OECD Emission Scenario Document

Additives in the Rubber Industry

IC 15 ('others')

Exposure Assessment of the environmental
Releases of Chemicals in the Rubber Industry

1. version

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Contents

1.   Introduction

This document deals with the emission (release) estimation of additives used in the rubber industry (IC-15 'others'), including additives like vulcanising agents (UC-53), anti ageing agents, fillers (UC-20), pigments, stabilizer (UC-49), plasticisers, processing aids, flame retardants (UC-22).

Additives used in the rubber industry may enter the environmental compartments water, air, and soil. The major pathway is the entry into water. This occurs in those processes, where water is used as cooling or heating medium, in vulcanisation, and where water has direct contact with the rubber mixture. Releases to water can also occur in the cleaning steps throughout these processes.

2.   Life Cycle Steps and Emission Pathways

The production of synthetic "raw rubber" is subsumed under industrial category IC 11 (polymers industry) and not dealt with in this document. The raw material is provided by the chemical industry. Countries outside of the EU export the raw material for the production of natural rubber products. An additive used in rubber production passes a number of life-cycle steps. Each step has to be characterised in terms of its emissions to water, air and soil.
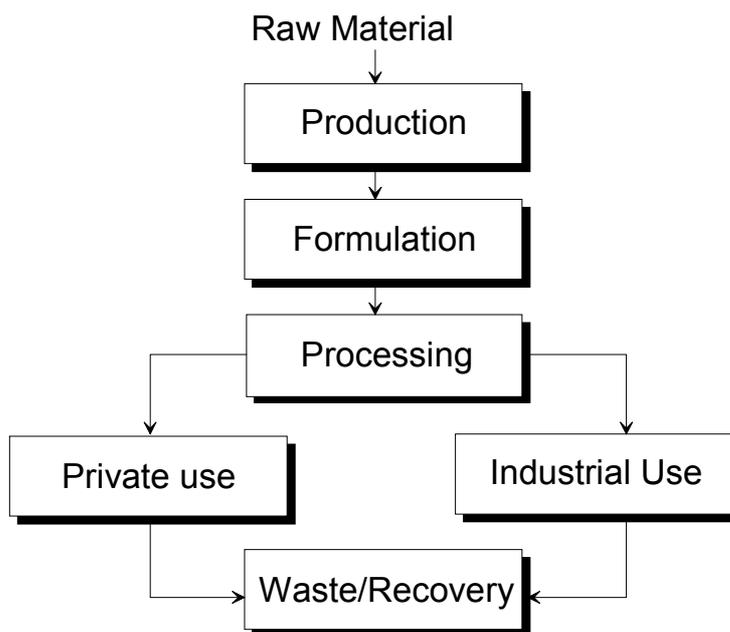
Figure 1:   Life Cycle Stages of Rubber Additives

The "production" of additives is not considered in this document. Any available production scenario can calculate the emissions. "Formulation" takes place in the chemical industry in the manufacturing of the rubber products and also in the rubber industry, where the formulation of the rubber mixture is performed.
The emissions during "processing" depend on the type and make-up of the additive and the processes used. The two steps "formulation" and "processing" can often not be separated and are therefore combined for the emission estimation. The processing steps taken into account are:

- the creation of the rubber mixture
- the mastication
- the shaping
- the vulcanisation
- the finishing.

The water pathway is the predominant one and is described in this document by a generic point source. For most additives, the air pathway is of minor importance.

A scenario of tyre abrasion describes the step „private use" with subsequent leaching into surface water and soil. The step "waste/recovery" means the leaching potential by rainwater to the soil and water compartment, while mainly volatile monomers and plasticisers escape to the air compartment. A specific waste disposal scenario is not covered in this document.

3.   Main Processes

The EU produces only synthetic rubber. The total production of synthetic rubber amounted to 2.51 million tonnes in the year 2001 (International Rubber Study Group 2003). This is about 25% of the world production. Unlike natural rubber, synthetic rubber is subject to strong fluctuations in the amount used and produced. This raw material was used to produce about 1 million tonnes of tyres and about 530.000 tonnes of rubber products ("technical and other elastomer products") in 1995. Table 1 shows the production of different EU countries, and Table 2 provides the market share of different synthetic rubber types characterised by the monomers used in the production. The world production of natural rubber grew from 3 million tonnes (1980) to 7.13 million tonnes in 2001 (International Rubber Study Group 2003).

Table 1   Production and Consumption of Rubber (1996)

Country	synthetic rubber production [1000 tonnes]	synthetic rubber consumption [1000 tonnes]	natural rubber consumption [1000 tonnes]
Spain	83.7	0	124.9
Italy	317.2	349	119.4
Turkey	40.1	0	0
France	565.8	403.6	209.7
Belgium	139.6	0	51.9
Germany	573.4	611.3	211.8
Netherlands	215.7	0	66.6
United Kingdom	306	268.9	140.4

Source: Rubber Statistics, Rubber Statistical Bulletin Vol. 51, No 10 (1997). http://www.rubberstudy.com - Statistics.

Table 2 Market Shares of different synthetic Rubber Types in the EU

Rubber type	Market share [%]
Styrene butadiene rubber (SBR)	58
butadiene rubber (BR)	14
ethylene propylene diene rubber (EPCM)	10
Isoprene rubber (IR)	1
Isoprene isobutylene rubber (IIR)	5
acrylonitrile butadiene rubber (NBR)	7
poly-2-chlorobutadiene (CR)	5

It is helpful to define three terms that are used in the rubber industry [Ullmann 1993]:

- **Rubber** (raw rubber) is a non-cross-linked, but cross-linkable (curable, vulcanisable) polymer with rubber-elastic properties at room temperature. At higher temperatures and/or, if exposed to deforming forces, rubber demonstrates an increasing tendency for viscous flow. Therefore, it can be formed under appropriate conditions.
- **Elastomers** are polymeric material that is cross-linked (vulcanised). They are hard and glassy at low temperature and do not exhibit viscous flow even at high temperature. Rubber elastic behaviour is characterised by a relatively low shear modulus with comparably low temperature dependency.
- **Vulcanisation** is a process in which rubber, through a change in its chemical structure (for example, cross-linking) is converted to a condition, in which the elastic properties are conferred or re-established or improved or extended over a greater range of temperature.

Natural rubber (NR) differs from synthetic rubber (SR). Natural rubber is made from the milk of the rubber tree. This rubber milk is also called latex and is a colloidal dispersion in an aqueous medium. The latex concentration after tapping is about 30%. Only a small percentage is used directly; the greatest part is processed into harder rubber. This is done through evaporation, hydro extracting, creaming and electro decanting. Thailand is the most important producer of natural rubber (1996). The raw rubber material is exported into the processing countries in the solid form.

Natural rubber demonstrates a balanced combination of physical attributes resulting in a broad range of possible uses (Ullmann 1993). A certain synthetic rubber surpasses every single attribute of a natural rubber. If these attributes are specifically required, the appropriate synthetic rubber will be used instead of the natural rubber.

The most important types of synthetic rubber are shown in Table 2 in relation to their market shares. Almost two thirds of the total rubbers currently used in industry are synthetic. These rubbers are made from monomers like butadiene, styrene, acrylonitrile and isobutene. Table 3 characterises the attributes of these rubbers. There are many more synthetic rubbers than shown in the Table. (Baumann and Ismeier 1998; Ullmann 1993).

Depending on the production procedure, one differentiates between polymerisates, polycondensates and polyaddition products. According to DIN ISO 1629 rubbers in the form of hard rubbers and latex are divided into groups depending on the chemical structure of the polymer chains. The rubber types are divided by their attributes (Lüpfert 1990) and their functions (Rentel et al. 1991). A rubber is made mainly of (Lüpfert 1991)

- polymers (elastomers)
- fillers
- plasticisers
- anti ageing agents
- linking systems.

The chemicals and raw rubbers used in rubber production pass through all the steps listed in Figure 1. Additives are added before and during processing. After the initial composition the mixture is created with an internal mixer, a rolling mill, a batch-off-facility and an extruder. The shaping of the semi-finished product is made with an extruder, a plunger type injection moulder, through calendaring, the spreading machine and assembly.

The compression process, the transfer moulding, and injection moulding are used for vulcanisation (curing) in presses. A continuous vulcanisation is made with the rotational vulcanisation, the steam pipe vulcanisation, the hot-air vulcanisation with microwave preheating, the boiling bed vulcanisation and the salt-bath vulcanisation. In addition to these, vulcanisation can also be accomplished in an autoclave.

Table 3 Rubber Type and Additives

Abbreviation	Rubber Type	Mixture
NR	**Natural Rubber**	
	proteins aliphatic acids resins ash sugar Water	(cis-1,4-polyisoprene) vulcanisation agents (sulfur) vulcanisation accelerators anti ageing agents fillers plasticisers processing aids
SR	**Synthetic Rubber**	
SBR	styrene butadiene rubber	monomers (1,3-butadiene, styrene) vulcanisation agents (sulfur) vulcanisation accelerators anti ageing agents fillers plasticisers factices resins processing aids
BR	butadiene rubber	monomers (1,3-butadiene) vulcanisation agents (sulfur) vulcanisation accelerators anti ageing agents fillers plasticisers processing aids
NBR	Acrylonitrile butadiene rubber	monomers (1,3-butadiene, acrylonitrile) vulcanisation agents (sulfur) vulcanisation accelerators anti ageing agents fillers plasticisers factices processing aids
CR	poly-2-chlorobutadiene (polychloroprene)	monomers (chloroprene) vulcanisation agents (sulfur) vulcanisation accelerators anti ageing agents fillers plasticisers factices processing aids
SR	**Synthetic Rubber**	
EPDM	ethylene propylene diene rubber	monomers (ethylene, propylene) vulcanisation agents vulcanisation accelerators anti ageing agents fillers

Abbreviation	Rubber Type	Mixture
		plasticisers processing aids
IR	isoprene rubber	monomers (isoprene) vulcanisation agents vulcanisation accelerators anti ageing agents fillers plasticisers processing aids
IIR	isoprene isobutylene rubber	monomers (isoprene, isobutylene) vulcanisation agents vulcanisation accelerators vulcanisation activators anti ageing agents fillers plasticisers processing aids
OTHER	special	monomers vulcanisation agents vulcanisation accelerators anti ageing agents fillers plasticisers processing aids

The following Table 4 presents different rubber products and their percentages of uses. This distribution does not fluctuate over the years; it is assumed to be valid for all EU countries.

Table 4 Distribution of Rubber Products

Rubber products	Percentage [%]
form products, rubber-metal-compounds	24
technical tubes and profiles	23
glues, mixture, repair material	13
other rubber products	11
foam, micro cellular and cellular rubber products	10
conveyor belts, flat and vee-belts	9
other technical products	5
sole material	3
sheet material, punching products	2
Sum	100

Source: Association of the German Rubber Industry 1990: leaflet

3.1 Natural- and synthetic Rubber

3.1.1 Mastication and creation of mixtures

The mastication is a process, in which the length of the polymer chains is decreased to lower the viscosity. This is necessary to get a better workability of the rubber (extrusion, calender and others) and to spread the rubber chemicals throughout the matrix. Rubber additives like

anti ageing agents, fillers, plasticisers are discussed in Subsection 3.4. Their functions are shown in Table 5.

An optimised viscosity is especially important for the production of rubber blends, since a homogenous distribution of the constituents is only possible within an equal viscosity. Unlike synthetic rubber, which can often be processed without a previous mastication, natural rubber is usually offered as a relatively hard product with little plasticity, making mastication a requirement.

The mastication can be achieved in a purely mechanical process, in a mechanic-thermal process, physically by adding plasticisers and/or processing agents as well as chemically. Machines like rolling mills, the internal mixer or the gordon-plasticator masticate the rubber substance but also serve as mixers, for cooling, to finish the mixing of half ready rubber mixtures and to preheat finished rubber mixtures. A possible contamination of the water compartment during the mixing process can happen through the cooling process, if water is used as a direct cooling agent. Finally sulfur is added to the mixture. The finished mixture is taken off as a rough sheet. This rough sheet is cooled by immersion in water to which an anti tack-agent has been added (rolling mill). Especially in larger companies, the internal mixer replaced the rolling mill. An internal mixer can hold 15 to 650 litres. For the homogenisation, cooling and working into sheets rolling mills are still used. In modern installations, a batch off machine is responsible for taking off the sheets and immersing them in a bath of water or a bath with an anti tack-agent.

3.1.2 Shaping

Rubber mixtures are usually not immediately reprocessed after the mixture was created, but are stored in a cool and dry environment. For the shaping, processes like extrusion, calendering, form moulding and transfer moulding are used.

Injection machines (extrusion plants) form rubber mixtures into bars, tubes, profiles, treads of tyres or sheets or encase cables and wires. In a continuous process, the rubber mixture is cured immediately after extrusion. If reprocessing as a semi-finished product is planned, the mixture is dusted and drawn through an adherent bath. Calendering is used to create sheets for the rubberisation of technical weaves (fractioning) and to coat weaves with a thin rubber sheet (skimming).

Formed articles are mass articles like stoppers, bottle caps, soles, packing rings, collars, membranes, valve balls, buffers and tyres. Processes like pressing, transfer moulding, flashless-processes and the injection-moulding process are characterised by shaping and vulcanisation in one step. An assembly line takes an important role in this context.

After the forms were used in a transfer moulding process over time, organic residues from the rubber mixture or from the anti tack-agents deposit as incrustation or carbonisation. A chrome sulfur bath (60°C) is used to clean the forms. The actual cleaning takes place in a lapping machine with a suspension of quartz flour. Water is used for rinsing the cleaned forms.

3.1.3 Vulcanisation / Curing

Vulcanisation is a chemical process to link the polymer structure. The term vulcanisation is used for a linking procedure, during which the macromolecules of the rubber are linked to each other over linking bridges. The linking is achieved with vulcanisation agents, usually sulfur, sulfur producers or peroxides as well as energy rich radiation.

If uncured, rubber is thermoplastic. The molecules are not connected to each other. During vulcanisation the rubber changes from the thermoplastic to the elastic condition. The number of links depends on the amount of linking substances, their activity and reaction time. The following procedures are used for vulcanisation:

- liquid curing method (LCM-Vulcanisation)

- boiling bed vulcanisation
- hot air vulcanisation after UHF-preheating
- vulcanisation in hot steam under high voltage (under lead).

The LCM-vulcanisation is commonly used for extruded profiles, tubes and similar processes. It is continuous and takes place in a liquid bath. The duration of the bath depends on the vulcanisation speed, the injection performance of the extrusion plant, the temperature of the heating bath and other factors. Eutectic salt mixtures (as well as glycerin, silicon oils, polyalkyleneglycole, metal mixtures) are used as a heating medium. For the boiling bed vulcanisation, small glass beads are used through which gas flows to lift them.

The HF vulcanisation uses a pressure-free continuous process. The vulcanisation in steam pipes is used almost exclusively in the cable industry. Immediately after coating, the cable is guided into a pipe, in which the steam pressure is usually between 0,5 and 1,2 MPa (max. 2 MPa). To reduce deformations, the vulcanisation is sometimes performed under a lead coating, this coating protects from the hydrolytical effect of the steam. After cooling with water, the lead coat is removed.

The vulcanisation of calendared products can be achieved with rotational vulcanisation or in vulcanisation presses. A high frequency preheating is possible. Assembled rubber products are vulcanised free (not in a mould) in an open heater (hot air vulcanisation).

To decrease the vulcanisation time, a steam heater is used. When using saturated vapour, in which water and steam are in balance, the temperature transference is more even. To avoid condensation spots, spreading agents are used. When vulcanising in water, the temperature transfer is extremely good compared to a medium of air or steam. This process will be used for big parts. It is used rarely for example for the vulcanisation of large rolls due to the complexity of the process. Vulcanisation can also be achieved through cold vulcanisation or energy rich radiation.

3.2   Latex

Natural latexes and synthetic latexes (as are created during the emulsion polymerisation) are processed, among other things into thin-walled products. The mixture is prepared by kneading the required chemicals into the latex solution. The coagulation process is used to create thick-walled products. The form is submerged in an alcoholic solution of a coagulation substance. This results in a thicker coagulum on the form as in other submerging processes. The vulcanised products must then be thoroughly rinsed with water to remove the coagulation substance.

For foam rubber, steam or hot water is used as a medium for the vulcanisation. The spinning process for the creation of latex strings also requires water to rinse out the coagulator. Products made from unwoven textiles are made from bonded fabrics impregnated with a latex mixture. The unused adhesive is pressed out and the damp fabric dried and vulcanised. Leftover chemicals (emulsifying agents, stabilisers) are removed by washing.

3.3   Rubber-Metal

Metal working takes place in the rubber industry wherever composite parts made of rubber and metal are needed. One example is the tyre, which, in addition to a multitude of rubber mixtures, includes stabilising material like steel, textiles or plastic. Other rubber-metal parts for the automobile industry are shock absorbers, elastomere springs, coupling parts, hangers and others. Rubberised parts, like rolls are used in other areas. For the preparation of composite parts, spray- and degreasing plants are used.

3.4   Chemicals and Additives

The rubber industry uses hundreds of different substances. The finished product gains the required attributes only by adding fillers, plasticisers, vulcanisation chemicals, anti ageing agents and adhesives. Table 5 lists the most important process steps for the production of rubber and the most important substance classes used in each step.

The rubber industry uses a special unit for expressing the components of a rubber mixture. They are presented in relation to 100 mass parts total rubber (parts per hundred rubber [phr]) or in wt-% (cf. Table 9).

Table 5 Function of Additives in Rubber Processing Steps

Process Step / Additive	Description	Function of the Chemicals used
Mastication	plastification of the rubber before working the molecular chain splitting through oxidative breakdown (usually aromatic or heterocyclic mercaptan). By using activators, the mastication can be shifted to a lower temperature spectrum (inner complex salts).	1. mastication agents 2. activators (Boosters)
Vulcanisation	For linking the rubber molecules And creating linking bridges (usually sulfur,	Vulcanisation agents Vulcanisation accelerators Vulcanisation activators
Vulcanisation	Sulfur producers, peroxides)	Vulcanisation retarders
Anti-ageing agents	finished product with protection against damaging influences.	1. antioxidants
		2. anti fatigue agents 3. anti ozonants 4. light protection agents 5. anti hydrolysis agents 6. agents against metal poisoning 7. desactivators 8. reversion protection agents 9. anti cyclisation agents
Fillers	substances which are insoluble towards the rubber matrix, non-volatile and inert towards the polymer. The fillers define the attribute of the vulcanisate. They extend and strengthen the vulcanisate.	inorganic fillers organic fillers
Plasticisers	plasticisers serve as extensions, they improve the fluidity, they distribute the fillers, improve the workability and the adhesiveness. They influence the physical attributes of the rubber like expansion and elasticity. Primary plasticisers have a loosening effect and lower the viscosity of non-vulcanised rubber mixtures. Secondary plasticisers serve as lubricants and improve the shapeability.	Plasticisers (categories: mineral oils, natural substances, synthetic plasticisers)

Pigments/ Colourants	pigments are in rubber de facto insoluble, organic or inorganic, coloured or white substances.	1. inorganic pigments:
		    a) white pigments
		    b) coloured pigments
		2. organic pigments

Processing aids	processing aids ease the working of the material: 1. lubricants are used mainly in the extrusion and the injection moulding processes 2. tackifiers serve to increase the assembly adhesion	1. lubricants − Metal soaps (like zinc-, calcium-soaps) − Aliphatic acids, Aliphatic acid ester 2. tackifier
Processing aids	3. among others to improve the stability, the injectability and calendrability	3. factice

Process Step / Additive	Description	Function of the Chemicals used
Processing aids	4. by using fillers activators, the activity of the fillers can be improved	4. fillers activators, adhesion agents

	5. blowing agents are used to produce porous rubber articles (decompose under gas splitting)
	6. bonding agents increase the adhesion of rubber on strengthening material (weaves, metal)
		5. blowing agents
		    α) inorganic
		    β) organic
		6. bonding agents

other agents	1. anticyclisation agents serve to prevent unwanted cyclisation during mastication 2. with the aid of replastication agents slightly vulcanized mixtures can be returned to a workable condition (desulfuring of the rubber mixture) 3. emulsifier disperse systems 4. flame retardant lower the flammability of the rubber 5. in solvent adhesives, in solvent dipping processes, for cleaning 6. for refining the surface 7. to increase the hardness of the vulcanisate 8. to nuance, neutralize and cover the rubber smell 9. to increase the electronic conductibility 10. to kill microorganisms 11. to prevent the spreading of germs	1. anti cyclisation agents 2. replastication agents 3. emulsifier 4. flame retardant 5. solvents 6. surface treatment agents 7. hardeners 8. odorants 9. antistatic agents 10. microbiocides 11. anti termite agents
Latex-chemicals	to add pulverised additives into the aqueous colloidal latex Solution	1. dispersion agents 2. emulsifier 3. stabiliser 4. wetting and foaming agents 5. foam stabilisers 6. thickeners 7. coagulating agents 8. preservatives 9. vulcanisation chemicals
Latex-chemicals		10. anti ageing agents 11. fillers 12. plasticisers
Release agents	1. prevent non-vulcanised rubber mixtures from adhering during storage 2. prevent rubber from sticking to the forming tools 3. release agents are added to the rubber mixture, migrate to the surface and improve the removability of the vulcanisate from the form	1. release agents for non-vulcanised rubber mixtures (anti tack agents) 2. mould release agents 3. mandrel release agents

4.   Emission Scenarios

This document describes three generic emission scenarios:

- formulation and processing: emissions to wastewater
- formulation and processing: emissions to air and soil
- private use of rubber products by tyre abrasion: emissions to surface water.

The contamination of water in the rubber industry takes place in several areas of the rubber production, whenever water comes into direct contact with the rubber product. This happens during cooling-, heating-, vulcanisation- and cleaning operations. Examples are direct cooling during the extrusion or direct contact of the material vulcanised with the steam in the vulcanisation processes.

The sources and possible contaminations of wastewater are presented in Table 6. The water can contain zinc (zinc compounds are often used in anti tack agents), cadmium, copper and lead from vulcanisation. Storing, weighing, mixing are other possible sources of exposure. By using wastewater free procedures (indirect cooling, circulation systems, dry cleaning) a releases to water can be avoided. A cascading arrangement of the rinsing baths and an optimised water flow can also decrease the volume of wastewater. The remaining wastewater is treated, if the pollutant concentration makes it necessary. Degreasing is accomplished with chlorinated hydrocarbons and complex builders like EDTA. The reduction or prevention of hazardous substances from the cleaning operations can be accomplished by degreasing on an aqueous basis.

Table 6   **Water Contamination of different Sources in the Rubber Industry [in mg L^{-1}] (Association of the German Rubber Industry 1999)** [1]

Source	COD[2]	C[3]	SLS[4]	AOX[5]	BTX[6]	NO$_2$	P$_{total}$	Pb	Cu	Zn	Cd
direct cooling water and anti tack agent contact after mixing	3000 (RP)						1.5 (RP)			0.54 (TI) 1.0 (RP)	<0,005 (TI)
direct cooling water after extrusion	5 (RP)						<0.05 (RP)			0.06 0.12 0.052 0.084 (TI) <0.1 (RP)	<0,001 (TI) <0,005 (TI) <0.02 (RP)
wastewater from the waste air treatment during the production and processing of solutions after pretreatment (stripping)	ca. 6000 35000 I 1000 (RP)		0.1 O (RP)	0.03-0.2 (RP)			0.1 I 8.1 O				
wastewater from vulcanisation in forms as well as subsequent washing and cleaning processes	282 (TI)							0.065 (TI)	0.14 (TI)	6.8 (TI)	

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[1] This table gives an overview about individual situations of special rubber manufacturers. It is not a generalisation of the emission situation.
[2] Chemical oxygen demand
[3] Hydrocarbons
[4] Semivolatile lipophilic substance
[5] Adsorbable organic halogen compounds
[6] Benzene, Toluene, Xylene

Source	COD[2]	C[3]	SLS[4]	AOX[5]	BTX[6]	NO$_2$	P$_{total}$	Pb	Cu	Zn	Cd
wastewater from vulcanisation in autoclaves as well as subsequent washing and cleaning processes	65 (RP) 1100 (RP)	301 (RP)	233 (RP)	0.23 (RP)	0.05 (RP)		3.3 (TI) 0.2 (RP)	<0.03 (TI) 0.16 (RP)	0.02 (RP) 0.05 (RP)	0.22 (RP) 0.87 (RP)	<0,002 (RP) ca. 0.05 (RP)
wastewater from vulcanisation in a salt bath as well as subsequent washing and cleaning processes						1500 (RP)					
wastewater from general cleaning of production tools	540 (TI) 2000 I 1200 O (RP)	87 (TI) 0.05 O (RP)	28 O (RP)	80 (TI) >3.0 I <1.0 O (RP)			<1 (TI) 41 I 6.6 O	5.8 I 0.01 O (RP)	2.2 I 0.02 O (RP)	38.5 I 0.2 O (RP)	
	I: inflow, O: outflow of the waste water treatment plant TI = type industry, RP = rubber products										

4.1 Definition of the generic Point Source

The generic point source for the rubber industry derived in this sub-chapter is related to the German situation. However, it is believed that the size for a typical manufacturer in the EU Member states is comparable, because of the general structure of the rubber industry. This generic point source is the reasonable worst case used in the environmental exposure assessment. Similar considerations can be followed in deriving the generic point source in non-EU Member states of the OECD.

The emissions of the generic point source are derived from a survey of the wastewater discharges in the rubber industry. This total wastewater stream can be divided into the following component waste streams: sanitary, rainwater, metalworking, rubber working and others. The total quantities discharged each day are shown in Figure 2. The Association of the German Rubber Industry (1999) provided these discharge volumes and the following assumptions.
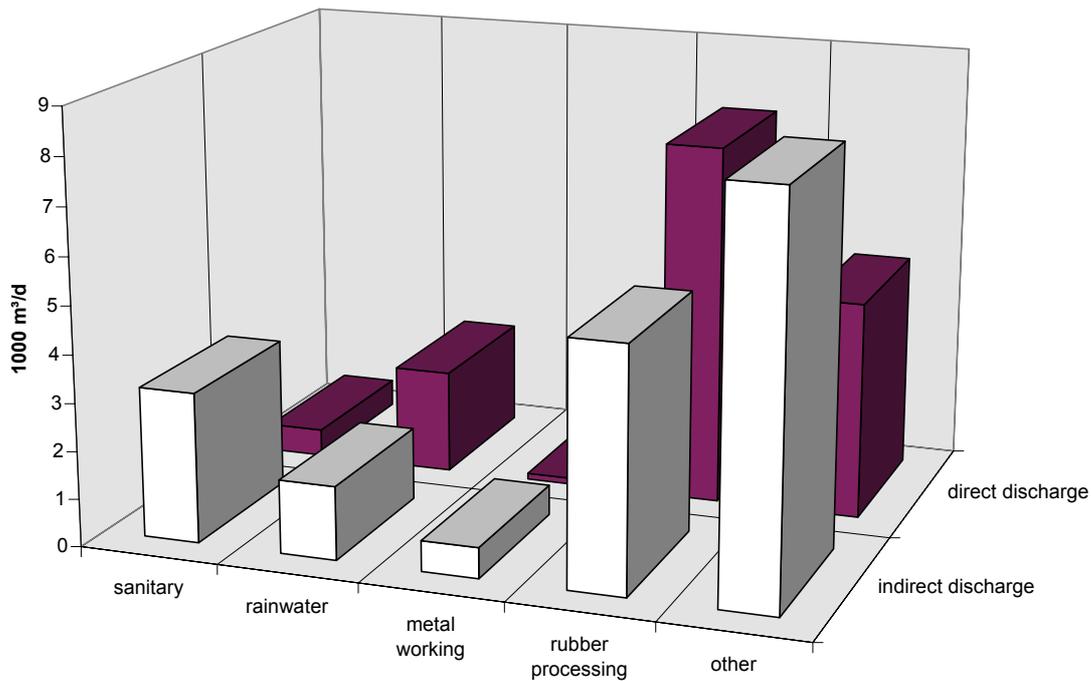
Figure 2: **Volume of Component Wastewater Streams of direct and indirect Discharge for Members of the Association of the German Rubber Industry (1999)**

The Association of the German Rubber Industry (1999) recorded these wastewater data for 18 direct dischargers (plant specific sewage treatment plant) and 70 indirect dischargers (via the municipal sewage treatment plant), 88 companies in total. It should be noted, that three companies discharged 75% of the total volume directly, and one indirect discharger made up for 30% of the corresponding discharge. This wastewater volume covers not only the rubber processing, but also the sanitary, metalworking and other areas. It is assumed that 37 % of the wastewater is attributed to rubber processing. Table 7 lists the statistical wastewater data for determining the generic point source.

Table 7 Statistical Data for Determining the generic Point Source

Quantity of Wastewater (1)	Flow rate [$m^3.d^{-1}$]
Q_{wwat} (arithmetic average of the waste water amount)	184
Q_{wwat} (minimum)	10
Q_{wwat} (maximum)	1154
Q_{wwat} (90-percentile)	438

(1) Sample size of 84 companies

Direct and indirect discharges were combined. The EU standard sewage treatment plant with a flow rate of 2000 $m^3.d^{-1}$ was used to eliminate from this sample those companies with a higher flow rate. 84 companies were below this threshold and discharged in sum about 15000 $m^3.d^{-1}$. Four very large companies that were above this threshold and discharged in sum also about 15000 $m^3.d^{-1}$. These four companies were eliminated in the evaluation of the 90 percentile. The 90 percentile value is 438 $m^3.d^{-1}$.

It was further assumed that about 3 m³ water is needed to produce one tonne of rubber goods. This water consumption accounts only for the rubber production.

With the assumption that the water consumption of the rubber production is 37% of the value for the 90-percentile of the wastewater volume (Table 7), a daily generic production (Q_{prod}) of about 55 tonnes according to the following calculation is derived as the reasonable worst case generic point source:

$$Q_{prod} \text{ (generic point source)} [t.d^{-1}] = Q_{wwat} \text{ (90 percentile)} [m^3.d^{-1}] * 0.37 / 3 [m^3.t^{-1}]$$
$$Q_{prod} \text{ (generic point source)} [t.d^{-1}] = 438 \ m^3.d^{-1} * 0.37 / 3 \ m^3.t^{-1} = 55 \ t.d^{-1}$$

A further assumption is that the production ratio for tyres : rubber products is 60 : 40. This assumption is considered in Table 8. The quantities compiled in this table are also used in example calculations (Section 5).

Table 8 Product Type and Amount produced at the generic Point Source

Product type	Amount $Q_{prod} [kg.d^{-1}]$	
Tyres	**33000**	
tyres total	26400	
car tyres		11600
Truck tyres		14800
re-treading	6600	
rubber products	**22000**	
technical tubes, profiles		5320
form products, rubber-metal compounds		5190
glues, mixtures, repair materials		2840
foam-, micro cellular- and cellular rubber products		2110
conveyor belts, flat- and vee belts		1940
sole material		550
sheet material, punching products		460
other technical products		1150
other rubber products		2440
Sum	**55000**	

Source: Association of the German Rubber Industry (1999)

4.2 *Emission Scenario for Formulation and Processing to Wastewater*

For the exposure calculation of the emissions to wastewater the following information is needed:
- function of the additive (Table 5)
- amount of product type produced per day [kg/d]. The default values for tyres and rubber products and their sub-divisions are presented in Table 8.
- the parts of the additive introduced as parts of 100 rubber. This figure is either given in weight percent [wt-%] or in parts per hundred rubber [phr]. The latter

figure is a special term used in the rubber industry. This figure can either be delivered by the applicant (A) or can be taken as default (D) from Table 9.
- fraction of additive remaining in the rubber product. This figure can either be provided by the applicant or be taken as default from Table 9.

The emission rate [kg/d] into wastewater is calculated from equation (1) as shown:

$$Elocal_{water} = Q_{prod} \cdot \frac{Q_{additive}}{100 \cdot F_{recipe}} \cdot \left(1 - F_{remaining}\right) \qquad \textbf{(1)}$$

Parameter:

Explanation	Symbol	Value	Unit	Origin [1]	Remarks
Input Amount of the product type produced per day	Q_{prod}		[kg.d^{-1}]	D	see Table 8
Parts of additive introduced per parts of 100 rubber	$Q_{additive}$		[wt-%] or [phr]	A/D	from applicant or Table 9
Recipe factor	F_{recipe}	2 resp. 1		A	2, when content is given in phr 1, when content is given in wt-% (seeTable 10)
Fraction of additive remaining in the rubber product	$F_{remaining}$		[-]	A/D	from applicant or Table 9
Output Emission of substance to waste water treatment plant	$Elocal_{water}$		[kg.d^{-1}]	O	

(1) A = based on information from applicant; D = default, O = Output

Table 9 lists for various additives the content in rubber products and tyres mostly expressed in parts per 100 rubber parts [phr]. For worst case calculations the upper range is used. If the content of the additive in the mixture is not specified, estimation can be used in the overall class (the gray field). The last column presents the fraction of the additive assumed to remain in the rubber product and therefore not being subject to emissions during processing.
These figures should not be seen as a possible recipe. The actual amount used depends on the function of the mixture and can vary within the given ranges. A tyre for example is composed of 10 different rubber mixtures and is made up of 48 wt-% natural and synthetic rubber, 23 wt-% carbon black, 18 wt-% steel, 3 wt-% woven glass fabrics and 8 wt-% additives.

Table 9 Functions and Content of Rubber Additives

Rubber additive	Use in rubber products [phr]	Use in tyres [phr]	$F_{remaining}$
mastication agents / peptiser	0.1-0.5 wt-% (NR), 1-3 wt-% (SR)	0.5	0.995
Activator	no data	4	
vulcanisation agents	0.25-5 (soft) 25-40 (hard)	up to 2	1.0
sulfur-containing cross-linking agents	2	2	

Rubber additive	Use in rubber products [phr]	Use in tyres [phr]	$F_{remaining}$
vulcanisation accelerator	0.1-2	0.2-2.0	
accelerator activator	1.5-5	1.3	
sulfur-free cross-linking agents	0.001-1	no data	
coagents for sulfur-free cross-linking agents	0.5-2.0	2.0	
other cross-linking agents	0.001-2.0	0.001-2.0	
vulcanisation retarders	1.5-2	0.3-2	
scorch inhibitors	0.1-1	0.5	
anti ageing and antiflex-cracking agents / antidegradants	0.8	0.8	0.98 (RP) 0.99 (TI)
Antioxidants	0-3	0.25	
antifatigue agents	1-5	0.2	
Antiozonants	1-7	0.2-1	
light protection agents	0-3	0.2	
anti hydrolysis agents	0.5-3	0,2	
heat protection agents	0.5-3	1	
agents against metal poisoning	0.5-3	0.2	
Deactivators	0.5	0.3	
reversion protection agents	0.5-3	no data	
anticyclisation agents	0.5	no data	
Quencher	0.5	no data	
other anti ageing agents	1.5-3.0	no data	
fillers and pigments[7][8]	Up to 30	up to 40	0.99
Fillers	10-30	40	
Pigments	1-5	no data	
Plasticiser	Up to 20	up to 3.8	0.95[9]

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[7] After formulation, fillers are not removed from the rubber matrix.
[8] Some fillers act as pigments and vice versa.
[9] Plasticisers do not enter into a chemical bonding with the rubber, migration to the surface is possible.

Rubber additive	Use in rubber products [phr]	Use in tyres [phr]	$F_{remaining}$
plasticiser, natural	10-20	2.4-3.8	
plasticiser, synthetic	10-20	2.4-3.8	
Processing aids	Up to 15	1-5	0.995
lubricants and flow improvers	5-15	no data	
Tackifier	5-15	no data	
Factices	5-15	no data	
filler activator	5-15	no data	
blowing agents	5-10	3	1.0(t)
bonding agents	1-4	3-4	1,0(t)
Stabiliser	1,5-3	1,5-3	
other agents	0.1-3	0.1-3	0.95
anticyclisation agents	No data	no data	
replastication agents	No data	no data	
Emulsifier	0.1-0.5	No data	
flame retardant	2-3	No data	
Solvents	No data	no data	
surface treatment agent	2-43	2	
Hardeners	No data	no data	
odour agents	0.25-0.1	no data	
Antistatic agents	0.1-0.6	5	
Microbiocides	No data	no data	
Antitermite agents	No data	no data	
Reinforcing agent	No data	no data	
Homogeniser	No data	no data	
latex-chemicals	Up to 5		0.95(RP)
dispersion agents	1-2		

Rubber additive	Use in rubber products [phr]	Use in tyres [phr]	$F_{remaining}$
Emulsifier	1-5		
Stabilizer	1-2		
wetting and foaming agents	1-2		
foam stabilizers	1-5		
Thickeners	No data		
coagulation agents	No data		
Preservatives	No data		
vulcanisation chemicals	0.5-2		
anti ageing chemicals	No data		
Fillers	No data		
Plasticisers	No data		
release agents	[10] up to 30	up to 30	0.95
..for non-vulcanised rubber	0.5-5	0.5-5	
mould release agents	5-30	5-30	
mandrel release agents	5-30	5-30	
Others	Up to 5	up to 5	0.95
cleaning agents	No data	no data	
other rubber chemicals	No data	no data	

An example for a recipe is presented to demonstrate the introduction of the F_{recipe} -factor for information about the composition in phr.

Table 10   Example for a Recipe (Sablowski Datacom 1996)

Compound constituent	Recipe [phr]	Recipe [wt-%]
Synthetic rubber butadiene (SBR)	80.28	43.63
Butadiene rubber (BR)	19.72	10.72
fatty acid	1.97	1.07
Plasticiser	11.97	6.50
Waxes	1.69	0.92
zinc oxide	3.10	1.68
carbon black	60.56	32.91
anti ageing agent	1.69	0.92

[10] These substances are not a part of the rubber mixture, but will be treated so

Accelerator	1.20	0.65
Sulfur	1.83	0.99
Sum	184.01	99.99

4.3 Emission Scenarios for Formulation and Processing to Air and Soil

Hardly any monitoring data for air and soil emissions of rubber additives are available. A simple emission scenario based on the A-Tables (emission factors) of the European Union Technical Guidelines (TGD 2002) from IC-11 (polymer industry) life cycle step "polymer processing" is presented here.

Table 11   Emission Factors (TGD 2002, A-Table for IC-11 "polymer processing")

Type of additive	Conditions		Emission factor
Air			
	Vapour pressure [Pa]	Boiling point [°C]	
Additives	< 1	< 300 / unknown	0.001
Pigments		> 300	0.0005
Colorants	1-100	< 300 / unknown	0.0025
Fillers		> 300	0.001
Processing aids	>100	< 300 / unknown	0.01
		> 300	0.005
Plasticisers		< 400 / unknown	0.01
		> 400	0.005
Vulcanising agents	<100		0.075
	100-1000		0.15
	1000-10000		0.25
	>10000		0.35
Soil			
Additives			0.0001
Pigments			
Colorants			
Fillers			
Processing aids			0.0005
Vulcanising agents			0.0001

The emission rate [kg/d] into air resp. soil is calculated from equation (2) as follows:

$$Elocal_{air} = Q_{prod} \cdot \frac{Q_{additive}}{100 \cdot F_{recipe}} \cdot F_{air} \qquad\qquad \textbf{(2)}$$

Parameter:

Explanation	Symbol	Value	Unit [1]	Origin [2]	Remarks
Input					
Amount of the product type produced per day	Q_{prod}		[kg.d^{-1}]	D	see Table 8
Parts of additive introduced per parts of 100 rubber	$Q_{additive}$		wt-% or [phr]	A/D	from applicant or Table 9
Recipe factor	F_{recipe}	2 resp. 1	[--]	A	2, when content is given in phr 1, when content is given in wt-% (see Table 10)
Emission factor to air resp.	F_{air}		[--]	A/D	from applicant or Table 11
Emission factor to soil	F_{soil}				from applicant or Table 11
Output	$Elocal_{air}$		[kg.d^{-1}]	O	
Emission of additive to air resp. To soil	$Elocal_{soil}$				

(1) A = based on information from applicant; D = default, O = Output

4.4 Emission Scenario for Use of Rubber Products by Tyre Abrasion

The use of tyres for motor vehicles produces tyre abrasion that is deposited as particles on streets and roads and on nearby soil (Baumann and Ismeier 1998, p. 307-353). These particles are exposed to short-range wind transport and to weathering so that hydrolysis, leaching, and biodegradation of constituents take place. Photo transformation by light is unlikely to play a major role on degradation. The following persistent leaching products were identified, when tyres were kept in water (pH 7) or acid rain water (pH 4) for eleven months [Baumann and Ismeier 1998, p. 341) (Table 12).

Table 12   Laboratory Experiments: Long-term Leaching Products of Tyres in Distilled and Acid Rain Water (Baumann and Ismeier 1998, p. 341)

Substance	Abbreviation	Concentration [mg/L]
2-Mercaptobenzothiazol	MBT	< 0.5
Benzothiazole	BT	15 – 1972
2-Methylbenzothiazol	MeBT	< 2
2-Methylthiobenzothiazol	MeSBT	< 2
Aniline		5.9 – 294
Dicyclohexylamine		< 35 – 218
Cyclohexylamine		< 1 – 423

These same substances were identified in monitoring of street run-off water. These monitoring experiments (Baumann and Ismeier 1998; p. 342 - 343; Schmegel 1995, p. 75; Spies et al. 1987; Kumata et al. 2000) show that tyre abrasion contributes significantly to run-off releases of persistent breakdown products of rubber additives, e.g. benzothiazole and methylated benzothiazole.

An approach to calculate the annual emission rate [kg/a] of a rubber additive and its breakdown products via tyre abrasion is presented in equation (3).

$$Eregional = Q_{ann\_prod} * F_{abrasion} * F_{mol\_weight} \qquad \textbf{(3)}$$

Parameter:

Explanation	Symbol	Value	Unit	Origin	Remarks
Input Amount of the additive produced per year in defined region	Q_{ann_prod}		[kg.a^{-1}]	A	The region needs to be defined.
Emission factor for abrasion from tyres	$F_{abrasion}$	0.12	[--]	D	Hartung and Koch 1989 p. 33 Ahlers et al. 1996, p. 73
Fraction of molecular weight for additive and breakdown product	F_{mol_weight}		[--]	O	If breakdown product is considered otherwise F_{mol_weight} = 1
Output Emission rate of additive or break-down product from tyre abrasion in defined region	$Eregional$		[kg.a^{-1}]	O	

The regional emission rate can be the input parameter in any Mackay level III fugacity model (e.g. SimpleBox) for calculating the regional concentrations in surface water and natural soil.

4.5   Emissions from Leachates of Landfill

Considerable amounts of used tyres (about 30%) are disposed off in landfills. Rubber additives may be found in the leachates of the landfill. It is at present difficult to quantify such leachates.

5.   Examples of Emission Estimation

5.1   Emissions from Formulation and Processing to Wastewater

A   Calculation of the emissions of a plasticiser
Additive in the manufacturing of a tyre; emission occurs during cooling after extrusion (equation 1).

Input Data

Q_{prod}	=	33000	kg.d^{-1}
$Q_{additive}$	=	3.8	phr
F_{recipe}	=	2	-
$F_{remaining}$ =		0.95	-

Output Data

$Elocal_{water}$	=	31.35	kg.d^{-1}

B   Calculation of the emissions of an antioxidant
Additive in the manufacturing of a rubber product (equation 1).

Input Data

Q_{prod}	=	22000	kg.d^{-1}
$Q_{additive}$	=	0.4	phr
F_{recipe}	=	2	-
$F_{remaining}$ =		0.99	-

Output Data

$Elocal_{water}$	=	0.4	kg.d^{-1}

C   Calculation of the emissions of an anti-ageing agent

Additive in the manufacturing of a truck tyre (equation 1).

Input Data

Q_{prod}	=	14800	kg.d^{-1}
$Q_{addtivie}$	=	0.5	wt-%
F_{recipe}	=	1	-
$F_{remaining}$	=	0.99	-

Output Data

$Elocal_{water}$	=	0.7	kg.d^{-1}

Example for the determination of the Clocal$_{water}$ with the data of example B

Additive in the rubber industry

Calculation of PEClocal for the aquatic compartment

Chemical: Example:

Amount of the product produced per day Q_{prod} = 22000 kg.d^{-1}
(from Table 8):

Parts of additive introduced in $Q_{additive}$ = 0.4 phr
parts of 100 rubber:

Recipe factor: F_{recipe} = 2

Fraction of additive remaining in the product: $F_{remaining}$ = 0.99

Fraction of emission directed to water: $Fstp_{water}$ = 100 %
(SimpleTreat Model)

Capacity of sewage treatment plant (STP) (TGD):   EFFLUENTstp = 2000 m^3.d^{-1}

Dilution factor (TGD):   DILUTION = 10

Factor (1+Kp*SUSPwater):   FACTOR = 1

Emission per day:

$$Elocal_{water} = Q_{prod} * \frac{Q_{additive} * (1 - F_{remaining})}{100 * F_{recipe}}$$ $Elocal_{water}$ = 0.44 kg.d^{-1}

Influent concentration:

$$Clocal_{inf} = \frac{Elocal_{water}}{EFFLUENTstp}$$ $Clocal_{inf}$ = 0.22 mg.L^{-1}

Effluent concentration:

$$Clocal_{eff} = Clocal_{inf} * Fstp_{water}$$ $Clocal_{eff}$ = 220 µg. L^{-1}

Concentration in surface water:

$$Clocal_{water} = \frac{Clocal_{eff}}{DILUTION * FACTOR}$$ $Clocal_{water}$ = 22 µg. L^{-1}

$$PEClocal_{water} = Clocal_{water} \text{ if } PECregional_{water} = 0$$

5.2 *Emissions from Formulation and Processing to Air resp. Soil*

A <u>Calculation of the emissions of a plasticiser</u>

Additive in the manufacturing of a tyre; emission occurs during cooling after extrusion (equation 1).

Input Data

Q_{prod}	=	33000	$kg.d^{-1}$
$Q_{additive}$	=	3.8	phr
F_{recipe}	=	2	-
F_{air}	=	0.01	-

Output Data

$Elocal_{air}$ =		6.27	$kg.d^{-1}$

5.3 *Emission from Abrasion of Tyres*

Input Data

Q_{ann_prod}	2030	$t.a^{-1}$
$F_{abrasion}$	0.12	
F_{mol_weight}	0.511	

Output Data

Eregional	124.5	$t.a^{-1}$

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7. Glossary

Adhesion
The strength of bond between cured rubber surfaces or a cured rubber surface and a non-rubber surface.

Anticoagulant (Natural Rubber Latex)
A substance added to field latex to retard bacterial action which would otherwise cause rapid coagulation of the latex. *Note:* The word "stabilizer" is often used in place of "anticoagulant" in latex terminology.

Antioxidant
A chemical used to retard deterioration due to heat, light, oxygen, or combinations thereof.

Autoclave
A pressure vessel into which materials or articles can be placed and exposed to steam under pressure. It is commonly used for vulcanization.

Butyl rubber
Isobutene-isoprene rubber (IIR), synthetic rubber, soft rubber, chlorobutyl and bromobutyl rubber as variants containing halogens with modified properties, high chemical resistance and high resistance to permeation, resistant to ozone and weathering

Calendering
Calendering is a process in tire manufacturing where steel or fabric is coated with rubber. Steel belts, body plies, cap plies and belt edge covers are all produced using the calendering process. Steel or fabric is placed between two large heavy rollers. Rubber is also inserted between the rollers at the top and bottom of the material being coated. As the material moved through the rollers the pressure forces the rubber to permeate and adhere to the steel or fabric.

Compound

A thorough mixture of natural and/or synthetic rubbers with various ingredients designed for each tire part.

Curing
he act of vulcanization; a description of a definite time and temperature of vulcanization.

Emulsifier
A surface-active substance used to facilitate the dispersion of an immiscible liquid compounding material in another liquid and to stabilize the emulsion thereby produced.

Extrusion
Extrusion is a process used to manufacture shaped solid rubber. Treads, sidewalls and bead fillers are manufactured using extruders.

Filler
A solid compounding material, usually in finely divided form, which may be added in relatively large proportions to a polymer for technical or economic reasons.

Mastication
To dissolve the rubber in solvents a technology was developed which is capable of softening the rubber by shearing it between two rotors.

Plasticiser
A compounding material used to enhance the deformability of a polymeric compound.

Rubber
Natural rubber or any synthetic, elastomeric material with physical properties similar to those of natural rubber.

Tack
The ability to adhere to itself; a sticky or adhesive quality.

Tackifier
Tackifier resins are processing aids, which help keep rubber components together prior to curing. They are especially useful in synthetic rubber compounds, which have low levels of inherent tack. Tackifiers are commonly incorporated into solvent base, water-base and hot melt adhesives with a base polymer such as natural rubber, styrenic block copolymer, ethylene vinyl acetate, polyethylene homopolymers, and butyl elastomers. The optimum type of elastomer will depend upon the final desired application and performance properties.

Thermoplastic Rubber
Rubber that does not require chemical vulcanization and will repeatedly soften when heated and stiffen when cooled; and which will exhibit only slight loss of its original characteristics.

Thickener
A substance used to raise or control the viscosity of latex with out the necessity for major changes in the total solids content.

Vulcanization
An irreversible process during which a rubber compound through a change in its chemical structure (for example, cross-linking) becomes less plastic and more resistant to swelling by

organic liquids and elastic properties are conferred, improved, or extended over a greater range of temperature.

8. Abbreviations

BR	butadiene rubber
CR	poly-2-chlorobutadiene
DIN	German Institute for Standardization
EDTA	ethylene diamine tetraacetate
EPDM	ethylene propylene diene rubber
ESD	Emission Scenario Document
EU	European Union
HF	high frequency
IC	industrial category
IIR	isoprene isobutylene rubber
IR	isoprene rubber
LCM	liquid curing method
Max	Maximum
NBR	acrylonitrile butadiene rubber
NR	Natural rubber
PEClocal	Predicted Environmental Concentration at the local stage
phr	parts per hundred rubber
RP	Rubber product
SBR	synthetic butadiene rubber
SR	synthetic rubber
STP	Sewage treatment plant
TGD	Technical Guidance Document of the European Union for Risk Assessment of New and Existing Chemicals
TI	Tyre industry
UC	use category
UHF	ultra high frequency
WDK	Association of the German Rubber Industry