overestimated the consumption of other ingredients such as perborate, EDTA, phosphate and polycarboxylates (**Table 3**). It is improbable that any of these sources accurately reflects the true consumption of these detergent ingredients in Sweden in 1988. The fact that our estimates were on the high end of the range for all ingredients except LAS indicates that this risk assessment probably erred on the conservative side as far as consumption is concerned.

Table 3: Consumption of common laundry detergent ingredients in 1988 estimated from Nielsen shares and from the Swedish Society for Nature conservation

	Nielsen shares, 1988 (metric tons)	Swedish Society for Nature Conservation, 1988 (metric tons)	
		Laundry	Total market
LAS	4712	4600	6300
EDTA[a]	129	110	120
Perborate tetra (as borate)	6333	5200	5200
Phosphate as STPP	15200	9500	11200
Polycarboxylates	1647	890	910

a) EDTA is no longer used in laundry detergents in Europe

Table 4: Risk quotients for the chemicals used in P&G detergents in Sweden (SW) and The Netherlands (NL) in 1988, 1992 and 1998. Removal and Long Term Effect (LTE) are from the DID List or P&G internal database. Empty cells indicate that the ingredient was not used in the formula

			The Netherlands			Sweden		
Ingredients	**(1-Fraction Removed in STP)**	**LTE**	**1988**	**1992**	**1998**	**1988**	**1992**	**1998**
Alcohol Ethoxy Sulfate (AE_3S)	0.03	0.15			0.015			0.017
Brightener 1	0.5	1	0.011	0.010	0.003	0.009	0.006	
Brightener 2	0.5	3.13			<0.001			
Butylated hydroxytoluene	0.12	0.05				<0.001		
C25 Alcohol Ethoxylate ($A_{25}E_5$)	0.03	0.18			0.047			0.023
C25 Alcohol Ethoxylate ($A_{25}E_7$)	0.03	0.24	0.099	0.054			0.031	
C25 Alkyl Sulfate	0.02	0.1			0.058			0.068
C28 Alkyl Sulfate	0.02	0.15			0.021			
Carboxy methyl cellulose	0.75	250	<0.001		<0.001	<0.001		<0.001
Clay	0.05	1000				<0.001		
Dye 1	0.04	0.02		<0.001	<0.001	<0.001	<0.001	<0.001
Dye 2	0.5	0.1	0.001		0.004	0.001		
Enzymes	0.09	25	<0.001	0.001	0.001	<0.001	<0.001	<0.001
Ethylene diamine disuccinate	0.05	10			<0.001			<0.001
Ethylene diamine tetraacetic Acid	1	11	0.003			0.001		
Fatty Alkyl Sulfate	0.02	0.55	0.014		<0.001			<0.001
Hydrogenated fatty acid	0.05	1.6				0.002		
LAS	0.05	0.3	0.146	0.140	0.086	0.090	0.081	
$MgSO_4$	1	800	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Na-carbonate	0.8	250	0.006	0.006	0.003		0.003	0.001
Na-citrate	0.04	85		<0.001	<0.001		<0.001	<0.001
Na-perborate	1	6	0.494	0.367		0.121	0.212	
Na-percarbonate	0.8	250			0.004			0.002
$NaSO_4$	1	1000	0.002		<0.001	0.001		<0.001
Na-toluene tulphonate	0.09	6.6				0.001		
P&G proprietary ingredient	0.03	0.12			0.024			0.012
P&G proprietary ingredient	0.03	0.03				0.055		
P&G proprietary ingredient	0.05	0.25			0.004			0.002
P&G proprietary ingredient	0.03	0.13				0.004		
Perfumes	0.1	0.02	0.031	0.065	0.043	0.009	0.030	0.089
Phosphates (as P)	0.6	1000				0.001		
Phosphonates	0.5	7	0.003	0.003	0.003		0.002	0.001
Polycarboxylates	0.5	124	0.002	0.002	<0.001	0.001	0.001	0.001
Polymer	1	100			<0.001			<0.001
Silica	0.05	100			<0.001			<0.001
Silicate	0.8	1000	<0.001	<0.001	V0.001	0.001	0	<0.001
Silicone	0.5	4.82	0.007	0.005	0.002	0.005	0.003	0.001
Soil release polymer	0.5	310			<0.001			<0.001
Sorbitol	0.09	100			<0.001			<0.001
Starch	0.07	250			<0.001			<0.001
TAED (bleach activator)	0.09	500	<0.001	<0.001	<0.001		<0.001	<0.001
Tallow-AE_{11}	0.03	0.05		0.070		0.049	0.040	
Tallow-AE_{25}	0.05	0.36	0.014	0.013			0.007	
Tallow-AE_{50}	0.75	2.5						0.005
Zeolite	0.05	120	0.001	0.001	0.001		0.001	<0.001
		n	20	18	32	21	18	28
		mean	**0.042**	**0.041**	**0.009**	**0.017**	**0.023**	**0.008**
		sum	0.84	0.74	0.32	0.35	0.42	0.22
		90th percentile	0.10	0.09	0.04	0.06	0.05	0.02

For all ingredients used in 1988, 1992 and 1998, the calculated risk quotients were all below 1 (**Table 4** and **Fig. 1**). In The Netherlands, the introduction of compact granular formulations had little if any impact on the risk quotient distribution (means, sums and 90th percentiles are equivalent). In Sweden, the risk quotients showed a slight overall

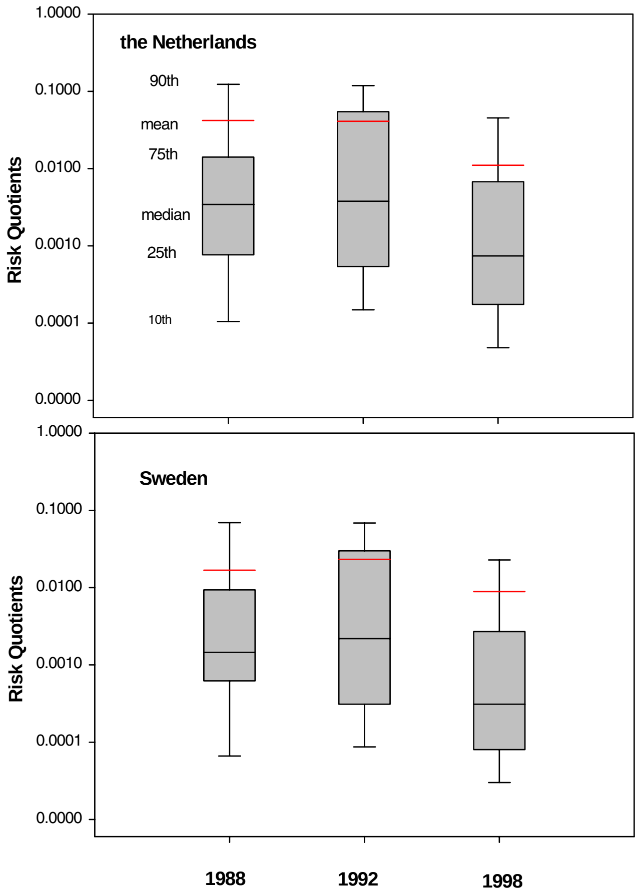

Fig. 1: Box plot of risk quotients for laundry detergent ingredients in The Netherlands and Sweden during 1988-1998. The boundary of the box indicates the 75th and 25th percentile and whiskers above and below the box indicate the 90th and 10th percentile. Mean and median are also indicated

increase in 1992, despite a clear decrease in detergent consumption (Table 2). This is due to a higher level of surfactants used in compact formulations compared with traditional granular products.

The risk quotient distribution changed drastically, although with the introduction of the 1998 formulations. A decrease in the means of 66 and 79% between 1992 and 1998 can be observed for Sweden and The Netherlands, respectively. A similar decrease can be observed for the 90th percentile or sum parameters, despite a significant increase of number of ingredients in the formulas (see Table 4).

The decrease in the risk quotients after 1998 is the result of both a lower detergent consumption and the introduction of a number of ingredients with lower individual risk quotients. It is important to note that the differences between the mean risk quotients values of 1992 and 1998 are not statistically significant in a t-test (a = 0.05).

The risk quotient distributions shown in Fig. 1 indicate lower values in Sweden when compared to The Netherlands in any given year. This can be explained by the lower recommended dosage of laundry detergent per wash load in Sweden, attributable to the lower water hardness in that country. When normalized for tonnage, population and per-capita water consumption, differences between the two countries

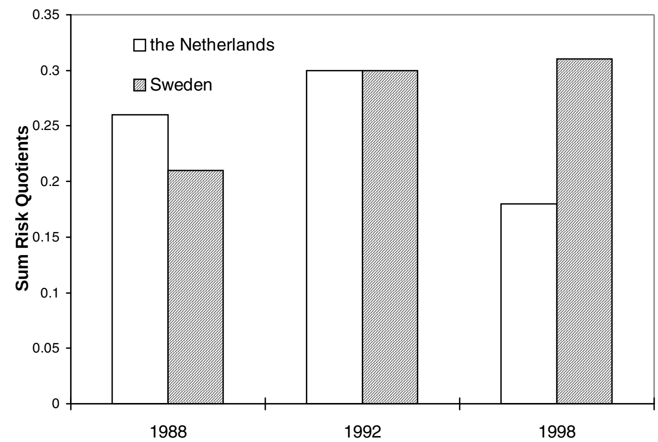

Fig. 2: Sum of average risk quotients in The Netherlands and Sweden in 1988, 1992 and 1998 after normalization for total tonnage and total water consumption. The only remaining variable is product composition

were canceled or even reversed (**Fig. 2**). The similarity between risk quotient distributions in 1992 indicates that products sold in both countries were identical in term of their risk quotient ratios. The reversal in 1998 is due to the higher level of surfactants in the Swedish product.

4.1 Sensitivity analysis

The data presented thus far were calculated based on point values without considering the uncertainty associated with them. For example, the value estimated for the detergent consumption in The Netherlands in 1988 was 10.48 kg per person annually. The real consumption, based on all the detergent tonnage information, may be higher. Variability is also associated with the other input parameters used to calculate risk quotients, such as the PNEC itself and the removal of each ingredient by sewage treatment. By assigning a theoretical variability to each input parameter, and using a Monte Carlo simulation technique [28], we estimated the potential impact of these uncertainties on the risk quotient distribution. A normal distribution with a ±15% fluctuation around the mean was used for the detergent consumption with a cut-off defined as 3 times the standard deviation. This means that the detergent consumption in The Netherlands would fluctuate between 8.91 and 12.05 kg per person annually. A ±15% variability in consumption was considered sufficient to count for the uncertainty associated with our tonnage estimation (A.C. Nielsen's market data may not cover the total market). For the LTE and removal figures, a normal distribution based on a fluctuation of ±30% with no cut-off was used. The variability of each of these parameters is actually dependent on the quality of the data generated, which is not available in the detergent ingredient database [27]. For reasons of simplicity (more than 130 individual RAs have been calculated), the same variability was assumed for all effect and removal parameters. In the context of a probabilistic risk assessment, the variability of the different parameters will need to be refined to represent the observed variability in laboratory or field experiments.

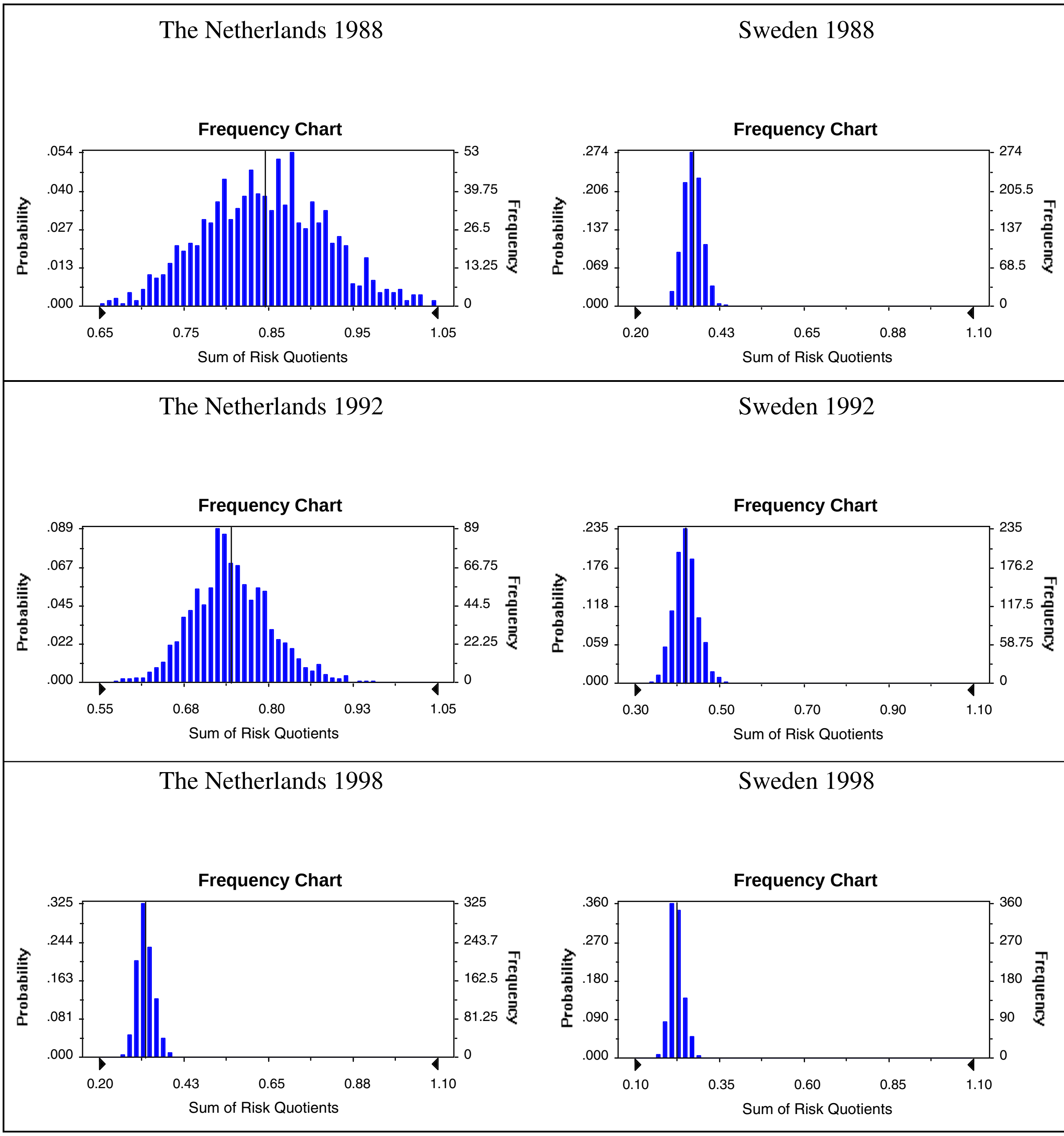

Fig. 3: Distribution of the sum of risk quotients based on a Monte Carlo simulation with Crystal Ball® (n=1000) for The Netherlands and Sweden (1988, 1992 and 1998), assuming a fluctuation of 15% for detergent consumption and a fluctuation of 30% for LTE and removal by sewage treatment. The horizontal shift between different risk quotient distributions reflects differences in the recommended product dose per wash load, per capita water consumption and product composition

Given these ranges for detergent consumption, effect and removals in sewage treatment, the Monte Carlo simulation was conducted with Crystal Ball® software (version 4.0). The simulation was halted after 1000 runs, as no more improvement could be obtained. The output requested was the sum of all risk quotient values in the form of a probability distribution. The underlying assumption here is that the toxic effects of the different ingredients would be additive.

The results presented in **Fig. 3** show that, even under the extreme high exposure scenario (i.e. the combination of highest consumption, highest toxicity and lowest removal), the probability that the sum of the risk quotients would be higher than 1 is nil (except for a few situations in The Netherlands in 1988). However, the 95th percentiles indicate a sum of risk quotients below 1 for all distributions.

4.2 Refinement of the ERA for selected ingredients

The risk assessments performed here are relative and representative of the type of data used to derive removal and effect values (mainly from the DID list). Input parameter distributions and therefore also risk quotient distributions can

Table 5: Risk quotients in The Netherlands for LAS, AS, AES and AE using either the DID List (Ecolabel 1995) or data from risk assessments performed in The Netherlands

		DID List input data			Input data from recent risk assessments		
Surfactant	Product	Average Removal %	LTE mg/L	Risk quotient	Removal Range (average) %	PNEC mg/L	Risk quotient
C_{10-13} LAS	NL88	95	0.30	0.13	98.0-99.6 (99.2)[a]	0.25	0.020
C_{25}AS	NL98	98	0.10	0.05	99.0-99.6 (99.2)	0.22	0.010
C_{25}AES	NL98	97	0.15	0.01	99.3-99.9 (99.6)	0.40	0.007
$C_{25}AE_{6-9}$	NL98	97	0.18	0.04	99.6-99.9 (99.8)	0.11	0.005

a) average removal values were used for the risk quotient calculation

be refined by using higher-tiered data. For example, the DID List contains the conservative removal estimates for surfactants by sewage treatment (95% for LAS, 98% for AS and 97% for AES and AE). Actual field monitoring studies, recently conducted in The Netherlands to support environmental risk assessments [14,26], showed that removals of LAS, AE, AES, AS and soap in the field were all higher than 99.0% at 7 selected sewage treatment plants, as a group considered to represent sewage treatment for the country. The removal at each individual facility was relatively constant and unaffected by plant operating parameters such as plant size, hydraulic retention time and sludge retention time [29]. The aquatic effect database was also re-analyzed to arrive at the most realistic PNECs for the same surfactants based on all available data at that time [19]. When the new PNEC and removal values were used for the assessment, instead of the values from the DID List, the risk quotient distributions for these surfactants improved towards a lower range of values (**Table 5**). This illustrates the relative nature of the ERA presented in Fig. 1 and our earlier point that comparative risk assessments between products should be based on the same-tier input data.

5 Discussion

Due to the tiered nature of the ERA process, which implies that the generation of new data stops as soon as safety have been demonstrated, the risk quotient of different ingredients cannot always be compared in absolute terms. An ingredient with a risk quotient of 0.1 is not necessarily 'less safe' than an ingredient with a risk quotient of 0.01. For example, a risk quotient of 0.1 may be changed to 0.05 or 0.01 when additional or higher-tier removal or toxicity data become available, leading to a more accurate estimation of the PEC and/or the use of a lower uncertainty factor. The risk quotients can be used as a relative measure of the environmental compatibility of different ingredients only if comparable, same-tier datasets are used for each. The product risk assessments presented here were intended to reveal trends and therefore did not always make use of the latest, highest-tier data available. As shown, comparative risk assessment between products at different points in time and/or place should take into account extra variables beyond the traditional toxicological and fate properties: per-capita detergent consumption, per-capita water usage, and representative sewage treatment practices are also part of the equation. Consumption is a reflection of product performance, local habits and local water characteristics. In Sweden, softer water leads to a lower detergent consumption per wash as compared to The Netherlands. Even though the intuitive conclusion may be that the laundering process would probably have less environmental impact in Sweden, a systematic comparison between two countries shows that risk quotients were virtually identical in 1992, while the situation in The Netherlands looked better in 1998, i.e. the risk quotients were lower there (Fig. 2). This could be explained by the higher surfactant content of the Swedish product in 1998.

The decrease of risk quotients during the last 10 years is attributable to new ingredients that are either more rapidly biodegradable, more highly removed, less toxic or more weight-efficient at times, but the principal factor is clearly the lower over-all consumption of detergents since the introduction of compact and super compact detergents (Table 2). Lower consumption has been driven by lower recommended dosage, but also less pre-wash, better washing machine efficiency, change in consumer habits (although these last 2 points are not documented here), etc. Because consumer acceptance of new products takes time, this shift has been gradual, as was apparent in 1998 in both Sweden and The Netherlands. Somewhat surprising perhaps is the fact that, despite clear environmental advantages, compact detergents were not originally developed for their environmental benefits, nor were these benefits recognized in the early days. When compacts first became popular in Japan, where space is at a premium and public transportation the norm, the smaller package that could contain products for the same number of wash loads was immediately seen as a real benefit to consumers. The appeal has since spread to other regions. Consumers gradually let go of the notion that volume equals value and came to recognize that compact products can offer equal or better value.

Evaluating products that are inherently mixtures of chemicals means that sooner or later one has to address the questions raised by the concept of 'mixture toxicity': what is the effect of the sum of all chemicals present in the environment? Are their effects additive? Synergistic? Antagonistic? Does it suffice to prove that each chemical alone is not expected to cause harm to the environment? The common approach in environmental risk assessment today is still to evaluate one chemical at a time, mostly because this is more straightforward, little is known about how to assess the toxicity of mixtures and old habits die hard. Additivity is a reasonable assumption if the modes of action of the ingredients are the same (i.e. narcotics) and there are no confounding factors (i.e. chemical reactions between the ingredients) [30,31]. The assessment is most straightforward for chemicals that are functionally and structurally similar and will therefore probably demonstrate additive effects. The environment, however, harbors a complex 'soup' of endogenous and man-made organic and inorganic chemicals. In the absence of scientifically validated approaches for the assessment of mixtures, additivity is assumed. The approach is conservative: it assumes that all chemicals have additive modes of action, whereas one would expect there to be a fair amount of antagonism. The criterion used here, namely that the sum of risk quotients of all ingredients should remain below 1 in order to demonstrate the safety of the product, is based on the assumption of additivity. Reassuringly, given the conservatism introduced by the assumption of additivity, the sum of the risk quotients for all products discussed here met that criterion.

6 Conclusion

Three conclusions can be drawn from this research:

1. Despite the very conservative nature of this risk assessment, all ingredients had a risk quotient well below 1 in both countries and at all times. Each individual risk assessment can actually be refined, as has been done in this article for the surfactants, but this is outside the scope of this article.
2. From 1988 to 1998, a decrease of more than 50% of risk quotients can be observed, due to a decrease in overall detergent consumption. This decrease has been made possible by the introduction of new technologies which are more weight-efficient, like compact and super compact detergents. Other factors like less pre-wash and better washing machine efficiency have certainly helped in reducing the detergent consumption. The development of compact and super compact detergents has been the result of many years of internal research trying to deliver a better product to consumers.
3. The difference observed in the distribution of risk quotients between countries is primarily due to the way the product is used and under which conditions (water hardness, water consumption, wash frequency, etc.), not to its chemical composition. Slight variations are observed from year to year due to a different balancing of ingredients; for example in 1998 where the Dutch product had a slightly lower risk quotient, due to a lower level of surfactant compared to the product sold in Sweden.

This analysis presents super compact detergents as a better alternative for the environment than the regular products manufactured and used in 1988. This conclusion may not apply to current regular detergents, manufactured and used in 2001, as they have also benefited from recent innovations. Also new product forms are now available on the market, like tablet and liquid pouch, which deliver new benefits like a better control of the dosage.

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