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Estimation of emissions and exposures to PFOS used in industry

A PFOS use inventory in metal plating and fire fighting

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An inventory of PFOS used in metal plating and fire fighting

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This investigation has been performed by order and for the account of VROM - Ministry of Spatial Planning, Housing and the Environment, within the framework of Socio-economic Analysis in REACH

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Abstract

Estimation of emissions and exposures to PFOS used in industry
An inventory of PFOS used in metal plating and fire fighting

By order of the Dutch Ministry of Spatial Planning, Housing and the Environment (VROM), the RIVM compiled an inventory of perfluoroctane sulfonate (PFOS) used in two specific applications. These are in use as mist suppressants/wetting agents in industrial chromium plating and as agents in fire-fighting foam. The primary aim of the inventory, with respect to the latter application, was to estimate existing stocks in the Netherlands.

In the Netherlands, the metal plating industry uses an estimated amount of 390 kg PFOS annually as a mist suppressant/wetting agent. The largest standing stocks of PFOS-containing fire-fighting foam are found at airports and chemical-industrial locations, such as petrochemical and paint manufacturing plants. The data compiled in the inventory were provided by suppliers.

The European Commission requested all Member States to compile an inventory of PFOS focusing on similar specific applications. The use of PFOS is restricted due to the risks it presents to both human and environmental health.

The methodologies used for data collection and analysis can be applied for other purposes, such as REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals).

Key words:
data collection, exposure estimation, release estimation, exposure methodology, PFOS, REACH, chemical use

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Rapport in het kort

Schatting van industriële emissies en blootstelling aan PFOS
Een inventarisatie van PFOS gebruik in de metaalbewerking en brandbestrijding

In opdracht van het ministerie van Volkshuisvesting, Ruimtelijke Ordening en Milieu (VROM) heeft het RIVM het gebruik in Nederland van de stof perfluoroctaansulfonzuur (PFOS) geïnventariseerd voor twee specifieke toepassingen. Het gaat om PFOS als nevelonderdrukker en bevochtigingsmiddel voor verchromingsdoeleinden, en om het gebruik ervan in blusschuim. Bij de laatste functie gaat het vooral om een schatting van de nog bestaande voorraden in Nederland.

In Nederland wordt per jaar naar schatting 390 kilo PFOS gebruikt voor deze toepassingen in de metaalbewerkingsindustrie. De grootste voorraden van blusschuim met PFOS zijn aanwezig op vliegvelden en op chemische industriële locaties, zoals verffabrieken en de petrochemische industrie. Het onderzoek is gebaseerd op gebruiksgegevens die leveranciers hebben aangeleverd.

De Europese Commissie heeft alle lidstaten van de Europese Unie gevraagd het gebruik van PFOS voor genoemde toepassingen te inventariseren. Vanwege de risicovolle eigenschappen van PFOS is het gebruik ervan aan banden gelegd.

De dataverzamelings- en analysemethodieken in dit rapport kunnen voor andere doeleinden worden gebruikt, zoals voor REACH. REACH staat voor Registratie, Evaluatie, Authorisatie en Restrictie van Chemische Stoffen.

Trefwoorden:
data verzamelen, blootstellingsschatting, emissieschatting, blootstellingsmethodologie, PFOS, REACH, stoffengebruik

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Summary

Directive 2006/122/EC of the European Parliament and the Council dated 12 December 2006 concerns the restriction of the placement on the market and use of perfluorooctane sulfonates (PFOS). This directive is part of Directive 76/769/EEC relating to restrictions on the marketing and use of certain dangerous substances and preparations. The latter will be replaced by Annex XVII of REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) starting on the 1st of June 2009. In Appendix I of the guideline 2006/122/EG it is mentioned that at the end of 2008 each EU Member State has to provide the European Commission with an inventory on 1) the amounts and releases of PFOS used as mist suppressants for non-decorative hard chromium (6^+) plating and wetting used in controlled electroplating systems and 2) existing stocks of fire-fighting foams containing PFOS. In reaction to this requirement, this document concerns the assessment of amounts and releases of PFOS and the size of foam stocks in the Netherlands.

Assessment of total stocks was done by using sales data and extrapolation based on market shares. Substantial effort was put into the data collection and many contacts were made with industry. The obtained information on sales volumes and use served as a basis to estimate emissions and stocks on national scale. Assumptions were made on the distribution of annual sales volumes, release and use. In addition, sales volumes were extrapolated to represent nation wide market quantities.

It is estimated that the worst-case maximum PFOS use for non-decorative hard chromium (6^+) plating is less then 0.4 ton per year (390 kg/year). Highest stored volumes of PFOS containing fire-fighting foam were found to be still present at airports and chemical industrial related locations being about 3300 m^3 and 3200 m^3 respectively, on a total of 18540 m^3.

The approach described in the current document is subject to uncertainty. More accurate estimations with known uncertainty can only be made on the basis of more data. Additional data of better quality are probably difficult to collect.

1 Introduction

Directive 2006/122/EG (Directive 2006/122/EC, 2006) concerns the restriction of the placement on the market and use of perfluorooctane sulfonates ($C_8F_{17}SO_2X$, of which X = OH, metal salt (O-M+), halogenide, amide and other derivates among which are polymers), further referred to as PFOS.

In Appendix I of the Directive 2006/122/EC it is mentioned that not later than 27 December 2008 each EU member state hands over to the Commission an inventory on:

- the amounts and releases of PFOS used as mist suppressants for non-decorative hard chromium (6^+) plating and wetting used in controlled electroplating systems and
- existing stocks of fire-fighting foams containing PFOS (i.e., foam present in firefighting storage equipment such as bottled foam and pipelines).

For the latter, it is important to bear in mind that existing fire-fighting foam that contains PFOS can still be used during a limited time frame in order to prevent possible further emissions into the environment. Foam that has been placed onto the market prior to December 2006 can still be used until 27 June 2011.

With this document the Netherlands fulfills the requirements mentioned in Directive 2006/122/EC. The approach to collect and analyze information and data used to estimate the required amounts and releases are described in the following chapters. The final estimates are summarized in chapter 5.

2 Method

Risk assessment information was collected to find information on the use and exposure of PFOS and PFOS related substances. The search strategy and information selection faced three stages being 1) literature search based on predefined terms and selection of a number of information resources facilitating a more focused search, 2) contacting branch organizations, (past) suppliers of PFOS for non-decorative metal plating, (past) suppliers of PFOS containing fire-fighting foam, and respective users on use quantities, 3) analysing literature and use data made available by industry facilitating quantitative estimation of emissions and stocks on a national scale.

2.1 Literature search

The first search criterion consisted of a selection of initial information resources most known to the authors to contain relevant information. Cross reference database search options offered by NLM's PUBMED, Scirus, European Commission and European Agencies' websites and institutional websites from WHO and RIVM were used. Information resources were screened by the use of one or a combination of the search terms 'chromium', '(non)decorative', 'environment', 'exposure assessment', 'fire-fighting foam', 'health', 'human', 'metal plating', 'mist suppressant', 'Perfluorooctane Sulfonates', 'PFOS', 'RAR', 'risk assessment', and 'workers'.
Collected information on the use of PFOS was split into two parts, i.e. one dealing with non-decorative material surface treatment processes with chromium (6^+) and one dealing with fire prevention and management.

2.2 Contacting branch organizations and suppliers

Branch organizations and associations allotting the specific domain of metal plating and fire-fighting applications were contacted for information on the quantities and releases of PFOS during metal plating with Chromium (6^+) and existence of stocks of PFOS containing fire-fighting foam in urban areas and important infrastructural locations such as petroleum industry, airports, harbors, railways, etc. Communication structures for obtaining data on the use of PFOS for metal plating and in fire-fighting foams are illustrated in Figure 1 and Figure 2, respectively.

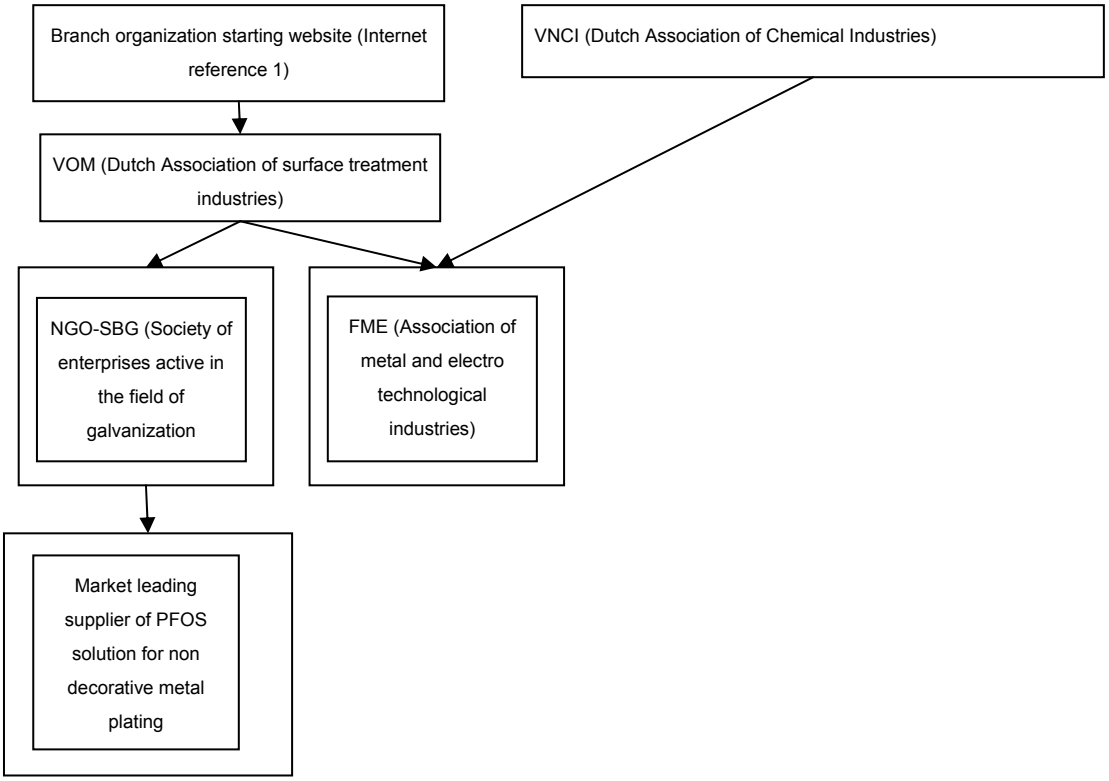

Figure 1. Communication structure used to obtain data on the use of PFOS in the metal plating industry.

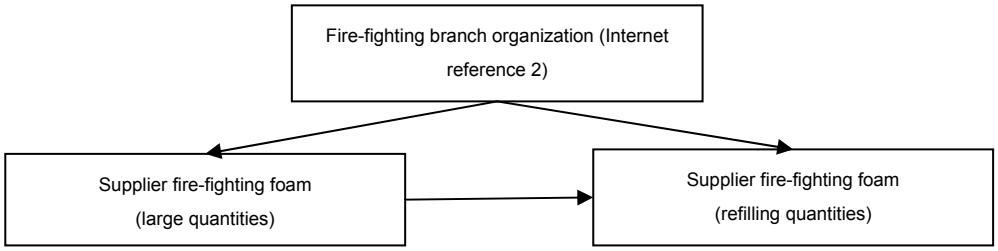

Figure 2. Communication structure used to obtain data on the use of PFOS in fire-fighting foams and existing stocks.

2.3 Analyzing literature and industry data to estimate releases and stocks

The data made available by past suppliers contained information on clients, locations and amounts. Data on PFOS in fire-fighting foam were stored in a for this task developed database (Microsoft Access, 2008). Calculations were done using Excel (Microsoft Excel, 2008) to estimate the total PFOS emissions during metal plating and PFOS containing fire-fighting stocks. The latter stock is presented as a total stock (national scale) and is - as a second step - categorized into stock estimations per branch. An in-depth description of the methodology with corresponding equations is described in Annex I. The results of the calculations are presented in the next chapter.

3 Results

This chapter describes the results obtained by applying the methodology described in Annex I. This chapter presents the results of estimated PFOS emissions into the environment from the metal plating industry and the estimates of existing stocks of PFOS containing fire-fighting foam per branch category.

3.1 PFOS emissions into the environment from the non-decorative metal plating industry

3.1.1 Process description

PFOS-related substances are used to lower the surface tension of metal plating solutions in order to prevent the formation of mists containing potentially harmful components from the baths. As such they are used in chromium plating, in anodizing and in acid pickling. Within the Netherlands, a major supplier of PFOS related substances for the Dutch metal plating industry supplied the substance tetraethylammoniumheptadecafluorooctanesulphonate (further referred to as TEA-PFOS) – a member of the PFOS family - as mist suppressant for the Dutch metal plating industry, more specifically for the non-decorative chromium surface treating industry. This substance is specifically selected for its resistance in a corrosive environment. Its use reduces the emission of aerosols formed during the surface treatment process from the chromium baths and remains sufficiently stabile in the corrosive solution within the chromium baths. The emissions of corrosive and carcinogenic material are minimized on the basis of economic analysis (Personal communication with a supplier of Fumetrol 140 on 29.07.2008). Contacted industry considered the use of an alternative substance, however, the bottleneck is the resistance against a corrosive environment caused by the chromium acidic electrolyte driving the stability of the mist suppressing property.

The substance TEA-PFOS is also being used for decorative chromium surface treatment applications. The market leader in the Netherlands announced that the use of PFOS for the production of non-decorative (and decorative) surface treatments will be terminated in 2008 for those applications that are not exempted as referred to in Directive 2006/122/EC. The market leader indicated the existence of a PFOS-free alternative for metal plating surface treatment applications.

3.1.2 Estimated amounts placed on the Dutch market

In 2006, the market leading supplier in the Netherlands placed 1560 kg of a solution (Fumetrol 140) containing TEA-PFOS (CAS: 56773-42-3, EINECS: 260-375-3) on the Dutch market for metal plating surface treatment mainly intended for the use of non-decorative purposes. The producer was located in Germany and therefore this solution was imported. On the basis of data provided on the Safety Data Sheet (No. 2096.6), this solutions contains 2.5-10% of the PFOS related substance TEA-PFOS. This equals to 39-156 kg. The market leader had a market share of 40-60% in 2006 in the Netherlands (data supplied by the supplier). It is assumed that this market share was similar in previous years.

3.1.3 Estimated releases into the environment

Aiming at the biggest range, this would equal to 39/60*100 - 156/40*100 = 65-390 kg/year of the substance TEA-PFOS that has been placed on the Dutch market. Assuming that 100% of the substance placed on the

market is released into the environment, the emission is estimated to be in the range of 65 – 390 kg/year. Given the limited data which has been provided, the worst-case approach is preferred. This resulted in a total yearly worst-case emission of 390 kg for the Netherlands (see Annex I Eq. 1).

3.2 Estimation of existing stocks of PFOS containing fire-fighting foam

3.2.1 Process description

Fluorised surfactants are used for specific fire-fighting applications. Surfactants need to quickly spread the water-based foam on top of burning hydrocarbon containing materials in order to avoid that the foam will arrive under the fire level. In this manner, the fire's oxygen supply is being blocked avoiding eventual burn-back. Both the surfactants as the foaming creating substances have to be stable under extreme conditions and have to resist the process of oxidation. Because the use of fire-fighting foam is limited to incidences foam products need to be long lasting and are being sold with a period of guarantee of 15-20 years. Industrial plants, areas and infrastructural sensitive locations like airports, tunnels and harbors, frequently contain storage locations of fire-fighting foam on PFOS basis in case of incidences.

3.2.2 Estimated amounts placed on the Dutch market

The market leader in the Netherlands placed between 1997 and 2002 in total more then 1 million kg of PFOS containing fire-fighting foam on the Dutch market. Sales volumes, however, decreased from 2001 onwards due to new legal requirements. As a first step, it was assumed that sales volumes are best represented by a lognormal distribution (having zero as an absolute minimum and infinity as a theoretical maximum (Ott, 1990). Consequently, on the basis of four sales volumes provided by the supplier (see Table 1) the annual mean, standard deviation and 95th percentile were found to be 239 m^3, 0.001m^3 and 309 m^3, respectively.

Table 1. Four consecutive sales volumes of PFOS containing fire-fighting foam placed on the Dutch market by the market leader.

Year	Sales volume (m^3)
1997	246
1998	212
1999	295
2000	212

As a next step, the 95th percentile is taken as a worst-case estimate of sales volume in the Netherlands. By assuming a shelf-life of 20 years – which is the maximum guarantee period – the total sales volume over a period of 20 years is estimated to be 20* 309 = 6180 m^3 of PFOS containing fire-fighting foam (see Annex I Eq. 2). The market leader had, however, an approximate market share of 25-50% in this period for the Dutch market (data supplied by the supplier). Assuming that the market share was 25% - and 75% of the fire-fighting foam was supplied by others -, this equals to a maximum of 6180/25 *100 = 24720 m^3 of PFOS containing fire-fighting foam that was placed on the Dutch market in 20 years time.

3.2.3 *Estimated existing stocks per branch*

It is estimated that 75 % of all stocks are still existing (expert judgment). This estimate is made by the market leading PFOS containing foam supplier and is based on the frequency of orders by clients to the suppliers after usage of foam, like in the case of an incident or fire-fighting training exercise. In reality the foam lasts longer then 20 years (Personal communication with a supplier of Fumetrol 140 on 29.07.2008). This translates into an estimate of 24720 m^3 * 0.75 = 18540 m^3 of PFOS containing fire-fighting foam that is still present in the Netherlands (see Annex I, Eq. 3).The rest of the foam (25%) is used. It is, however, unknown under which conditions the foam is used, i.e., under controlled conditions with assumed appropriate risk management measures, like collection and treatment of used foam in the case of an emergency exercise with negligible or low emissions into the environment, or uncontrolled with probably emissions into the environment like in the case of real incidences, like a fire.

The market leader also sold PFOS containing fire-fighting foam to smaller refilling distributors (assumed to include in the above estimate). One of the market leading smaller distributors, sold small (refilling) volumes PFOS containing fire-fighting foam throughout the Netherlands with an estimated market share of 40-60% for the period prior to 2002. The sales data were analysed by adding a category or branch label to each sale volume (see Table 2). Percentages of category sales were then calculated by dividing each category sales volume by the total sales volume of all branch categories (see Annex I, Eq. 4). The estimations of sales percentages of refilling PFOS containing fire-fighting foam are presented in Table 2 per branch category.

Table 2. Categorized estimation of sales volume percentages of refilling PFOS containing fire-fighting foam.

Branch category	**Percentage[1] (%)**
Aviation/airport	17.9
Industry of chemicals	17.5
Fire prevention/protection	11.2
Industry of coatings/paints	8.2
Ministry of defence/military purposes	8.1
Transportation, storage, and distribution of chemicals (containers,trucks,boats)	6.1
Engineering industry (construction, material and products)	4.9
Industry of petrochemicals	4.6
Waste industry	3.4
Food Industry	2.4
Industry of consumer products	2.2
Packaging industry	2.0
Car industry	1.8
Insulation producing industry	1.8
Public buildings, hospitals and universities	1.1
Other	6.8

1. Within this exercise, it is assumed that the sales data for a period of ten years is representative for a twenty year period.

Based on Table 2, it could be concluded that highest volumes were sold to aviation/airports, the chemical industries (and related transportation) and military applications. By combining the assumed volume of fire-fighting foam in the Netherlands (18540 m^3) and percentages per branch category (Table 2), the amounts of foam that is still present per branch is estimated as shown in Figure 3. Since the percentages as presented in Table 2 are based on a supplier with an approximately market share of 50%, this calculation is in fact an extrapolation of the derived supply distribution among industrial branch categories.

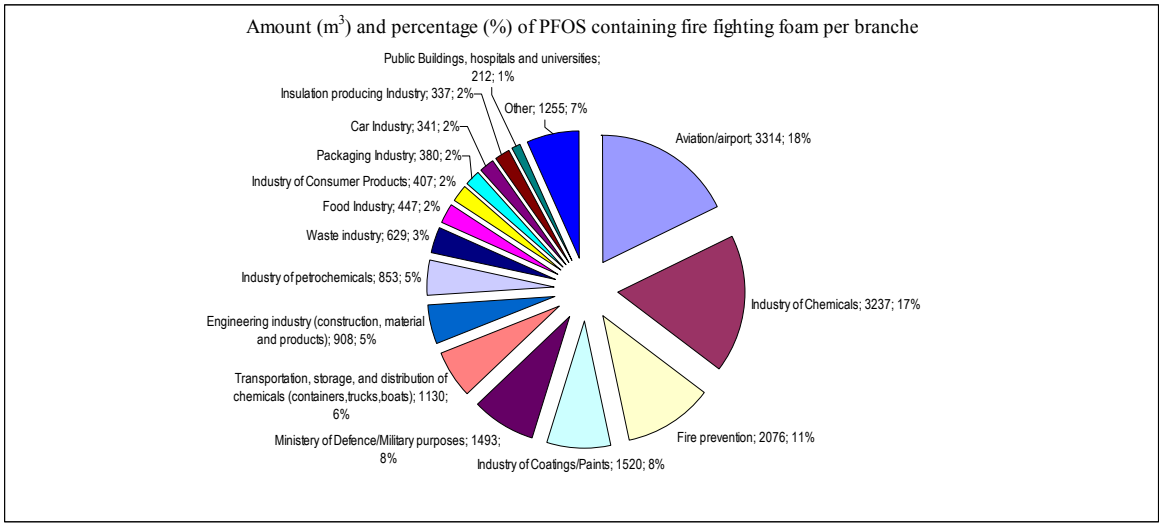

Figure 3. Estimated amount (m³) and percentage (%) of PFOS containing fire-fighting foam per branch category.

Highest volumes were found to be still present within the category aviation/airport – mainly existing of airports – of more then 3300 m^3 and the chemical industry of more then 3200 m^3. Percentages < 1% were added and categorized into the category 'other'.

4 Discussion

Assessment of emissions and stocks can be done using direct (measurements) and indirect (modeling) methods. Substantial effort was put into the data collection phase making many contacts with industry. The obtained information on sales volumes and use is essential to estimate emissions and stocks. In order to come up with estimates on a national scale assumptions were made on the distribution of annual sales volumes, release and use. In addition, sales volumes were upgraded to represent nation-wide market quantities. The approach described in the current document is, however, subject to uncertainty which is difficult to quantify. More accurate estimations with known uncertainty can only be made on the basis of more data. This process, however, is time consuming and additional data of better quality are difficult to collect.

4.1 Use and emissions from the metal plating industry

The emission of PFOS (TEA-PFOS) from the metal plating industry using chromium (6^+) is based on sales volume data provided by industry assuming 100% release into the environment. Industry indicated that the provided volumes are mainly used for non-decorative metal plating purposes. In the light of the worst-case approach used in this report, it is assumed that the provided data only accounts non-decorative purposes. Because of limited availability of sales data – only for the year 2006 - the assumption was made that the 2006 data are also representative for other years. In reality, sales volumes are subject to fluctuations and therefore this assumption introduces uncertainty to some extent. Also the 100% emission assumption introduces uncertainty. It is even unrealistic and it can be assumed that the emissions are in reality substantially lower, possibly being a fraction of this amount. The release is also estimated on a national scale being the Netherlands. In reality, however, emissions occur on the production locations. Without more data on the metal plating process this uncertainty can, however, not be quantified.

Other EU Member States, however, faced similar challenges and also made estimations of PFOS use within the metal plating industry on the basis of various assumptions. Within the UK the amount of PFOS has been estimated to be less then 0.5 ton per year (RPA, 2004). The Dutch estimated amount of about 0.4 ton (390 kg) per year is in the same order of magnitude and in general agreement with the British estimation. In terms of EU production and use of PFOS related substances, the total EU-wide estimated use quantities by the metal plating industry is between 8.6 and 10 tonnes per year (RPA, 2004).

Brooke et al. (2004) produced emission data of PFOS salt type solutions and assumed that due to its relatively simple formulation process – mainly existing of dilution processes - emissions only occur during their use. Local emissions were estimated to be 180 mg/day to waste water and 0.33 mg/day to air. Brooke et al. (2004) assumed that emissions only occur during 240 days per year resulting in an annual local emission of 43g/year to waste water and 79 mg/year to air. In addition to local emissions, regional emissions to waste water were estimated to be 1000 kg/year. The estimated worst-case Dutch environmental release – being a combination of emissions to all compartments (390 kg/year) – is larger than the local scale emissions, but smaller than the regional scale emissions used in the risk assessment made by Brooke et al. (2004). In EU risk assessment, local scale is commonly defined as the direct vicinity around the source (100-1000m) and regional scale as an area of 200 by 200 km (TGD Part II, 2003). Assuming that Brooke et al. (2004) used these specifications, the Netherlands' surface is larger than the local scale surface used by Brooke et al.

(2004), however, slightly larger compared to the regional scale surface. Solid quantitative conclusions can, however, not be made since both methods are too different.

Assuming that placement on the Dutch market equals the actual use and that no reactions occur of PFOS during the non-decorative chromium process, the assumption is that placement on the Dutch market equals environmental release. Environmental releases include emissions to air, water, soil, and waste. Emissions to air are expected to be low. Air concentrations during work on production sites of chromium plating were assumed to be 3.9 ng/m^3 according to Brooke et al. (2004). Although air concentrations are low, inhalation exposure cannot be excluded. In fact, Olsen et al. (2003) measured PFOS concentration in body serum of workers on a Perfluorooctanesulfonyl Fluoride Production Site of 0.941 ml/l (Geometric mean). Although the latter production site is different compared to chromium plating sites, occupational exposure and intake of PFOS via inhalation cannot be excluded for chromium plating workers. Most emissions are, however, expected to occur to waste water, mostly via the process of drag out (Brooke et al., 2004; OECD, 2004). How much of the PFOS can or is filtered out of the waste water via the application of risk management measures, like the placement of an active charcoal filter, is not known. Consequently, also emissions in the form of waste can not be excluded. Data reviewed within the frame of this task, however, is lacking specifications to assess compartment specific emissions. As a result, possible health risks related to industrial chromium plating cannot be indicated, however, can also not be excluded.

4.2 Assessment of national PFOS containing fire-fighting stocks

The existing stocks of PFOS containing fire-fighting foam in the Netherlands is based on sales volume data provided by two market leading suppliers, i.e., one supplier of large quantities and one supplier of smaller quantities mainly used for refilling purposes. Large quantity sales data covered multiple years (in total four) providing set of data to derive a sales volume distribution. This distribution is, however, subject to uncertainty and can only be improved by getting more (detailed) data on sales volume.

In the light of a realistic worst-case approach, the subsequent 95th percentile was taken as highest annual sales volume of PFOS containing fire-fighting foam and multiplied by the maximum guarantee period of 20 years. In this way the theoretical maximum stock was estimated. The Scientific Committee on Health and Environmental Risks (SCHER) assumed a use rate of 15% for estimating environmental releases from the use of PFOS containing fire-fighting foam (SCHER, 2004). On the basis of additional assumptions on the actual foam use - based on expert judgment – in this work a lower estimate based on data supplied by the PFOS fire-fighting foam supplier was made of the total stock of foam that still exists in the Netherlands, being 75%.

To enable additional estimations of branch specific stocks, detailed company specific sales data provided by the market leading refilling supplier were used to categorize sales volume percentages branch-wise. These percentages were then multiplied by the estimation of the total stock in the Netherlands to obtain branch specific stock estimations. Also this approach is subject to unknown uncertainty and can only become more realistic with detailed data.

Directive 2006/122/EC specifies that PFOS containing foam placed on the market prior to December 2006 can still be used until 27 June 2011. On the basis of 25% use of PFOS containing fire-fighting foam since 2000 until 2008, a period of 8 years, this translates to a use quantity of 24720-18540/8 = 772.5 m^3/year. Assuming similar use quantities per year this leads to a remaining stock in 2011, an additional 3 year

period, of 18540 – (3*772.5) = 16222.5m^3 that will still be present in the Netherlands. By multiplying this number with the percentages presented in Table 2, branche-specific estimations of the PFOS containing fire-fighting foam stocks can be made.

5 Conclusion

Directive 2006/122/EC of the European Parliament and the Council dated 12 December 2006 concerns the restriction of the placement on the market and use of PFOS. In appendix I of the guideline 2006/122/EC it is mentioned that at the end of 2008 each EU Member State has to provide the European Commission with an inventory on 1) the amounts and releases of PFOS and related substances used as mist suppressants for non-decorative hard chromium (6^+) plating and wetting used in controlled electroplating systems and 2) existing stocks of fire-fighting foams containing PFOS. In reaction to this requirement, this document concerns the assessment of consequent emissions and stocks in the Netherlands.

On the basis of information provided by industry and assumptions on the distribution of annual sales volumes, release and use, it is estimated that the maximum (worst-case) PFOS emission for non-decorative hard chromium (6^+) plating is less then 0.4 ton per year (390 kg/year). Highest stored volumes of PFOS containing fire-fighting foam, were found to be still present (annum 2008) at airports and chemical industrial related locations being about 3300 m^3 and 3200 m^3, respectively, on a total of 18.540 m^3.

The assessment described in the current document is considered as a best estimate for the described emissions and stocks bearing in mind that there is limited data available. More accurate estimations with known uncertainty can only be made on the basis of more data. Additional data of better quality are probably difficult to collect.

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6 Annex I. Theoretical concepts for the estimation of industrial PFOS emissions and stocks of PFOS containing fire-fighting foam

I.1 Estimation of PFOS emissions during metal plating

Data were made available by the market leading supplier on a PFOS containing solution for the metal plating industry using chromium (6^+). These data were used to estimate the total emission of PFOS into the environment according to Equation 1 (Eq. 1).

$$E_{PFOS} = \frac{V_{total} * C_{PFOS}}{f_{market}} * f_{release} \qquad \text{Eq. 1}$$

in which:

E_{PFOS} = Estimated maximum worst-case emission of PFOS into the environment [kg]
V_{total} = Total volume of solution placed on the Dutch market by the market leader [kg]
C_{PFOS} = Concentration of PFOS related substance in solution (%)
f_{market} = Fraction of Dutch market share based on the low end of the estimated range
$f_{release}$ = Fraction of release (unrealistic worst-case)

I.2 Estimation of PFOS containing fire-fighting stocks

Data that were made available by the market leading suppliers of PFOS containing fire-fighting foam were used to estimate of total volumes for several industry branches that still exist on the Dutch market. The estimation was made in three subsequent steps using specific equations.

I.2.1 Estimation of total volume that is still present on the Dutch market using data from the past market leading supplier of large quantities.

Estimation of total volumes of PFOS containing fire-fighting foam was based on data made available by the past market leading supplier of large quantities. The total volume was calculated on the basis of Equation 2 (Eq. 2), assuming a shelf life equal to the guarantee period of delivered foam.

$$V_{total} = \frac{V_{sale} * t_{guarantee}}{f_{market}} * f_{remaining} \qquad \text{Eq. 2}$$

in which:

V_{total} = Estimated realistic worst-case total volume that is still existing on the Dutch market [m^3]
V_{sale} = Estimated realistic annual worst-case sales volume (95th percentile) [m^3]
$t_{guarantee}$ = Maximum guarantee period representing maximum shelf life
f_{market} = Fraction of Dutch market share based on the low end of the estimated range (personal communication, 2008)
$f_{remaining}$ = Fraction of remaining fire-fighting foam (personal communication, 2008)

I.2.2 Estimation of the fraction of delivered refilling quantities per branch on the Dutch market using data from the market leading supplier of small quantities.

Fractions of delivered PFOS containing fire-fighting foam per branch were calculated by Equation 3 (Eq. 3).

$$f_{branch(i)} = \frac{V_{branch(i)}}{\sum_{i=1}^{n} V_{branch(i)}} * 0.01 \qquad \text{Eq. 3}$$

in which:

$f_{branche(i)}$ = Fraction of delivered refilling quantities per branch i on the Dutch market
$V_{branche(i)}$ = Refilling volume per branch [m^3]

I.2.3 Estimation of national amounts per branch combining data from market leading suppliers of large and refilling quantities.

By multiplying the total volume of foam placed on the Dutch market by branch fractions, the volumes per branch that still exist are estimated using Equation 4 (Eq. 4).

$$Q_{branch(i)} = V_{total} * f_{branch(i)} \qquad \text{Eq. 4}$$

in which:

$Q_{branche(i)}$ = Estimated realistic worst-case volume per branch that still exists on the Dutch market [m^3]
V_{total} = Estimated realistic worst-case total volume that is still existing on the Dutch market [m^3]
$f_{branche(i)}$ = Fraction of delivered refilling

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