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Leaching of zinc from rubber infill on artificial turf (football pitches)

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Abstract

Many artificial football pitches are treated with a rubber infill made of recycled tyres. From this material zinc leaches to the soil, groundwater and surface water. [This can present significant environmental risks, particularly for aquatic life.] Human health risks posed by leaching of zinc are negligible as zinc concentrations in the water do not exceed drinking water standards.

In the present study a targeted environmental risk assessment is performed for zinc in rubber infill on football pitches. The main focus was on the release of zinc from the rubber infill, i.e. the zinc load, the distribution of zinc between the soil, groundwater and surface water. Ageing of the rubber crumbs appears to be of major importance for estimating zinc releases. The study used simple calculation models and realistic input values, and assumed an increasing zinc release due to ageing of the rubber.

Environmental quality standards for zinc in surface water and groundwater are exceeded. The study shows that zinc from the rubber infill is either emitted mainly to the surface water (when a drainage system has been constructed on clay or peat soils) or mainly to groundwater (in naturally well-drained sandy soils). The study showed that the predicted concentrations of zinc in soil, under typical Dutch drainage conditions, also exceeds environmental quality standards.

The predicted zinc load is relatively high. For comparison, the zinc criteria in the Dutch Building Materials Decree are exceeded, and the leaching rate of zinc from rubber crumbs is up to 20 times greater than the local leaching of zinc from agricultural applications of manure and pesticides. The results can be used to assist decision-making on the use of rubber infill on football pitches where an environmental risk assessment is required.

Keywords: Rubber infill, artificial turf, zinc, leaching, ageing, aging, critical load, soil, groundwater, surface water, drainage water, emission.

Rapport in het kort

Veel kunstgrasvelden zijn ingestrooid met rubbergranulaat dat is gemaakt van gerecyclede autobanden. Uit dit soort rubbergranulaat lekt zink naar de bodem en naar grond- en oppervlaktewater. Dit kan aanzienlijke risico's voor het milieu met zich meebrengen, vooral voor het leven in oppervlaktewater. De risico's voor de mens zijn te verwaarlozen, omdat de hoeveelheid zink in het water onder de daarvoor gestelde (drinkwater)norm blijft.

Dit onderzoek haakt in op de discussie die momenteel gaande is over mogelijke schadelijke gevolgen van gebruik van rubbergranulaat uit autobanden voor kunstgrasvelden. De studie richt zich op de vraag hoe groot de uitloging van zink uit dit soort rubbergranulaat is. Het blijkt dat veroudering van rubber van grote invloed is op de hoeveelheid zink die uit het materiaal lekt. Daarnaast is onderzocht tot welke milieu-effecten dit uitlogen mogelijk leidt voor bodem, grond- en oppervlaktewater. Het onderzoek is verricht met rekenmodellen op basis van realistische waarden. Het gaat daarbij uit van het gegeven dat de hoeveelheid zink die uitloogt toeneemt door veroudering van rubber.

Milieunormen voor zink in de bodem en in grond- en oppervlaktewater kunnen worden overschreden. Bij ondoorlaatbare bodems, veen en klei, wordt het zink in het regenwater naar het oppervlaktewater getransporteerd. Bij van nature goed doorlatende zandgronden zakt het grotendeels naar het grondwater, of vloeit het weg naar het oppervlaktewater.

De uitloging van zink uit rubbergranulaat is relatief groot. Ter vergelijking: de berekende zinkuitloging overschrijdt criteria uit het Bouwstoffenbesluit. Ook is de lokale uitloging van zink uit rubbergranulaat ongeveer tot twintig keer zo groot als die van mest en bestrijdingsmiddelen in de landbouw. De studie draagt bij aan de besluitvorming over het gebruik van rubbergranulaat op kunstgrasvelden.

Trefwoorden: rubber, instrooirubber, kunstgras, zink, veroudering, emissie, immissie, uitloging, bodem, grondwater, oppervlaktewater, drainage.

Acknowledgements

Contents

Samenvatting

Van rubbergranulaat afkomstig van gerecyclede autobanden, waarmee veel kunstgrasvelden zijn ingestrooid, loogt veel zink uit naar bodem, grond- en oppervlakte water. Dat is de uitkomst van berekeningen in dit rapport over zink uit rubbergranulaat. De risico's voor de mens hiervan zijn te verwaarlozen. Voor milieukwaliteit, en met name voor het grond- en oppervlaktewater, kunnen de risico's echter aanzienlijk zijn.

De studie richt zich op de vraag hoe groot de uitloging van zink uit rubbergranulaat is, en welke milieueffecten voor bodem, grondwater en oppervlaktewater mogelijk aan die uitloging verbonden zijn. Daartoe zijn met behulp van eenvoudige rekenregels twee soorten schattingen gemaakt:

- Inschatting van de hoeveelheid zink (mg zink/m^2 kunstgras/jaar) die uit rubbergranulaat uitloogt en vergelijking daarvan met normen uit het Bouwstoffenbesluit.
- Inschatting van de zinkconcentraties in bodem, oppervlaktewater en grondwater als gevolg van uitloging uit rubbergranulaat, en vergelijking daarvan met milieukwaliteitscriteria.

Bij de aanleg van kunstvoetbalvelden wordt de bodem vijftig centimeter afgegraven. Wanneer de natuurlijke ondergrond slecht doorlaatbaar is (bijvoorbeeld klei- of veengrond) wordt hierop meestal een drainagesysteem aangebracht. Bij een goed doorlaatbare ondergrond (zand) is dat vaak niet nodig. Vervolgens wordt vijftien centimeter zand gestort en daarop een lavalaag van 35 centimeter dik, bestaande uit vijf procent rubber. Op die lavalaag komt de kunstgrasmat. Deze wordt ten slotte ingestrooid met drie centimeter rubberkorrels. Deze rubberkorrels kunnen gemaakt zijn van gerecyclede autobanden of van ander materiaal. Deze studie richt zich op de milieueffecten van rubbergranulaat uit gerecyclede autobanden.

Laboratoriumproeven en metingen aan rubbergranulaatmonsters uit het veld geven aan dat de hoeveelheid zink die uit het materiaal lekt toeneemt in de tijd (Hofstra, 2007). Dit komt door chemische en fysische veranderingen in het rubber (=veroudering). Het meenemen van de gevolgen van veroudering van rubber op de uitloging van stoffen is cruciaal voor de risicobeoordeling van rubbergranulaat. Bij het berekenen van de hoeveelheid zink die in het milieu terecht komt is rekening gehouden met deze veroudering.

In het onderzoek is een aantal aannames gedaan. Zo is er vanuit gegaan dat de hoeveelheid zink die in de lavalaag achterblijft te verwaarlozen is. Deze aanname is gebaseerd op de fysische eigenschappen van de lava, die maken dat water snel door deze laag heen zakt.

Daarnaast is ervan uit gegaan dat het zink nauwelijks in de natuurlijke ondergrond achterblijft. Bij slecht doorlaatbare ondergrond wordt namelijk door het drainagesysteem overtollige neerslag met daarin opgelost zink snel afgevoerd naar het oppervlaktewater. Bij een goed doorlatende zandondergrond zakt overtollig regenwater hier snel doorheen naar het grondwater, of vloeit het weg naar het oppervlaktewater.

Bij de berekeningen is er ten slotte vanuit gegaan dat het drainagewater van de voetbalvelden in kleine sloten terecht komt. Dit is van belang om te bepalen hoe groot de verdunning van

het drainagewater met het oppervlaktewater is. Bij grote sloten, met veel water, is de verdunning groter dan bij de kleinere sloten die vaak rond sportterreinen liggen.

Op grond van berekeningen is de geschatte zinkuitloging uit rubbergranulaat ongeveer 800 mg/m^2/jaar. Dit betekent dat een kunstgrasveld dat is ingestrooid met rubbergranulaat uit oude autobanden al na ongeveer drie jaar de immissie-eis uit het Bouwstoffenbesluit overschrijdt. Dit besluit geeft aan hoeveel zink er in 100 jaar mag uitlogen uit steenachtige materialen. Het betekent ook dat de emissie van zink uit rubbergranulaat lokaal ongeveer tot twintig keer zo hoog is als de emissie van zink uit mest en bestrijdingsmiddelen in de landbouw.

De berekende zinkconcentraties in oppervlaktewater en grondwater overschrijden milieukwaliteitscriteria. Ook de voorspelde concentraties in de natuurlijke ondergrond kunnen volgens de berekeningen de milieunorm overschrijden.

De conclusie van het onderzoek is dat de hoeveelheid zink die uit rubbergranulaat op kunstgrasvelden in het milieu terecht komt milieurisico's met zich mee kan brengen. Aanvullend onderzoek, kan de onzekerheden in deze studie verkleinen. De verwachting is niet dat daardoor de ingeschatte concentraties of risico's zullen verdwijnen. Bij de risico-inschatting in dit onderzoek is al uitgegaan van realistische waarden.

Summary

As the number of artificial turf football pitches in the Netherlands has grown, attention has focused on the potential effects of components in the rubber infill on public health and the environment. This report examines the emission of zinc from rubber infill and its distribution into the environment. The risks of zinc to public health are of no concern: the human toxicity of zinc is low and WHO drinking water criteria are not exceeded. What this report addresses are the potential ecotoxicological effects of zinc leached from rubber infill. The assessment follows two paths:
1. Estimation of the zinc load (mg/m^2 artificial turf/year) and comparison with critical loads in the Dutch Building Materials Decree.
2. Estimation of zinc concentrations in drainage water, soil, surface water and groundwater, and comparison with ecotoxicological risk limits.

Rubber crumbs from recycled tyres are used as a 3 cm infill on artificial turf. A water permeable foundation, consisting of a 35 cm lava layer (containing 5% rubber) and a 15 cm layer of sand, is laid down below the artificial turf. This study estimates the risks of a single lifecycle of artificial turf, which is approximately 10 years. The distribution of zinc in the environment was calculated using simple equations. The calculation of concentrations in the environment was not limited to a period of 10 years.

Laboratory experiments and measurements of field samples of the rubber infill show that the emission of zinc increases over time, due to chemical and physical changes of the rubber particle. This 'ageing' has been accounted for in the calculations of zinc load. Ageing of the rubber crumbs appears to be of major importance for estimating zinc releases.

As the water (with dissolved zinc) flows quickly through the permeable lava layer and the lava particles are quite large, adsorption of zinc is considered to be very small and has been disregarded. Moreover, proper data on the binding of zinc to the lava are not available.

The Netherlands can roughly be divided into two areas:
1. Clay and peat soils, where in general drainage systems have been installed. Precipitation is discharged by drainage pipes to the surface water, which means that exposure of the natural subsurface layer will be minimal. Drainage water is diluted when it enters the surface water, the dilution factor being dependent on the dimensions of the ditch. A dilution factor of 5 is considered representative for small ditches in the Netherlands (<5 m wide, <1 m deep).
2. Naturally well-drained sandy soils without a man-made drainage system. In these areas the precipitation infiltrates to the groundwater (which on average is 1 m below the soil surface) or is discharged to surface water by natural drainage. As the adsorption capacity of sand for zinc is quite small, the concentrations of zinc in the upper groundwater will in some years time reflect those in the infiltration water. As sand has a low adsorption capacity, accumulation of zinc in the subsurface sand does not occur.

The estimated zinc load is 800 mg/m^2/year. The critical load stated in the Dutch Building Materials Decree is defined as an acceptable load over a period of 100 years. This critical load (2100 mg/m^2/100 years) is exceeded after approximately 3 years. The zinc load from rubber crumbs is locally approximately twenty times higher than from agriculture (manure

and pesticides). The predicted concentrations in surface water and groundwater may exceed environmental risk criteria. Limits for the concentration of zinc in the natural subsurface sand may also be exceeded.

Additional investigation can reduce the uncertainties in this risk assessment, but this does not necessarily mean that the predicted concentrations or risks will also be reduced. The risk assessment approach reflects a realistic 'worst case' scenario.

1.   Introduction

In the Netherlands the risks of rubber infill on artificial sports (football) pitches are under discussion. The assessment focuses on human health and environmental risks.

A stakeholder group (sport federations, branch organisation of tyre industry, building contractors, accreditation institute and users/consumers) has commissioned Intron to perform literature research and measurements in order to assess the risks of rubber infill. A first assessment report was published in 2006 (Hofstra, 2006), which indicated (amongst others) the need for additional research to the risk of zinc leaching.

Intron performed experiments to leaching of zinc under laboratory and field conditions. Their study showed that ageing of rubber has a large impact on the emission of zinc. The extrapolation of the emission (mg/kg product) to a zinc load (mg/m^2) as described by Intron in the follow-up report (Hofstra, 2007) does not take into account the effects of ageing. In the present report the data from Intron are combined with information from other public sources in order to assess a more realistic zinc load to the soil, the subsequent exposure of groundwater and surface water and a preliminary comparison with environmental quality standards.

> Aim of this study is to estimate the emission of zinc from rubber infill in artificial turf of football pitches and to give an indication of the environmental risks related to the zinc emission.

2.   Characteristics of artificial football pitches

Rubber infill is used on artificial turf in order to improve the physical properties for sport (elasticity and resistance). The crumbs are a recycling product of car and truck tyres. A typical application consists of a 3 cm layer of rubber infill on a thin layer of sand. The crumbs are irregular shaped granules of approximately 2-3 mm large.

In the Netherlands the artificial turf is constructed on a well-drained foundation. This foundation consists of a 35 cm layer of lava mixed with rubber crumbs (5%) and a 15 cm layer of sand. A schematic overview of a typical situation in the Netherlands is presented in Figure 1. An overview of the installation of an artificial turf football field is shown in Appendix 1.

In the clay and peat areas in the Netherlands a drainage system is applied beneath the artificial turf on a depth of 50 cm. Drainage pipes are usually at a distance of 4 m from each other. Drainage water will be discharged to adjacent ditches. In natural well-drained sandy areas a man-made drainage system is not necessary. In these sandy areas infiltration water can enter the groundwater and will partially be discharged to the surface water as well.

An amount of 15 kg rubber/m^2 is applied; this equals approximately 100 tons per sports field (100x70 m). Rubber has a bulk density of 500 kg/m^3, so the application of 15 kg/m^2 equals a layer of 3 cm. Good properties of the artificial turf are guaranteed for a period of 10 years, and replacement is recommended after 10 years. However, practically it is expected that the artificial turf will not be replaced with that frequency, for example for financial reasons. The risk assessment in this report is based on the effects of 10 years of leaching of rubber crumbs under outdoor conditions.
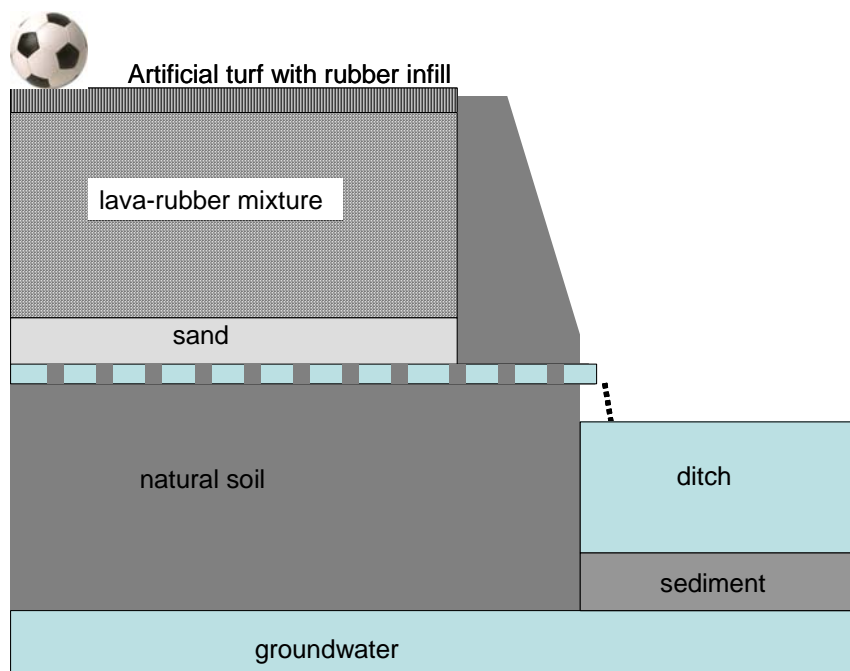

Figure 1 Schematic representation (not on scale) of the environmental compartments around artificial football pitches

3.   Ageing of rubber and emission of zinc

3.1   Ageing of rubber

Rubber is a polymer material. Zinc is used in the production process as an accelerator of the vulcanization process. Different amounts of zinc also change the vulcanization process, and as a consequence affect the performance properties of rubber, such as the stability and resistance of the material. Several qualities of rubber exist, with different zinc contents. Car tyres for example, contain less zinc than truck tyres. As a result the truck tyres are less susceptible to decomposition. Amounts of 7,000-16,000 mg zinc/kg have been reported. Zinc is chemically bound to sulphur structures of the polymer and zinc itself is not likely to be susceptible to desorption or diffusion.

Rubber is a product that is susceptible to ageing; the polymeric structure of rubber is gradually decomposed due to UV-radiation and oxidation processes. Ageing is considered as a process changing the chemical and physical properties of the rubber crumbs. Several processes involved in the ageing process are visualized in Figure 2. During ageing zinc is liberated from the polymer and could be present as dissolved zinc species or e.g. ZnS precipitates. Adsorption of zinc to the polymer is unlikely because the polymer is a neutral material, whereas zinc needs negatively charged sites for its adsorption. However the formation of polymer fragments which are charged can not be excluded.
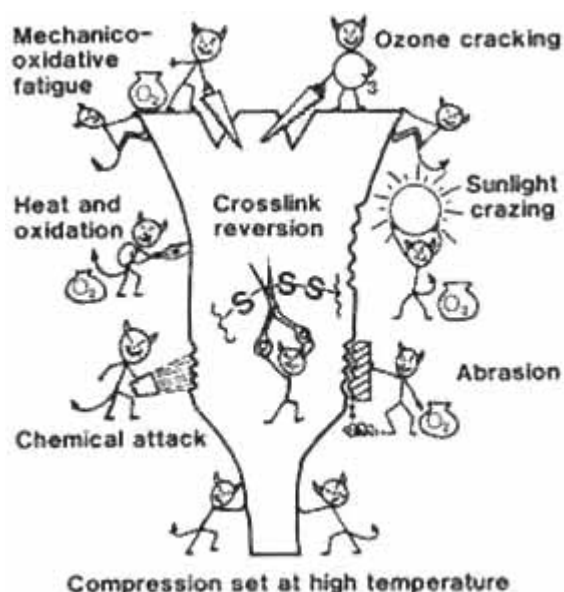
Figure 2 Types of degradation in natural and synthetic rubber vulcanisates (from: http://www.bouncing-balls.com/chemistry_tech_conservation/ageing.htm)

The decomposition or degradation kinetics of rubber has been studied by Keursten and Groenevelt (1996). They mixed 10-40% rubber crumbs (mean particle size 2.5 mm) with soil and measured CO_2 production as an indication for biodegradation. They showed that the biodegradation of rubber is a first order exponential kinetic process with rates of 0.009-0.03 day^{-1} (equivalent to half-lives of 23-77 days).

Smolders and DeGryse (2002) used 100 μm tyre debris particles to study pore water concentrations of zinc in soil. The tyre debris was either mixed with soil (2.5% w:w) or placed on top of it (1 cm layer on top of 9 cm) in a column. Columns were kept outdoors for 11 months and in that period the rainfall was 816 mm. They showed that 10-40% of the zinc from tyres is transformed in 1 year to a zinc species with the same solid-liquid distribution (so the same adsorption) as freshly added Zn^{2+}. An increase in pH over a period of 11 months was observed: in soils amended with truck tyre debris the pH increased with 1.5 units, in soils amended with car tyre debris an increase of 0.3 units was observed. This effect of rubber weathering on the pH has also been observed along roadsides (Post and Beeby, 1993; Pagotto et al., 2001; Kocher et al., 2001). Since the solubility and adsorption of zinc is very dependent on the pH this can be of great significance to transport rate and distribution over environmental compartments.

Based on literature found the following can be concluded:
- Ageing affects zinc emission
- Ageing of rubber changes the soil pH
- Ageing can result in mineralization of the rubber polymer

3.2   Emission of zinc from rubber crumbs

Due to ageing the progressive release of zinc from the polymer and the increasing porosity of the rubber crumbs lead to increasing zinc emissions as is demonstrated by measurements of Intron (Hofstra, 2006 and Hofstra, 2007). Intron measured the emission of zinc by a column leaching test (NEN 7383). This test has been developed for the assessment of the leaching of inorganic substances from building materials. In this test a vertical column is filled with rubber crumbs (<4 mm) and eluted by an upwards flow of deionized water for approximately 3 weeks until a liquid to solid ratio (L/S-ratio) of 10 has been achieved. The pH is not controlled, but is determined by properties of the material (rubber). Measurement of the inorganic compounds, pH and conductivity is measured at a L/S-ratio of 10, sometimes at lower L/S-ratios also.

In the Intron report 'fresh' production samples and aged samples have been tested. Two types of aged samples were measured:
1. aged samples that are produced by laboratory exposure to UV equivalent to 1 or 3 years sunlight exposure (ISO 4892-3).
2. aged samples that were taken from artificial turf fields of different age (1 and 3 years old).

A summary of the INTRON data is given in Table 1.

Table 1 Effect of ageing on zinc emission (mg/kg at a L/S-ratio of 10) from rubber crumbs

Ageing time	Emission at a L/S-ratio of 10 (mg/kg)			
	Car tyres		Truck tyres	
(years)	Laboratory ageing test	Field ageing	Laboratory ageing	Field ageing
0	4.6		12	
1	33	32 (n=5)	43	37 (n=3)
3	45	57 (n=3)	62	26 (n=3)

The data in Table 1 show an increasing emission for rubber crumbs of car and truck tyres aged in the laboratory. In samples aged under field conditions the emission increases for the car tyre crumbs but not for truck tyre crumbs. Trends of field emissions are more difficult to interpret because the variety in field samples is high.

In Table 2 data from column leaching studies in other countries are summarized. It is difficult to interpret these data because test methods are unknown or not identical and the origin of the samples is not described. It is important to know if it concerns production samples or aged field samples. The emission values of the Intron study fall within the range of values measured in other studies. Because of the uncertainty about the age of the samples in the other studies the data in Table 2 will not be used for the exposure estimations in the present report. Data on the zinc content in rubber crumbs are summarized in Appendix 2.

Table 2 Overview of zinc emissions of rubber crumbs from studies performed in countries outside the Netherlands

	pH	L/S-ratio	method	Test concentration liquid phase (mg/L)	Emission (mg zinc/kg rubber)
Minnesota (1990)[1]	7	?	?	?	2.3-7
Wisconsin (1992)[1]	7	?	?	?	0.44-13
Virginia (1992)[1]	7	20	?	6.1	121
Westerberg[1]	7	10	?	1.3	13
Haoya[1]	7	10	?	0.2	1.9
LUT (2004)	7	10	EN 12457	1.3	13
LUT (2004)	13.6	10	EN 12457	7.1	71
NBI (2004)	7.7	10	EN 12457	2.3	23
NBI (2004)	7.8	10	EN 12457	1.2	12
NBI (2004)	7.5	10	EN 12457	0.6	6

[1] Taken from Hofstra, 2006

With the test results in Table 1 concentrations in the outdoor environment (infiltration water, drainage water, surface water, groundwater and soil) will be computed, taking into consideration precipitation, dilution and adsorption. Equations are described in the chapters 4 and 5.

Emission of zinc from rubber products is already known from studies with tyre debris and from tyre weathering alongside highways. A study on a plot where tyres were piled showed that zinc concentrations in the groundwater were up to 6 times higher than the concentration in a control plot (Groenevelt and Grunthal, 1998). This indicated that zinc emission and leaching do occur.

A risk assessment on zinc and zinc compounds was carried out within the framework of Council Regulation 793/93/EEC on Existing Chemicals (EC, 2006). This document shows some tests, submitted by industry, on the fate of tyre debris (containing ZnO) in water. Dissolution tests have been carried out following the OECD protocol (OECD, 2001). Tyre debris (fraction <100 μm) was added to ISO 6341 medium at loading rates ranging from 10 to 100 mg/L and after 7 days the dissolved Zn concentrations were measured in the media. A maximum of 276 μg/L Zn (dissolved) was measured at a loading of 100 mg/L debris. It is important to notice, however, that 100 mg/L tyre debris only corresponds to 0.8 mg/L Zn as the amount of ZnO in car tyre debris is stated to be 1% (2% for trucks). This means that approximately 35% of the available Zn in tyre debris is already released within 7 days. Also these data indicate that the potential release of zinc from rubber products is large. The quantities emitted in this study are not used for the following estimation of emissions from

rubber crumbs because the particle size of the tyre debris in this study is much smaller than of rubber crumbs on artificial turf.

It can be concluded that there are various data showing the release of zinc into the environment from rubber to be substantial. A mechanistic understanding of the zinc emissions would further help extrapolating these test results to environmental exposure.

3.3   Mechanism of zinc emission

A mechanistic explanation for emission of zinc from rubber, is not clearly described in the literature. Based on studies using a series of different construction materials, ECN describes the following mechanisms (Van der Sloot and Dijkstra, 2004):
- Dissolution
- Desorption
- Diffusion
- Surface wash-off

These processes are largely dependent on chemical conditions (such as pH, buffer capacity, speciation, presence of other salts and the salt strength) and physical conditions (such as permeability, size and shape of the particle, tortuosity and porosity).
Ageing is considered as a process changing the chemical and physical conditions that influence the processes mentioned above.

Based on this information and the information in section 3.1 it is expected that ageing of rubber can be described by chemical transformations (oxidation) of the rubber polymer. In that process inorganic zinc species, organic zinc fragments and other organic fragments will be released. Besides, the breakdown of the polymer will result in a reduction of the size of the particles and in an increase of the porosity of the material. These processes explain the increasing emission of zinc, as reported in Table 1 in this study (from: Hofstra, 2007).

3.4   Environmental quality standards

3.4.1   Criteria for construction products

Mineral granular construction materials in the Netherlands are to fulfill the requirements in the Dutch Building Materials Decree, soon (Spring 2007) to be replaced by the Soil Quality Decree. The Building Materials Decree gives a Critical Load of 2100 mg Zn/m^2/100 years. Values in the Soil Quality Decree will be expressed as emission limit values (mg/kg product in a leaching test) but aim at the same protection level.

Although the Building Materials Decree is not intended for synthetic materials such as rubber, the applicability of rules and equations of the Building Materials Decree has been used by Intron (Hofstra, 2007) as a first indicative approach. This can be understood in the light of similarity in the granular appearance of the crumbs with stony and earthy materials for which the Building Materials Decree is intended and the use of the crumbs in an outdoor 'construction'. Moreover, column leaching tests as according to NEN 3783 as prescribed by the Building Materials Decree are available (Hofstra, 2006). However, equations (to calculate the load of a substance from the results leaching test) in the Building Materials Decree and

also in the Soil Quality Decree, do not account for ageing processes, and assume a diffusion controlled release rate only. In Figure 3 it is shown by a dotted line what the trend of the zinc emission will be when the assumptions of the Building Materials Decree are used. It is a hypothetical line, but the first order exponential decreasing shape is characteristic for diffusion controlled release. Also shown in Figure 3 are the actual measured emissions (taken from Table 1) which on the contrary show a first exponential increasing shape. Measurements and standard deviations are given as well as the trend (by a solid line). The mismatch between the curve of diffusion-controlled release and the measured emission is a strong indication that other processes determine the actual release of zinc from rubber crumbs.
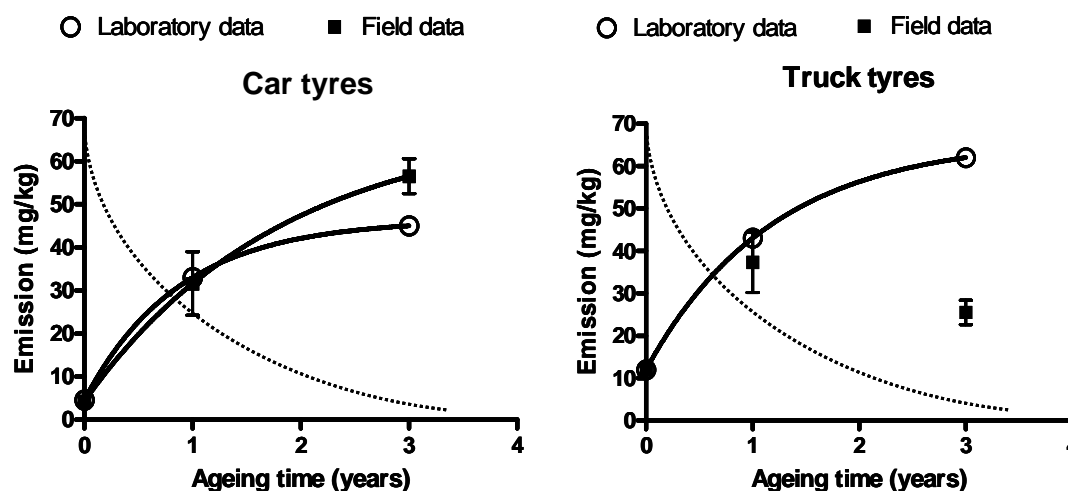

Figure 3 Measured emission of zinc from rubber crumbs of car tyres and truck tyres in laboratory and field ageing experiments. Solid lines show the statistical trend, the dotted line is a hypothetical trend assuming diffusion-controlled release only (as in the Dutch Building Materials Decree)

If ageing is neglected the zinc emissions from rubber crumbs will be seriously underestimated. Besides the issue of ageing, the applicability range of the Building Materials Decree is restricted for construction layers of 0.2-10 m in thickness. The equations are explicitly not intended to be extrapolated outside this range in order to prevent the use of contaminated materials in very thin layers. Usually such thin layers of mineral aggregates would show fast emissions within one or some years only. This would not correspond with an approach used in the Building Materials Decree, focusing on a calculation period of 100 years. The period of 100 years was considered to be adequate for the general way of use of construction materials taken into account by the Building Materials Decree. The use of 3 cm layers of rubber crumbs on artificial turf is therefore not within the applicability range of the Building Materials Decree.

Based on these graphs it is clear that equations from the Dutch Building Materials Decree are not valid for the extrapolation of zinc emission from rubber crumbs of artificial turf because the process of ageing is not taken into account.

The critical load mentioned in the Building Materials Decree has been derived based on the assumption that an input of 1% of the Soil Target Value, calculated over 1 meter of soil, will usually be acceptable due to a mix of adsorption to soil particles, solution in groundwater and time of the process. In general soil and ground water are expected to be protected at a level of target values. Equations and scenarios used in the Building Materials Decree are described by

Aalbers et al. (1996). The corresponding critical load is 2100 mg/m^2/100 years. When the zinc load of rubber crumbs is assessed more realistically (so, not using the equations from the Building Materials Decree) a comparison with the critical load of 2100 mg/m^2/100 years can still be used as a first indicative criterion to assess the potential risk of the rubber crumbs (which is done in chapter 4).

The critical loads in the Building Materials Decree will be replaced by emission limit values in the Soil Quality Decree. New critical loads have been computed for the Soil Quality Decree which are protective for soil, groundwater and surface water. Equations and scenarios used for the Soil Quality Decree are described by Verschoor et al. (2006). For these computations environmental variability and relationships between soil and groundwater have been taken into account. For zinc it is shown that exceedance of the criterion can result in exceedance of the MPC (Maximum Permissible Concentration) in groundwater in acid, sandy soils or exceedance of MPC in clay. Critical loads mentioned in the Soil Quality Decree are in principle related to *potential* risks, since they are derived from Maximum Permissible Risk levels[1]. *Actual* risks are very dependent on the specific situation and might be lower than the potential risks.

3.4.2   Quality standards for environmental compartments

The European Water Framework Directive and the daughter Directive for Groundwater demand amongst others that *inputs* of chemicals into surface water and groundwater must be prevented or limited by all reasonable means. Threshold values for selected substances are to be defined by member states by the end of 2008.

As a consequence of EC in Directive 76/464/EEC - Water pollution by discharges of certain dangerous substances, the Netherlands determined a MPC (Maximum Permissible Concentration) for zinc in surface water of 40 μg/L. The WHO criterion for Drinking Water is not adopted by the EC in the Drinking Water Directive 98/83/EC.

Other values are limits used in Dutch policy or scientific risk limits used for risk assessments. In Table 3 an overview of relevant criteria is presented. Criteria used for soil or groundwater remediation policy (Dutch Intervention values) are considered not relevant for rubber, as they are also not considered for other building materials (except in some cases for the relocation of contaminated soil). Still, intervention values are used, as a possible concentration exceedance of intervention value is important to notice. However, the intervention value must not be considered as an opportunity to increase the permissible addition. Up to date information on criteria for soil, groundwater, surface water and sediment can be found on the internet:
- http://www.rivm.nl/rvs/normen/milieu/
- http://www.rijkswaterstaat.nl/rws/riza/wateremissies/Thema/Normen_voor_het_waterbeheer/tabellen.html

Zinc is a naturally occurring element. In the MPC (Maximum Permissible Concentration) the natural background concentration (BC) is included. In the calculation of risks, the concentrations measured in the leaching tests are added to the natural background concentration. For that reasons an MPA (Maximum Permissible Addition) is derived from the MPC according to:

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[1] For certain components socio-economic considerations or harmonization with other regulatory frameworks prevailed over the MPA based emission limit values, this is also the case for zinc
[3] L/S-ratio (L/kg) = precipitation excess(L/m^2) /amount of rubber (kg/m^2)

$$MPC = MPA + BC$$

$$MPA = MPC - BC$$

Table 3 Environmental risk limits (scientific status) and legal criteria

Compartment	Criterion
Building materials	**mg/m^2/100 years**
Critical load	2100
Drinking water (WHO)	3000 µg/L
Soil	**mg/kg**
Background concentration (Target Value)	140
MPC	160
MPA	16
Intervention Value (SRC)	720
Groundwater	**µg/L**
Background concentration (>10 m bss, dissolved, Target Value)	24
Background concentration (<10 m bss, dissolved, Target Value)	65
MPA[1]	28
Intervention Value (SRC)	800
Surface Water	**µg/L**
Background concentration (dissolved)	2.8
MPC (dissolved)	9.4
MPA (dissolved)	6.6
Background concentration (total) (Target Value)	12
MPC (total)	40
MPA(total)	28

[1] An official MPC or MPA for groundwater does not exist, therefore the MPA(total) of surface is used for groundwater also. This has also been done for the derivation of new emission limit values for the Dutch Soil Quality Decree.

MPCs are derived from experiments with a diversity of species. The MPC is defined as the HC5, the hazard concentration at which more than 95% of the organisms or biological processes (i.e. nitrification) are protected.The Intervention Value is based on human and ecotoxicological Serious Risk Concentrations (SRCs). The latter is defined as the HC50, i.e. the hazard concentration at which more than 50% of the species or processes are protected. The MPC does not take the combined effect of a cocktail of substances into account.

The toxicity of zinc for humans is not very high, however zinc may pose a risk for ecosystems at relatively low concentrations as can be seen from the environmental quality standards. This is illustrated by the high drinking water standards in comparison with the lower MPCs for surface water and groundwater.

4.   Assessment of zinc load

Emission is a property of a product; it provides information about the release of a substance per kg of product. The load gives information about the input to the soil as a result of the use of the product in a construction on the soil surface. For the computation of the load, information is required about the properties of the product (emission), but also about the amount of product used in the construction.

The risk assessment of zinc from rubber crumbs on artificial turf starts with an estimation of the zinc load (mass of zinc/m^2 soil). The zinc load can then be compared with values of the Building Materials Decree or Soil Quality Decree. In Table 1 a number of emission data is available. The zinc load can be computed from these emission data. From the measurements a good estimation of the trend in emission should be made. Extrapolation of emission values should be based on an understanding of the underlying processes and a proper description of these processes. A rough description of processes involved in the emission is made in section 3.1. However, based on the current information, no mathematical or quantitative description of all these processes is possible. Therefore two approaches have been elaborated here: 1) assuming a constant zinc emission and 2) assuming an increasing zinc emission. Both approaches include decomposition as a major process in the zinc emission.

4.1   Approach 1: A constant zinc emission.

In this approach it is assumed that ageing is continuing, but that exhaustion of available zinc is balanced by the release of zinc by decomposition of the matrix. It is unknown if the zinc emission will increase further after 3 years. Therefore, the maximum values for car tyre crumbs of 45 and 57 mg/kg are used as measured in respectively laboratory and field ageing and for truck tyre crumbs maximum values of respectively 62 and 37 mg/kg have been used (see Table 1).

The estimated annual release of zinc in the environment for this scenario is:

Equation 1

$$Load_{zinc} = \frac{E_{L/S10}}{M_{rubber}}$$

$Load_{zinc}$ = annual release of zinc into the environment (mg/m^2/year)
$E_{L/S10}$ = emission of zinc in a laboratory column leaching test (mg zinc/kg rubber)
M_{rubber} = amount of rubber applied on the artificial turf (kg/m^2)

In Figure 4 the cumulated load and the amount of residual zinc in the rubber are shown.
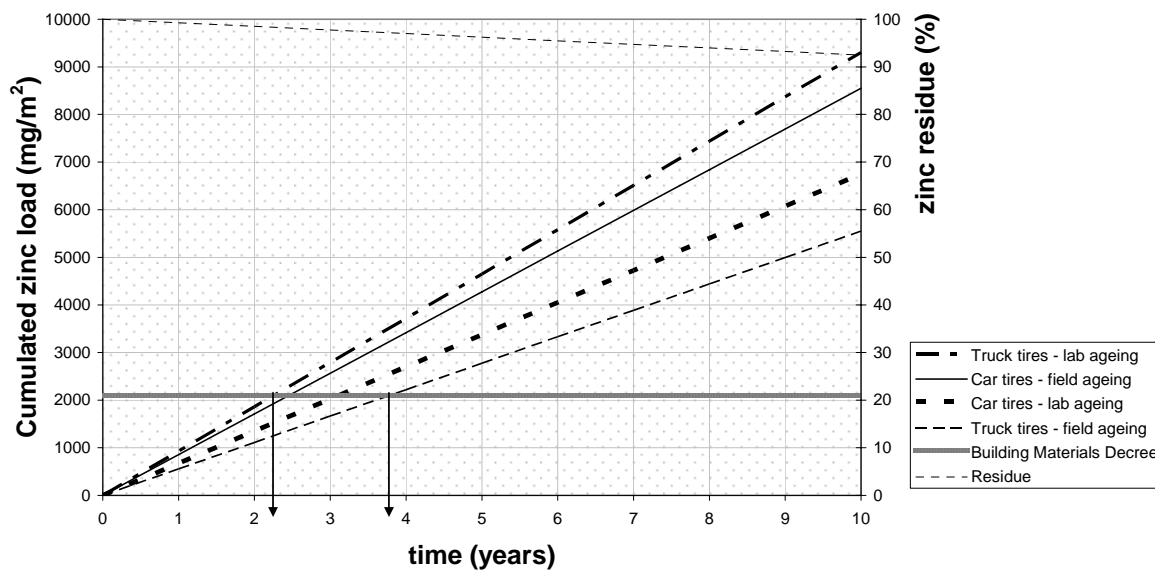
Figure 4 Cumulated zinc load (mg/m^2) assuming a constant emission due to ageing and critical load from Building materials Decree on the left y-axis. Residual zinc (average) in rubber (%) on the right y-axis over a period of 10 years

For computation of the zinc residue an average zinc content is taken of 7560 mg/kg, as determined by Intron in 5 field samples of 1 year old. The figure shows that exhaustion of rubber is not occurring within the life-time of the rubber infill (after 10 years the infill is to be replaced). In ten years the total amount of zinc in the rubber crumbs is estimated to be decreased by approximately 8%. The fact that 92% of the zinc is still present supports the assumption that zinc emissions can be constant or increasing for a longer period than 3 years.

4.2   Approach 2: An increasing zinc emission

A statistical analysis performed by Intron (Hofstra, 2006) showed that the increase of themeasured emission in the ageing test was significant. A stabilization of the emission to a constant value has not been shown within a period of 3 years of ageing. The question is how the emission will develop after 3 years. Will it be constant like in the previous scenario, or will it continue to increase. It is very likely that at some time the emission will decrease, e.g. after some decades. The latter will be the case when rubber is almost completely degraded and zinc is becoming exhausted. As is shown for approach 1, exhaustion of zinc is not relevant within the life-time of the rubber infill.

This second scenario is based on the hypothesis that zinc is released by a rate that is related to the rate of degradation rate of the polymer. Photochemical, chemical and microbiological processes contribute to the ageing of rubber. The most widely acknowledged empirical approach to describe chemical as well as microbial degradation kinetics is a first order exponential equation. Moreover, first order exponential kinetics have been observed for rubber (Keursten and Groenevelt, 1996). When a first order exponential kinetic is applied to degradation of the polymer, this will also be valid for the release rate of its transformation products. Since zinc is part of the polymer structure its release can be considered as the production of a transformation product.

In Figure 3 the trend lines are shown according to a first order exponential pattern. It shows that in laboratory ageing studies the zinc emission increases for rubber from both car and truck tyres. In field studies this effect is only seen for rubber from car tyres. A possible explanation for this could be that field samples are more variable because they originate from different fields, i.e. samples aged for 3 years originate from other fields than samples aged for 1 year.

Using the increasing zinc emission, an average zinc load of 8000 mg/m^2/10 years is predicted, see Figure 5. This may vary from 6700-9300 mg/m^2/10 years when data for car and truck tyre debris are treated separately. Approach 1 assuming a constant emission also falls within this range, with 8550 mg/m^2/10 years. The criterion of the Building Materials Decree is then exceeded within 2.5-3.5 years after application of the rubber infill.
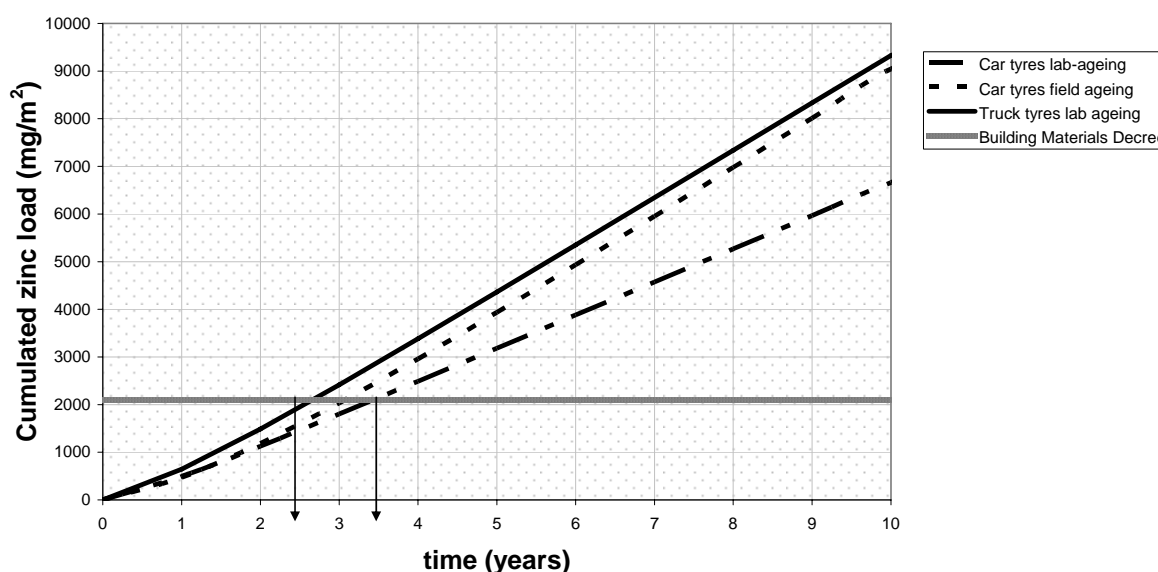
Figure 5 Cumulated zinc load (mg/m^2) assuming an increasing emission due to ageing

4.3   Uncertainties in the estimation of the zinc load

The two approaches from the previous section show that the cumulated zinc load is likely to be between 6,000-10,000 mg/m^2/10 years. The difference in estimated zinc load between the two approaches is not large. Approach 1 which assumed a constant emission due to ageing falls within the estimated range of approach 2, which assumed an increasing emission due to ageing.

Estimation of the zinc load shows that the criterion of the Building Materials Decree is exceeded after approximately 3±0.5 years. There are still a lot of uncertainties about the mechanism of zinc emission and measurement of the effect of ageing on rubber that has aged for longer periods of time (>3 years) are not available. Approach 2 is the most likely scenario based on the current state of knowledge.

The following aspects have not been taken into account, possibly leading to underestimation of the zinc load:

1. Difference in L/S-ratio between column experiments and the field situation.
2. Difference in pH between column experiments and field situation
3. The presence of rubber in the foundation. 5% of rubber is present in the 35 cm lava layer.
4. The supplement of 10% rubber after 1 year.
5. The possibility of accelerated zinc emission after 3 years due to progressive ageing.

The following aspects have not been taken into account, possibly leading to overestimation of the zinc load are:
6. The possibility of a decreasing emission after a period of 3 years
7. The possibility that the zinc emission is retarded due to diffusion.

Ad. 1 Liquid-solid ratio
The measured emission values are the result of column leaching experiments with a L/S-ratio of 10. In the field the annual L/S-ratio is dependent on the amount of rainfall and irrigation and will be at least 20^3.
Since artificial turf is designed for a quick transfer of water to the subsurface and withdrawal of water by plant growth is not present, it is very likely that effective infiltration will be much more than the average precipitation excess of 300 mm which is usually considered as a representative value for the Netherlands. A minimum downward flux of 600 mm is assumed, and this corresponds with a L/S-ratio of 40.
It is uncertain if the difference in L/S-ratio between the column experiments and the field situation will affect the emission in the field. We assume that the transformation of rubber and the release of zinc is time-dependent and does not depend upon the amount of infiltration water.

Ad. 2. pH
In the column experiments performed by Intron, deionized water is used as eluate. This has a neutral pH of 7. In contact with rubber, the pH in the solution will be determined by the properties of the material (rubber). The solubility of zinc is very dependent on the pH. The solubility of zinc-species increases exponentially with decreasing pH. Under field conditions rubber is exposed to rain. Clean rain water has a pH of approximately 5.6, so the solubility and emission under field conditions could be considerably higher than in a column leaching experiment. There are studies that suggest that rubber has a increasing effect on soil pH. When this also happens on the artificial turf, the effect of a lower pH might be partially compensated. As yet, is recognized that the emission of zinc is could be very sensitive to the pH, but there is not enough information to quantify the impact under field conditions.

Ad. 3 and 4. Additional rubber sources
The contribution of zinc emission from the 35 cm lava-rubber layer below the artificial turf has not been taken into account. One reason is that ageing of rubber in the subsurface can be very different from ageing of rubber on the surface. Exposure to sunlight is an important factor in the ageing of rubber. If the same emission trend for rubber in the subsurface is assumed as for rubber on the soil surface, the estimated zinc load can increase with 60-70%. Also the additional supply of 10% rubber after 1 year has not been quantified. Taking this contribution into account can raise the zinc load by approximately 10%.

Ad 5 and 6. Extrapolation of zinc emission
With the current information of zinc emission from aged rubber crumbs it is most likely that the zinc emission is increasing within the lifetime of the rubber infill (10 years). The emission seems to stabilize after approximately 4 years, but no data are available for the period after

3 years to verify this assumption. Considering the fact that the zinc content in rubber is very large in comparison to the amount that is released, exhaustion of zinc is not likely within the lifetime of the rubber infill. Accelerated emission after 3 years due to progressive transformation of rubber or a decrease in emission due to reduced transformation of rubber can not be excluded. A change in physical properties of rubber due to ageing leading to reduced emission of zinc can also not be excluded. More research to the mechanisms and effects of rubber ageing is required to reduce those uncertainties.

Ad 7. Diffusion
It is likely that diffusion and decomposition are simultaneous processes. Transformation processes attack on the surface of the particle. Zinc released during that process will mainly dissolve (depending on the pH) directly into the water phase. Diffusion is assumed to be of minor importance. A better quantification of all the release processes and the development of a mathematical model that addresses these mechanisms can refine the present assessment.

Estimation of the zinc load shows that the criterion of the Building Materials Decree is exceeded after approximately 3±0.5 years. Removal of the rubber infill after its lifetime of 10 years (or earlier) does not reduce the risks of zinc that has already leached into the soil. For the prediction of exposure of soil, groundwater and surface water an average emission of 800 mg/m^2/year is used (range 600-1000 mg/m^2/year). It is shown that in the first 3 years the emission is increasing. For the estimation of exposure of soil, groundwater and surface water, longer time periods are taken into consideration. Therefore it is justified to use an average of 800 mg/m^2/year as a constant value.

The next chapter will go into more detail for the exposure of the environmental compartments.

5.   Exposure assessment

5.1   General assumptions

In this chapter the estimated zinc load from section 4 is used as an input to the environment and concentrations in drainage water, soil, surface water and groundwater will be computed, based on simple equations.

Once zinc has been released from the rubber into the water phase it is transported to the layer directly below the artificial turf. Artificial turf fields are constructed in such a way that a quick vertical transfer of water to the subsurface is warranted, so that surface run-off can be neglected. This subsurface is very permeable and consists of a 35 cm lava-rubber mixture (see Figure 1). Downward transport of dissolved zinc can be retarded due to adsorption to the lava. This means that emissions on the soil surface are not immediately visible in for example drainage water.

Concentrations in the environmental compartments are no fixed values. Input and transport processes are both dynamic. Concentrations may rise and fall, due to transport from one compartment to another, for example from soil to groundwater, or from drainage to surface water to sediment. Concentrations of zinc are dependent on the time point of interest and the depth or distance from the input source. The risk assessment approach in this report uses simple equations to compute potential risks. These simple equations do not provide a good insight in changes in depth and time. They give an idea of average concentrations that can be found after some time. When possible an indication is given of the relevant time-scale.

A more refined assessment using numerical models can provide detailed depth and time profile. However these models also require more specific input information of soil properties, precipitation, hydrology, pH, adsorption etcetera. This information is not available at this moment.

5.2   Infiltrating water

Since artificial turf is designed for a quick transfer of water to the subsurface and withdrawal of water by plant growth is not present, it is very likely that effective infiltration will be much more than the average precipitation excess of 300 mm which is usually considered as a representative value for the Netherlands. A minimum downward flux of 600 mm (L/m^2) is assumed. Using Equation 2 with an annual zinc load of 800 (±200) mg/m^2/year the average concentration in the infiltration water is 1.3 mg/L (range 1.1-1.6 mg/L).

Equation 2

$$C_{in} = \frac{Load_{Zinc}}{N_i}$$

C_{in}	= dissolved zinc concentration in infiltration water (mg/L)
$Load_{zinc}$	= zinc load (mg/m^2/year)
N_i	= effective infiltration = precipitation excess (mm=L/m^2/year)

5.3   Soil

For this risk assessment only the natural soil layer below the artificial turf is considered as soil. The lava-rubber and sand layer applied as a foundation are considered no part of the soil. It is assumed that these layers are removed, if the land-use changes. When the artificial turf and its foundation are removed, it is likely that a new layer of approximately 50 cm of soil has to be applied in order to keep the same level as the surrounding area. Thus the upper layer is replaced and has not been exposed.

The quality of the subsurface is mainly relevant because it is a possible source of groundwater contamination.

Question is to what extent the natural subsurface is exposed to zinc. Two situations can occur:
1.   clay and peat soils, with a drainage system on top of it.
2.   sandy soil low in organic matter, with no drainage system because the natural permeability is good

Ad. 1. Clay and peat soils
It is assumed that exposure of these soils to zinc leached from rubber is negligible, because infiltrated water will be mainly discharged by the drainage system to the surface water.

Ad. 2. Sandy soil, low in organic matter
Infiltration in a permeable sandy soil is a vertical transport process to the groundwater. These sands have a low adsorption capacity for zinc. Adsorption in the lava-rubber layer is neglected, because of the high vertical flux and preferential flow paths in this material. To get a rough impression of potential zinc exposure of the natural sandy subsurface, a worst case assumption is made, that the complete load of zinc enters this natural sandy subsurface. The natural subsurface is thus continuously exposed to infiltration water with a concentration of 1.3 mg/L (range 1.1-1.6 mg/L).

Concentrations on the solid phase, are computed by Equation 3 which requires information on the adsorption coefficient of zinc.

Equation 3

$$Zn_{soil} = Kf \cdot [Zn]_{solution}^{n}$$

Zn_{soil}	= concentration of zinc on the solid phase (mg Zn/kg soil)
Kf	= adsorption coefficient (L/kg)
$[Zn]_{solution}$	= concentration of zinc in the soil solution (mg Zn/L water)
n	= Freundlich exponent

Since adsorption is very variable due to the large variation in soil properties in the environment, two approaches are elaborated to deals with this variation.
A.   A fixed adsorption coefficient which is thought to be representative for sandy soils in the Netherlands and which has been derived from a dataset with measurements in 47 soil types (Verschoor et al., 2006)

B. The variation in adsorption coefficients is computed by applying relationships between the adsorption coefficient and the pH, organic matter and clay content (Römkens et al., 2003).

Ad. A
A realistic lower limit of the adsorption coefficient of zinc in sandy top soils in The Netherlands is 11 L/kg. The concentration-dependency of the adsorption is neglected, so n=1. Concentration to the solid phase will become in equilibrium with the concentration in the liquid phase according to Equation 3.

Assuming linear adsorption based on a dataset of 47 Dutch soil types the concentration increase of zinc on the solid phase of the natural subsurface sand is 15 mg/kg.

Ad B.
There are equations describing the relation between the adsorption coefficient of zinc with soil pH, organic matter and clay content. (Römkens et al., 2003). It is shown that the adsorption of zinc is very sensitive to pH and the organic matter content. The equation 3 is based on a regression of measurements in 60 Dutch soil samples.

Equation 4

$\log Kf = -4.59 + 0.49*pH + 0.61*\log(SOM) + 0.39 * \log(\%clay) - 0.19*\log (DOC)$

SOM   = Organic matter content in the solid phase (%)
DOC   = Dissolved organic carbon (%)

In Table 4 the variation of Kf with pH and organic matter is shown. The Freundlich coefficient n has been estimated to be 0.78. Table 4 shows that within the group of sandy soils the adsorption can vary considerably.

Table 4 Adsorption coefficients (L/kg) as a function of pH and organic matter content. % clay is kept constant at 5%

pH \ %OM	4	5	6	7
0.05	1	2	7	21
0.2	2	5	16	49
0.5	3	9	27	85
1	4	14	42	130

The consequence of this variation is that the estimated increase in zinc concentrations on the solid phase, based on non-linear adsorption (Equation 3) and taking variation in pH and organic matter into account, are expected to vary from 1-188 mg zinc/kg soil.

As mentioned above, this is a rough worst case estimate. A natural horizontal drainage is neglected and adsorption to the lava-rubber mixture and sand foundation is also not taken into account. The assessment can be improved by information on the physical/chemical properties of lava with respect to binding and water fluxes. However, for a generic risk assessment aiming to protect a heterogeneous environment, this is an acceptable approach.

5.4   Groundwater

As mentioned before, a drainage system may not be present on naturally well-drained sandy soils. About 50% of the Netherlands (roughly the south and eastern part) consists of these sandy soils. In absence of an artificial drainage system, infiltration water is supplies the groundwater resources. To obtain a first impression about the risks it is assumed that 100% of the infiltration water enters the groundwater.
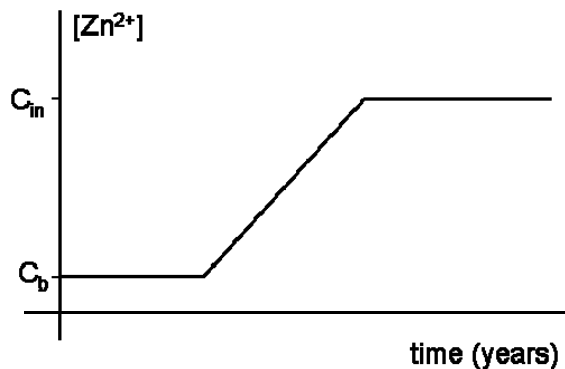
Figure 6 Schematic drawing of breakthrough of zinc concentrations in groundwater. C_b = background concentration, C_{in} = concentration in infiltration water

Effects of zinc emission on groundwater are expected to become visible after a few years to up to decades because of retardation of zinc during transport in the sand. This means that effects can continue for a prolonged period, also when the artificial turf field is removed. Exact periods of time are very dependent on properties of the soil below the artificial turf. After a period of time breakthrough of zinc occurs. Due to continuing zinc load the zinc concentrations in the upper groundwater will approach the concentration in the infiltration water. Predicted concentration in the upper groundwater is 1.3 mg/L (range 1.1-1.6 mg/L).

The retardation factor is calculated by:

Equation 5

$R = 1 + (\rho \cdot K / \theta)$

R = retardation factor
ρ = bulk density of the soil (default 1.5 kg/dm^3)
θ = porosity (default 0.35)
K = linear adsorption coefficient (L/kg)

From Equation 5 it follows that with an estimated Kp of 11 L/kg (in the natural subsurface sand), the travel time of zinc is 48 times longer than the travel time of water. As in section 5.3 and 5.5 it is assumed that the lava-rubber and sand foundation do not adsorb zinc. Expected travel times of zinc to the upper groundwater under sandy soils is in the order of months-years, depending on local soil properties.

5.5   Surface water

The exposure route of zinc from rubber infill to surface water is by drainage. The construction of the artificial turf prevents surface run-off. An amount of 600 mm infiltration water with 1.3 mg zinc/L (range 1.1-1.6 mg/L) is predicted to enter the lava-layer. Typical travel times of water in the lava layer are estimated to be in the order of hours-days, depending on moisture conditions. The adsorption of zinc to the lava is considered to be negligible.

Common dimensions of artificial turf football fields are 70*100 m.
The total annual discharge of water is 7000 m^2 * 600 mm = 4200 m^3 water.
The total annual mass of zinc entering the surface water over a length of 70-100 m is 5.6 kg.

Estimated *total* surface water concentrations can then be estimated by Equation 6, adapted from Technical Guidance Document on Risk Assessment (European Commission, 2003):

Equation 6

$$C_{SurfaceWater} = \frac{C_{drainage}}{dilution}$$

C$_{SurfaceWater}$ = dissolved concentration in the water during the emission episode (μg/L)
C$_{drainage}$ = concentration in the drainage water (μg/L)
dilution   = dilution factor

Estimated *dissolved* surface water concentrations can then be estimated by Equation 7, adapted from Technical Guidance Document on Risk Assessment (European Commission, 2003):

Equation 7

$$C_{SurfaceWater} = \frac{C_{drainage}}{(1 + Kp_{susp} \cdot SUSP_{water} \cdot 10^{-6}) \cdot dilution}$$

Kp_{susp} = solids-water partition coefficient of suspended matter (L/kg)
$SUSP_{water}$ = concentration of suspended matter in the surface water (ditch) (mg/L)
10^{-6} = unit conversion factor

An assessment of dilution factors is done using the 'CIW- Mixing zone model', which is a spreadsheet model for the computation of surface water concentrations at a certain distance from the discharge, dependent on the volume of discharge water and the dimensions of the surface water. The model has standard characteristics for polder ditches, canals, rivers and lakes and dimensions and discharges can be defined by the user.

Computations with the 'CIW-Mixing zone model' show dilution factors that are higher than the default dilution factor of 10. Based on the concentrations mentioned in Table 6 dilution factors of approximately 50-90 are computed (see Appendix 3).

The polder ditch is still a relatively large ditch. This ditch has also been used by Verschoor et al. (2006) in the derivation of new emission limit values for the Soil Quality Decree (2007). However, in other regulatory frameworks i.e. for pesticide registration (pesticide drift to surface water) ditches of 1 m wide and 30 cm deep are used. The impact of zinc leaching on the water quality of these small ditches, which are abundant in the Netherlands, is higher than on larger ditches, canals and rivers. In small ditches the concentrations in the water may even approach concentrations in the drainage water, because drainage is the main input source of water. Expert judgement indicated a realistic dilution factor of 5 to be likely for small ditches in the Netherlands (Peter Vermij, RIZA, pers. comm.). The zinc concentration in surface water is, as for soil and groundwater, very dependent on local conditions. For the risk assessment a *total* concentration *increase* of 22-32 µg/L for polder ditches and 220-320 µg/L for small ditches are considered (Equation 6). These surface water concentrations are total added concentrations, and include dissolved zinc as well as adsorbed zinc to suspended matter.

Equation 7 computes the dissolved amount of Zn by considering adsorption to suspended organic matter. A relatively low $SUSP_{water}$ for these small ditches of 15 mg/L is taken from Appendix 5 (NL-ditches). This value is also used in the EUSES model as a default value. The EU zinc RAR used a Kp_{sus} of 110000 ($logKp_{sus}$ =5.04). Taking an average drainage water concentration of 1100-1600 µg/L, the estimated *dissolved* surface water concentration varies from 83-120 µg Zn/L in small ditches and from 8-12 µg/L in polder ditches.

5.6   Discussion of exposure assessment

The further the distance from the input source, the more uncertainties are involved in the risk assessment. The amount of infiltrating water (precipitation excess) is quite uncertain and this could have a substantial effect on the estimated concentrations. Moreover, assumptions on the retardation of zinc on the lava layer are very uncertain and affect the predictions of drainage water concentrations, surface water concentrations, soil concentrations and groundwater concentrations.

Precipitation excess
A precipitation excess of 600 mm/year is assumed in this study. The total average precipitation is 800 mm/year. An average value for effective infiltration to the groundwater in sandy areas in the Netherlands is 300 mm. This concerns precipitation reduced by evaporation and transpiration (plant uptake). Calculations for bare soil indicate that under normal Dutch weather conditions the effective infiltration can increase to 450 mm/year. Artificial turf fields are very permeable for water and also the subsurface is prepared in such a way that water is quickly transported to deeper layers. Adjustment of the hydrology in the model computations in combination with soil properties mentioned above is recommended.

Retardation of zinc on lava
The fact that high infiltration rates reduce the adsorption of zinc is amongst others described by Miretzky et al. (2006). The high flow rate in combination with preferential flow through macropores will reduce the contact of zinc with potential binding places. Therefore as a worst case assumption in the first step of the risk assessment it is assumed that no adsorption in this lava matrix takes places. However, there is information about the potential high sorption of rubber to scoria (lava) powder (Kwon et al., 2005). Particles < 0.1 mm showed a removal of 80% of zinc from the solution. The removal decreased drastically to less then 10% for

particles of 0.5-2 mm. They even recommended the use of scoria powder as a material for sorption of zinc in contaminated water. Scoria powder showed a larger capacity and affinity for Zn^{2+} than commercial powdered active carbon. Lava stone used as a foundation is much larger than the powder used in the tests (see Appendix 1, picture 5). Moreover, it is unknown if the chemical composition of lava used in the foundation is similar to that of the scoria tested. The scoria showed a non-linear adsorption, indicating that saturation of adsorption sites can occur. The chemical composition of scoria, used in the experiments of Kwon et al. (2005) is described in Appendix 4.

6.   Risk assessment

Building Materials Decree
In chapter 4 it was already indicated that the predicted zinc load from 10 years leaching of rubber crumbs on artificial turf exceeds the criterion in the Dutch Building Material Decree by a factor 3-5. Estimation of the zinc load shows that the criterion of the Building Materials Decree, which is defined as the acceptable load over a period of 100 years, is exceeded after approximately 3 ± 0.5 years. When rubber crumbs will be exposed on the fields for a period longer than 10 years, the exceedance of criteria in the Building Materials Decree will increase further.

Environmental Risk Limits
Based on the current assessment of zinc leaching from rubber crumbs and zinc behaviour in the subsurface layers, zinc concentrations in surface water, soil and groundwater can be compared with environmental quality standards. An overview of the risk assessment in which predicted concentration increases (ΔC) in various compartments are compared with corresponding environmental quality standards, is given in Table 5. The ratio of ΔC/MPA gives an indication of potential ecotoxicological effects (ratio >1 is potential risk).

Table 5 shows that environmental risk criteria in soil, groundwater and surface water may be exceeded, by leaching of zinc from rubber infill on artificial turf. Theoretically the ratio ΔC/MPA for dissolved concentration and total concentrations should be identical. The difference between exceedance factors for dissolved and total concentrations in surface water is explained by the fact that the Kp_{susp} used in this study (taken from the zinc RAR) is not identical to the Kp which originally was related to the derivation of the MPC(total) and MPC(dissolved).

Table 5 Overview of the risks of leaching of zinc from rubber crumbs on artificial turf. Criteria, predicted concentration increase (ΔC) and the ratio ΔC/MPA are shown. A ratio ΔC/MPA>1 is printed in bold and indicates potential risk

Compartment	Criterion	ΔC	Ratio ΔC/MPA[1]
Soil	**mg/kg**	**mg/kg**	
Background concentration	140		
MPC[2]	160	1-188[4]	
MPA	16		0.1-**12**
Intervention Value	720		
Groundwater	**µg/L**	**µg/L**	
Background concentration (>10 m bss, dissolved, Target Value)	24		
Background concentration (<10 m bss, dissolved) / Target Value	65	1100-1600[5] (total)	
MPA (total)[3]	28		**39-57**
Intervention Value	800		
Surface water (small ditches)	**µg/L**	**µg/L**	
Background concentration (dissolved)	2.8		
MPC (dissolved)	9.4	83-120[5] (dissolved)	
MPA (dissolved)	6.6		**13-18**
Background concentration (total)	12		
MPC (total)	40	220-320[5] (total)	
MPA (total)	28		**8-11**
Surface water (polder ditches)	**µg/L**	**µg/L**	
Background concentration (dissolved)	2.8		
MPC (dissolved)	9.4	8.3-12[5] (dissolved)	
MPA (dissolved)	6.6		**1.3-1.8**
Background concentration (total)	12		
MPC (total)	40	22-32[6] (total)	
MPA (total)	28		0.8-**1.1**

[1] MPA = Maximum Permissible Addition, MPA=MPC-background,

[2] MPC = Maximum Permissible Concentration, see section 3.4

[3] The MPA surface water is adopted because MPA groundwater is not available

[4] range due to variation in adsorption capacity of soils, and different qualities of rubber

[5] range due to different qualities of rubber

'CIW emission-immission impact assessment'
The 'CIW mixing zone model' has been used to compute the dilution of drainage water in polder ditches. The 'CIW mixing zone model' and the 'emission-immission impact assessment' are used in the Netherlands as a tool in allowance of discharges of waste water to surface water. The concentration increase at the end of the mixing zone should meet $0.1*MPC_{surface water}$ (= 4 µg/L as total). The concentration increase in polder ditches due to drainage of zinc from artificial turf is given in Table 5, and amounts 22-32 µg/L. This means that the CIW criterion is not met; an exceedance factor of 4.5-8 is predicted. The impact of zinc in drainage water on smaller ditches (220-320 µg/L) is considerably higher, the CIW criterion may be exceeded by a factor 45-80.

Bioavailability
The last decade a lot of discussion has emerged on the bioavailability of metals in the environment. The recent EU risk assessment on zinc (European Commission, 2006) pragmatically incorporates a bioavailability concept for the compartments surface water, soil and sediment.

For *surface water* the Biotic Ligand Model (BLM) is used for determining the fraction of dissolved zinc that is bioavailable to the target species. With these BLMs so-called bioavailability factors (BioF) were estimated which depend on the 'key' water characteristics pH, hardness and DOC (Dissolved Organic Carbon) of a typical water. This BioF is subsequently used for correcting the dissolved concentration into the actual bioavailable concentration. The corrected concentration is then compared with the PNEC (Predicted No Effect Concentration). The table in Appendix 5 gives an overview of the range of BioF for a number of EU waters. BioF range from 0.1 to 1 (no correction). The values in bold point to the values actually used in the zinc risk assessment. Although the zinc risk assessment is not officially finalised yet in the EU and bioavailability is not officially introduced in national legislation, the BioF can indicatively be used in the risk assessment of rubber crumbs. Assuming that zinc discharges from fields will mainly go to ditches, the BioFs amount to 0.6 (realistic worst case conditions) and 0.2 (average conditions) (see Appendix 5, under 'NL ditches'). These BioF are relatively high compared to other water types which is mainly due to the high DOC levels in such lowland ditches. The predicted concentrations from Table 5 (i.e. 83-120 μg/l) should be multiplied with above-mentioned BioFs before comparing them with the relevant environmental quality standard for surface water (MPA). This results in values of 17-24 μg/L with the average BioF and 51-72 μg/L with the realistic worst case BioF. The exceedance factors then become 2.6-3.6 (average) and 8-11 (realistic worst case). Conclusion: the introduction of a bioavailability correction for zinc as proposed in the EU zinc risk assessment leads to lower exceedance factors but these exceedance factors are still clearly above one.

For the bioavailability correction in *soil* the EU risk assessment of zinc and zinc compounds uses a tiered approach in which both a lab-to-field correction (ageing aspects of zinc) and a soil properties correction step is applied on the PEC side. The lab-to-field factor has a standard value of 3, whereas the factor for soil properties on average ranges between 1 (sandy soils) and 1.5 (river clay/peaty soils). As the soil scenario for rubber crumbs is conducted for sandy soils the overall soil bioavailability correction factor amounts to 3 (= 3*1). Present (uncorrected) soil concentrations (1-188) should be divided by 3 to get corrected concentrations (0.3-63). Comparing these concentrations with the MPA of 16 mg/kg would result in exceedance factors of 0.03-4. So similar to water it can be concluded that the introduction of a soil bioavailability correction for zinc as proposed in the EU zinc risk assessment leads to lower exceedance factors, but they are still above one.

7.   Conclusions and discussion

As the number of artificial turf football pitches in the Netherlands has grown, attention has focused on the potential effects of components in the rubber infill on public health and the environment. This report examines the emission of zinc from rubber infill and its distribution into the environment. The risks of zinc to public health are of no concern: the human toxicity of zinc is low and WHO drinking water criteria are not exceeded. What this report addresses are the potential ecotoxicological effects of zinc leached from rubber infill. The assessment follows two paths:
1.   Estimation of the zinc load and comparison with critical loads stated in the Building Materials Decree.
2.   Estimation of zinc concentrations in drainage water, surface water, soil and groundwater and comparison with environmental quality standards.

Estimation of zinc load
Measurements in field-aged rubber crumbs and laboratory-aged rubber crumbs show an increase in emission values in column leaching studies. This increase can only be explained by ageing: as the chemical and physical properties of the rubber change over time, emissions increase.

The phenomenon of ageing and increasing emission of zinc cannot be described by the current equations given in the Building Materials Decree for stony and earthy granular materials, which take only diffusion into account and not ageing. Neglecting the observed ageing of rubber crumbs will lead to a substantial underestimation of zinc loads. The critical loads mentioned in the Building Materials Decree are considered to represent a first rough indication for the potential environmental risks.

Two methods for taking ageing into account have been developed. The first assumes a constant and relatively high emission after 3 years of ageing. The second takes the actual increase in zinc emissions into account. Total zinc loads in these methods do not differ very much, but the latter does more justice to the observed emission pattern. A total zinc load of 6,000–10,000 mg/m^2/10 years is estimated, depending on the quality of the rubber. An average zinc load of 800 mg/m^2/year was used for the risk assessment.

The uncertainties in the estimation of the zinc load are due, among others, to the following:
•   Measurements for ageing cover periods up to only 3 years; figures to 10 years have been extrapolated.
•   The L/S-ratio in column leaching experiments will reflect leaching under field conditions.
•   The impact of pH differences in column leaching experiments (approximately 7) on predicted zinc emissions under field conditions (pH approximately 5.6).

Under the assumptions made for these uncertainties, which are not conservative, emissions stabilise after 3 years. Column experiments are used as representative systems for the field situation despite the fact that L/S-ratio is lower and the pH is higher.

In a very recent report from Alterra research centre (De Vries et al., 2004) a prediction was made on the (potential) risks of zinc accumulation in agricultural soils in the Netherlands.

Major sources of zinc inputs varied only slightly between different land uses and soil types. In grassland animal manure contributes most (more than 90%) to the input of zinc. Fertilizers are a comparatively small source of zinc, whereas atmospheric deposition is also limited. Other sources, mainly compost and pesticides are a substantial source in arable land (approximately 15%). The Alterra report further concludes that the zinc input data for the Netherlands are high in comparison with EU countries having a less intensive agriculture activity. On the other hand they are shown to be representative for regions in North-Western Europe characterized by a similar, intensive agriculture.

In comparison with other types of land use, the local zinc load of rubber crumbs is relatively high and exceeds average zinc loads from agriculture. The amount of zinc from agricultural sources will partly be taken up by plants. The zinc load from rubber infill is not subjected to plant uptake. The effective input of zinc from agriculture that is available for distribution in soil and water is on average 50 mg/m^2/year. In comparison with the effective input of zinc from agriculture the zinc load from rubber crumbs is a factor 12-20 higher. In Appendix 6 the zinc balance in agriculture is presented for several soil types and land uses (data, De Vries et al., 2004)

Estimated exposure of drainage water, surface water, soil and groundwater.
The concentrations of zinc in soil, groundwater and surface water were assessed using simple equations and assumptions. Drainage water concentrations are estimated to vary from 1.1 to 1.6 mg/L. Further distribution of zinc depends on its mobility in the lava layer beneath the artificial turf. Although this material has a strong binding capacity for zinc when present in very small particles (<0.1 mm), the present gravel-like structure will minimise the possibilities for binding. Water will probably flow quickly to either the drainage system (on clay or peat) or the natural subsurface sand.

Zinc concentrations are diluted in surface water bodies, depending on the dimensions of the ditch. Typical small ditches are assumed to have a dilution factor of 5. An estimated surface water concentration of 220-320 µg/L (total zinc) was used for the risk assessment; in polder ditches the concentration increase is 10 times lower (dilution factor of 50). Concentrations in the groundwater may reach values approximately equal to that in the drainage water, because adsorption to sand is quite low. However, it can take some years before the zinc appears in the groundwater. Clay and peat are assumed to be not exposed to zinc because these soil types will have a surface drainage system. Concentrations in sandy soils could reach 1–188 mg zinc/kg soil.

Risk assessment
The estimated zinc load of 800 mg/m^2/year will result in exceedance of the critical load stated in the Building Materials Decree (2100 mg/m^2/100 year) within 3 years. One infill with a lifetime of 10 years will exceed the critical load by a factor of 4. Exceedance of critical loads implies potential risks to soil, surface water or groundwater. This is confirmed by the exposure assessment addressing the various 'receiving' compartments.

Drainage water concentrations (1.1-1.6 mg/L) exceed the legal criterion for surface water (MPC(total) = 40 µg/L) by a factor of 27-40. Drainage water concentrations will be diluted in the surface water, but in small ditches the MPA(dissolved) may be exceeded by up to a factor of 18 and MPA(total) by up to a factor of 11. A general rule in the waste water policy (CIW emission-immission impact assessment using the mixing zone model) is that only 10% of the

permissible concentration (= 4 μg/L) may be consumed by a particular source. This would imply that the exceedance factor in surface water is 45-80 in small ditches.

In sandy areas the emission to groundwater may exceed the Intervention Value by a factor 1.5-2.2. In sandy soils, infiltration of water with dissolved zinc will result in weak binding of zinc to the soil matrix and could cause the MPA to be exceeded by a factor of 12.

When rubber crumbs will be removed after 10 years and is replaced by materials with a lower zinc emission, the concentrations in drainage water and surface water will drop quickly. In soil and groundwater concentrations will decrease more slowly, due to the retardation of zinc in the soil. The concentrations calculated in this risk assessment are 'steady state' concentrations. This implies that concentrations do not increase further when emissions continue. However, the extent of environmental contamination will increase so larger bodies of groundwater, surface water and also sediment will become contaminated. The exceedance of environmental risk limits will not increase.

This risk assessment identifies that potential ecotoxicological risk in surface water, groundwater and soil may occur. It further indicates that application of rubber crumbs does not meet the critical load standards in the Building Materials Decree and does also not comply with criteria for the discharge of wastewater (CIW).

8.   Recommendations

Uncertainties in the risk assessment can be reduced by further studies. The following options are suggested:

Behaviour and ageing of rubber
Mechanisms of behaviour and ageing of (different types of) rubber should be investigated to obtain a better understanding of the risks of zinc and other components leaching from rubber. Better knowledge of such mechanisms should improve explanations of the behaviour of rubber during use and interpretation of the values measured in the laboratory and in field situations. It should also improve predictions of the risks of other or new types of rubber considered for use in artificial football pitches or other situations.

Monitoring drainage water
It is recommended that measurements are first taken in drainage water from existing artificial turf with rubber infill of differing age and quality. Sampling at several time intervals in different seasons is preferred. Besides measurement of zinc concentrations, records should be kept of weather conditions in the period before and during sampling, as well as registration of the pH and DOC of the drainage water. Parallel measurements of the leaching characteristics of the rubber crumbs by column tests is preferred. These experiments can verify the mobility of zinc in the lava layer and estimated drainage water concentrations.

Bioassay with drainage water
Bioassay is recommended to assess the toxicity of the drainage water. Bioassays show the effect of the total cocktail of leached substances as well as information on the effects of individual substances.

Semi field experiment (lysimeter)
A mini artificial turf field (1x1x1 m) can be built and exposed to outdoor weather conditions in a lysimeter. Water samples can be taken at different depths, preferably right below the lava rubber layer at 35 cm depth, and below the natural subsurface (a representative sandy soil) at 1 m depth. Besides measurement of zinc concentrations, records should be kept of weather conditions in the period of the experiment, as well as registration of effluent volumes, the pH and DOC of the drainage water. Parallel measurement of the leaching characteristics of the rubber crumbs in the lysimeter by column tests is preferred. Extrapolation of laboratory to field emission would be aided by measurement of zinc leaching from a control lysimeter, without rubber but with a known addition of soluble zinc. This experiment should give more insight into the emission and mobility of zinc under field conditions and its exposure to groundwater.

Modelling
When more and better information has been obtained, for example from the experiments mentioned above, more advanced models can be used for a refined risk assessment.

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Appendix 1 Installation of artificial turf

Overview of different steps in the construction of an artificial turf football pitch in Culemborg (the Netherlands). Pictures from: www.voetbal-clubs.nl/voetbal/vriendenschaar/

1. Removal of topsoil, 2. Removal of old drainage system, 3. Installation of new drainage system on natural subsurface and covering with sand, 4. Application of lava on sand, 5. Structure of the lava, 6. Application of artificial turf on a white filter layer, 7. Vehicle with rubber/sand mixture, 8. Application of rubber/sand mixture
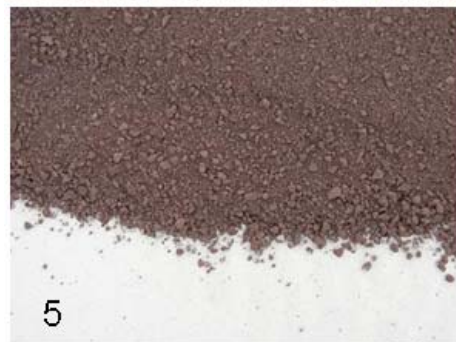
Appendix 2 Zinc content in rubber crumbs

Zinc content in rubber crumbs (mg/kg)

Sample I[1]	Sample II[2]	SampleIII[2]	Sample IV[2]	Sample V[2]
174	7500	7300	17000	9500

[1] LUT (2004); Westenberg and Máscik (2001)
[2] NBI (2004)

Zinc content in Dutch rubber crumb fresh production samples (Hofstra, 2006)

Sample	Origin	Type	Zinc content (mg/kg)
Product 1	Re-Tyre Lommel (B)	Car tyres	9400
Product 2	Granuband Amsterdam	Car tyres	8800
Product3	Rumal Weert	Truck tyres	19000

Appendix 3 'CIW Mixing zone model'

For this exposure assessment the increase in zinc concentrations due to drainage of artificial turf to a (standard) polder ditch of 5 m wide and 1 m deep with a water flux of 0.15 m^3/s has been computed.

Based on information of ISA Sport (Mr. Peterman, pers. Comm.), the effect of two types of drainage systems has been taken into account:
1. Drainage tubes in longitudinal direction with a diameter of 65 mm and 4 m distance from each other. So there are 18 discharge points over a length of 100 m.
2. The water of individual drainage pipes is collected in one larger drainage tube with a diameter of 160 mm. So in this case, there is one point of discharge.

For the discontinuous precipitation it is assumed that rainfall occurs 8% of the time.

In Figure 7 and Figure 8 the changing zinc concentrations are outlined as a function of the distance from the drainage pipe. Predicted increase in zinc concentrations in polder ditches due to leaching of zinc from artificial turf are shown in Table 6.

Table 6 Predicted increase in zinc concentrations (µg/L) in polder ditches due to leaching of zinc from rubber crumbs on artificial turf. The increase is computed at the end of the mixture zone (ΔC_L) which is at 50 m beyond the last drainage pipe

	C_0 = 1.3 mg/L	
Discontinuous precipitation 600 mm	**18 drains**	**one collection pipe**
ΔC_L (µg/L)	14.8	25.3
Dilution factor	90	53

Results of the 'CIW-Mixing zone model' show that the drainage by a collection pipe results in higher concentration than drainage by individual pipes.
Computations with the 'CIW-Mixing zone model' show dilution factors that are higher than the default dilution factor of 10. Based on the concentrations mentioned in Table 6 dilution factors of approximately 50-90 are computed.
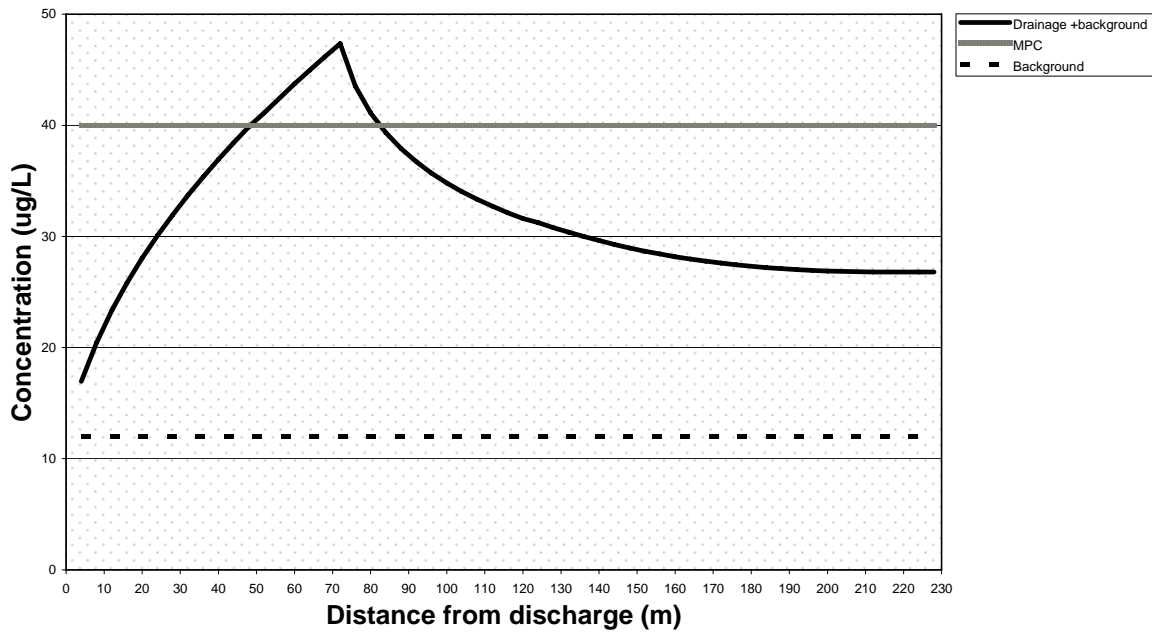
Figure 7 Concentration of zinc (µg/L) in a polder ditch as a result of drainage of artificial turf by 18 individual drainage pipes. Also shown are the MPC (total) and the background concentration of zinc in surface water
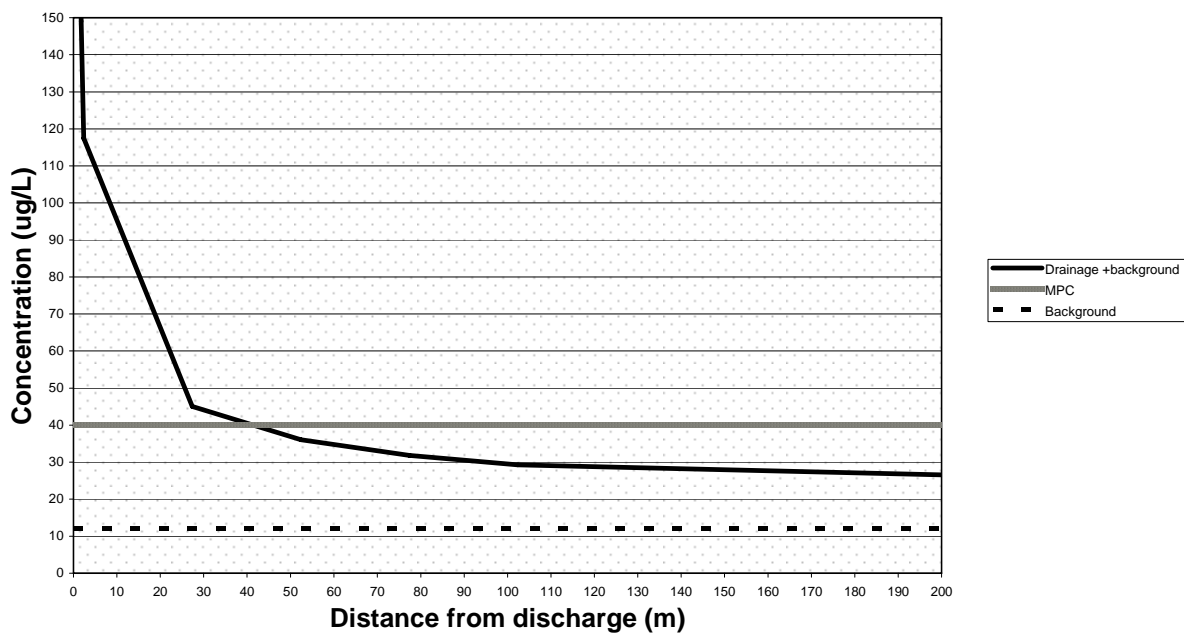
Figure 8 Concentration of zinc (µg/L) in a polder ditch as a result of drainage of artificial turf by a collection pipe. Also shown are the MPC (total) and the background concentration of zinc in surface water.

Appendix 4 Chemical composition of scoria (lava)

Chemical composition of scoria, used in the study of Kwon et al. (2005)

Components	Wt.%
SiO_2	49.84
Al_2O_3	14.07
Fe_2O_3	9.14
CaO	11.80
MgO	4.38
K_2O	1.77
Na_2O	2.18
TiO_2	2.04
MnO	0.30
P_2O_5	0.47

Appendix 5. Characteristics of EU regional waters for deriving BioFs

Characteristics (DOC, hardness and pH) of EU regional waters for which PEC/PNEC surface water exceeds one (without correction for bioavailability). Corresponding bioavailability factors (BioF$_{water}$) are calculated with Biotic Ligand Models for algae and fish. BioF$_{water}$ in bold represent the values that will be used in the risk characterization for, respectively, average (50P DOC and 50P inorganics) and realistic worst case (algae 10P DOC and 90P inorganics and fish: 10P DOC and 10P inorganics) conditions.(from: EC, 2006)

	DOC (mg/l)		pH			Hardness (CaCO$_3$ mg/l)			BioF algae 10-90	BioF algae 50-50	BioF fish 10-10	BioF fish 50-50
	10P	50P	10P	50P	90P	10P	50P	90P				
Meuse B/NL	1.9	2.6	7.4	7.6	7.9	143	205	244	**1.0**	**0.8**	0.7	0.5
German rivers [2]												
Aller	4.5	5.5	7.6	7.7	7.9	161	179	189	**0.6**	**0.5**	0.4	0.3
Elbe	4.8	5.5	7.5	7.8	8.3	308	415	512	**0.6**	**0.5**	0.3	0.2
Ems	6.7	7.1	7.3	7.5	7.8	223	253	297	**0.4**	0.3	**0.7**	0.4
Main	2.5	3.2	7.7	7.9	8.3	238	287	314	**0.9**	**0.7**	0.5	0.3
Mosel	2.4	3.2	7.6	7.8	8.1	325	409	510	**0.9**	**0.7**	0.3	0.2
Mulde	3.4	4.2	7.0	7.3	7.6	145	179	205	**0.7**	**0.5**	**0.7**	**0.5**
Oder	4.8	5.6	7.5	7.8	7.9	216	229	240	0.5	0.4	**0.6**	**0.4**
Saale	4.5	5.5	7.7	7.9	8.1	511	775	1042	**0.6**	**0.5**	0.2	0.1
Saar	3.4	4.0	7.6	7.8	8.0	228	277	322	**0.7**	**0.6**	0.5	0.3
Weser	3.8	5.0	7.6	7.8	8.1	490	750	1012	**0.8**	**0.6**	0.2	0.1
Flanders [1]	2.4	4.1	7.0	7.8	8.1	42	151	308	0.9	**0.6**	**1.0**	0.4
Flanders, add. DOC	9.3 (90P)		7.0	7.8	8.1	42	151	308	0.3 (90-90)	0.3 (90-50)	**0.6** (90-10)	**0.3** (90-50)
Walloon Region	1.7	2.9	7.1	7.8	8.3	31	134	341	**1.0**	**0.7**	1.0	0.5
NL large lakes	5,3	7,6	8,3	8,6	8,9	160	220	280	**0.5**	**0.3**	0.4	0.1
NL small lakes	4,1	9,9	7,6	7,8	8	160	220	280	0.6	**0.3**	**0.8**	0.3
NL streams/brooks	13,9	18,2	7,3	7,4	7,5	118	220	322	0.3	0.2	**0.6**	**0.2**
NL ditches	15,3	27,5	6,1	6,9	7,7	260	350	440	0.2	0.1	**0.6**	**0.2**
NL sandy spring	1,2	2,2	6,6	6,7	6,8	76	79	82	0.7	0.6	**1.0**	**1.0**
NL Rhine	2,1	2,8	7,7	7,8	7,9	201	220	233	0.9	**0.8**	**1.0**	0.4
NL Meuse	2,3	3,3	7,5	7,7	7,9	165	220	275	0.9	**0.7**	**1.0**	0.4
NL 'average'	6.3	10.2	7.3	7.6	7.8	163	218	273	0.4	0.2	**0.8**	**0.3**
France Rhin Meuse	2.0	3.4	7.5	7.8	8.0	168	248	312	**1.0**	**0.6**	1.0	0.5

[1] No specific data available. GEMS-A database is taken as substitute.

Appendix 6 Estimated zinc load (total input) from agricultural sources

Land use	Soil type	Zn flux (g.ha^{-1}.yr^{-1})			
		Input	Uptake	Leaching	Accumulation
Grass	Sand	938	700	228	10
	Sand calcareous	853	510	66	277
	Clay	969	474	34	460
	Clay calcareous	885	390	16	479
	Loess	1013	636	117	260
	Peat	889	455	126	308
Arable	Sand	1039	392	377	271
	Sand calcareous	868	319	86	463
	Clay	911	347	43	521
	Clay calcareous	899	238	19	642
	Loess	993	405	178	410
	Peat	836	317	271	248
All		926	425	152	349

(De Vries et al., 2004)