An update of the lists with compounds that are relevant for the drinking water production from the river Meuse - 2015

Titel:	An update of the lists with compounds that are relevant for the drinking water production from the river Meuse - 2015
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Executive summary

The goal of this study is to identify compounds that are (potentially) relevant for the production of drinking water from the river Meuse. The existing lists of compounds that are (potentially) relevant for the drinking water function of the river Meuse are re-evaluated and updated based on water quality measurements carried out at nine monitoring stations and intake points along the Meuse in the period 2010-2014.

Before evaluating the compound lists, the criteria that were set by Fischer et al. (2011) for selecting the relevant and potentially relevant compounds were reviewed. This resulted in some adaptations of the methodology. The most notable changes are the renaming of the lists from C, B, A into 1, 2 and 3, the deletion of list B (list of potentially relevant compounds) , and the introduction of a new list to keep an overview of these compounds that were evaluated, but no longer fulfill the criteria for list 1 and 2. The following lists are now in use:

- List 1: Drinking water relevant compounds
- List 2: Candidate drinking water relevant compounds
- List 3: No longer drinking water relevant compounds

The compounds on the former list A, B and C were re-ranked using the adapted criteria. Based on i.e. their detection frequency, occurrence in concentrations above the ERM target value and their scores based on removal by water treatment, toxicity, odor/taste threshold and public perception, compounds were placed on either list 1, 2 or 3.
In the second part of the study, candidate compounds for the list of "drinking water relevant compounds" were identified based on monitoring studies described in literature, and measurement data from monitoring campaigns, e.g. obtained with screening techniques. This resulted in a list of 72 compounds. In case affordable measuring techniques are available for the potentially relevant compounds, they were included in list 2.

The adapted criteria and the literature and monitoring review resulted in the following lists:

List 1: Drinking water relevant compounds (including scores)					
Pharmaceuticals		**Pesticides**		**Industrial compounds**	
Ibuprofen	15	Desphenyl chloridazon	11	Acetone	11
Metformin + Guanylurea	29	DEET	10	DIPE	19
Metoprolol	10	DMS	11	DTPA	13
Paroxetine	16	Glyphosate + AMPA	11	EDTA	13
Sotalol	10	Isoproturon	16	Fluoride	>18
X-ray contrast agents		Nicosulfuron	11	NTA	19
Amidotrizoic acid	11	Terbuthylazine	16	**Plasticizer**	
Iohexol	12	**Polycyclic aromatic hydrocarbon**		DEHP	17
Iomeprol	12	Benzo(a)pyrene	18		
Iopamidol	12	**Hormone disturbing compounds**			
Iopromide	12	ER-Calux (bioassay for estrogenic activity)	27		

List 2: Candidate drinking water relevant compounds			
Pharmaceuticals[1]		**Industrial compounds**	**Hormone disrupting compounds**
4-FAA (metabolite metamizol)	Gabapentin	Benzylalcohol	Anti-AR CALUX
Propyphenazone	Lamotrigine	Aniline	GR-CALUX
Tramadol	Citalopram	Melamine	Bisfenol A
4-AAA (metabolite metamizol)	O-Desmethylvenlafaxine	Pyrazole[2]	**Pesticides/biocides**
Amoxicillin	Venlafaxine	Tert-butyl alcohol (metabolite MTBE)	3,5,6-TCP (chlorpyrifos + triclopyr metabolite)
Ciprofloxacin	Fluconazole	Urotropine	Metazachlor ethane sulfonic acid
Clarithromycin	Irbesartan		Metazachlor oxalic acid
Clindamycin	Telmisartan		Metolachlor ethane sulfonic acid
Erythromycin	Valsartan		Oxadiazon
Roxithromycin	Amisulpride		

[1] The compounds that are depicted in orange are selected for the RIWA monitoring programme of 2016
[2] Pyrazole has a known emitting source and it is proposed to monitor the compound only at the monitoring stations downstream from this source.

List 3: No longer drinking water relevant compounds			
Pharmaceuticals	**Pesticides**	**Industrial compounds**	**Plasticizers**
Aspirin	2,4-D	4-n-Nonyl phenol	BBP
Carbamazepine	BAM	Diglyme	DBP
Diclofenac	Carbendazim	Benzotriazole	DEP
Galaxolide	Chloridazon	BPS	DIBP
Lincomycin	Chlorotoluron	DMSA	N-butylbenzenesulphonamide
Naproxen	Dimethenamid	ETBE	TBP
Phenazone	Diuron	MTBE	TCEP
Salicylic Acid	MCPA	NDMA	TCPP
Sulfamethoxazole	Mecoprop	Surfynol 104	**Perfluorinated compounds**
Drugs of abuse	Metazachlor	Tolyltriazole	PFBA
Barbital	Methyl-desfenylchloridazon	**Fragrances and musks**	PFBS
Pentobarbital	Metolachlor	AHTN	PFHxS
Phenobarbital	**Artificial sweeteners**	Musk (ketone)	PFOA
Hormones	Acesulfame-K	Musk (xylene)	PFOS
Estrone	Sucralose		

Abbreviations

2,4-D	2,4-Dichlorophenoxyacetic acid
3,5,6-TCP	3,5,6-Trichloropyridinol
4-AAA	4-acetylaminoantipyrine
4-FAA	4-formylaminopyrine
ADI	Acceptable daily intake
AMPA	Aminomethylphosphonic acid
BAM	2,6-Dichlorobenzamide
BBP	Benzylbutylphthalate
BP-4	Benzophenone-4
BQ	Benchmark quotient
BRA	Brakel (RIWA monitoring station)
BTO	Joint Water Sector Research Programme for Dutch water companies (bedrijfstakonderzoek)
CALUX	Chemically Activated Luciferase Expression
C_{max}	Maximum concentration
CTGB	Dutch Board for the Authorization of Plant Protection Products and Biocides
DBP	Dibutyl phthalate
DEET	N,N-Diethyl-meta-toluamide (diethyltoluamide)
DEHPA	Di-(2-ethylhexyl)phosphoric acid
DEHP	Di(2-ethylhexyl)phthalate
DEP	Diethyl phthalate
DIBP	Diisobutylphthalate
DIPE	Diisopropylether
DMR	Danube, Meuse and Rhine
DMS	N,N-dimethylsulfamide
DMSA	Dimethylaminosulfanilide
DnBP	Di-n-butyl phosphate
DIPE	Di-isopropyl ether
DPhP	Diphenyl phosphate
DTPA	Di-ethylene-triamine-penta-acetic acid
EDTA	Ethylenediaminetetraacetic acid

EYS	Eijsden (RIWA monitoring station)
ER-CALUX	Estrogen Receptor Chemical Activated Luciferase gene eXpression
ETBE	Ethyl tert-butyl ether
EWM	Expertgroup Water quality Meuse
GR	Glucocorticoid Receptor
HEE	Heel (RIWA monitoring station)
HEU	Heusden (RIWA monitoring station)
HWL	Het Waterlaboratorium
ICPR	International Commission for the Protection of the Rhine
KEI	Keizersveer (RIWA monitoring station)
KWR	KWR Watercycle Research Institute
LC-HRMS	Liquid chromatography-high resolution mass spectrometry
LUI	Luik/Liège (RIWA monitoring station)
MCPA	2-Methyl-4-chlorophenoxyacetic acid
MCPP	Methylchlorophenoxypropionic acid (mecoprop)
MRI	Magnetic Resonance Imaging
MTBE	Methyl-tertiair-butylether
NDMA	N-Nitrosodimethylamine
NOAEL	No observed adverse effect level
NTA	Nitrilotriacetic acid
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutanesulfonate
PFHxS	Perfluorohexane sulfonate
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctanesulfonate
pGLV	Provisional drinking water guideline value
RIVM	National Institute for Public Health and the Environment
RIWA	Association of River Waterworks
STE	Stellendam/Scheelhoek (RIWA monitoring station)
TAI	Tailfer (RIWA monitoring station)
TBEP	Tris-(butoxyethyl)-phosphate
TBA	Tert-butyl alcohol
TBP	Tributylphosphate

TCEP	Tris(2-chloroethyl)phosphate
TCPP	Tris(chloropropyl)phosphate
TDCP	Tris[2-chloro-1-(chloromethyl)ethyl]phosphate
TDCPP	Tris(1,3-dichloro-2-propanol)phosphate
TDI	Tolerable daily intake
TEP	Tri-ethyl phosphate
TiBP	Tri-iso-butyl phosphate
TnBP	Tri-n-butylphosphate
TTC	Threshold of toxicological concern
UK	United Kingdom
USA	United States of America
UV	Ultraviolet light
WBB	Waterwinningbedrijf Brabantse Biesbosch
WHO	World Health Organization

Contents

1. Introduction

The rivers Rhine and Meuse are important for the Dutch drinking water production. To assure the quality of the river water and also the drinking water produced from these rivers, the Dutch association of river waterworks (RIWA) was established to monitor and protect the water quality of the Rhine and Meuse. One of the topics of concern for drinking water companies along the Meuse is pollution of the river by organic micro pollutants. Sources of pollution are for example waste water treatment plants (WWTPs), diffuse emissions by agriculture, but also industrial plants located near the river, and occasional dumping of chemicals near or in the river. New chemical compounds are continuously found in the river, partly because new compounds are introduced into the market, and partly because of the development or improvement of analytical techniques, which makes it possible to analyze additional groups of compounds or to achieve lower detection limits.

To be able to follow the development of pollutants in the Meuse, in 2007 RIWA Meuse started to compile lists of compounds that - based on a set of criteria - are considered relevant or potentially relevant to the drinking water function of the river Meuse (Van den Berg et al., 2007). Compounds on these lists should be measured in a common monitoring campaign of the drinking water companies along the Meuse. The lists were updated in 2011 (Fischer et al., 2011). A new evaluation of the lists is performed in this project.

Specifically, the goals in the project are:

- To re-evaluate the current lists of relevant and potentially relevant compounds for the drinking water function of the river Meuse based on the monitoring data in the period between 2010 and 2014.
- To review the current criteria of the compound lists and highlight potential issues of the current methodology, propose solutions, and provide a new set of criteria for the lists to resolve these issues.
- To propose new compounds for the list of potentially relevant compounds for the drinking water function of the river Meuse (the candidate compounds). These candidate compounds were derived from recent literature, measurement results, screening results, and monitoring campaigns from drinking water companies.

In the report, first the methodology is discussed: issues regarding the criteria that were set in 2011 are indicated together with the proposed solutions and the responding changes that were made to the criteria. Next, the results from the re-evaluation of the compounds based on the new criteria are discussed and an updated list with relevant compounds is proposed. Finally, a literature review on additionally emerging compounds is given. Based on this literature review and monitoring data, a selection of compounds that are considered as potentially relevant to the drinking water function is proposed for addition to the list of candidate compounds.

2. Available data

The measurement data of compounds was obtained from the RIWA Meuse database. This database is assembled using data provided by drinking water companies and water management agencies located near the Meuse (Figure 1). The monitoring stations are shown in Table 1.

Table 1. *RIWA monitoring stations located near the Meuse, in order of downstream appearance.*

Monitoring station/intake point	Abbreviation	Drinking water company/ water management agency
Talifer	TAI	Vivaqua
Namêche	NAM	Water-link
Liège/Luik	LUI	Water-link
Eijsden	EYS	Rijkswaterstaat Water, Verkeer en Leefomgeving
Heel	HEE	NV Waterleiding Maatschappij Limburg
Brakel	BRA	Dunea
Keizersveer	KEI	Evides NV/WBB
Stellendam/Scheelhoek	STE	Evides NV
Heusden	HEU	Dunea

To obtain the data necessary for calculating the score of drinking water relevant and candidate compounds the program EPI Suite 4.1[1] was used. It concerned the following parameters:

- The octanol/water partition coefficient Log K_{ow} as an indicator of polarity. The log K_{ow} was taken preferably from experimental data, but if experimental data was not available, the Log K_{ow} was estimated using the EPI Suite "KOWWIN v1.68 Log K_{ow} estimate".
- The vapor pressure of the compound as an estimate of volatility. The vapor pressure was obtained as experimental result from the EPI Suite database if such data was available, or estimated using the "mean vapor pressure of Antoine & Grain methods" in EPI Suite.
- The biodegradability was derived from estimations using the BioWIN3 Ultimate Survey Model in EPI Suite.

[1] The program EPI Suite 4.1 is available on the website of the US environmental protection agency: http://www.epa.gov/oppt/exposure/pubs/episuite.htm

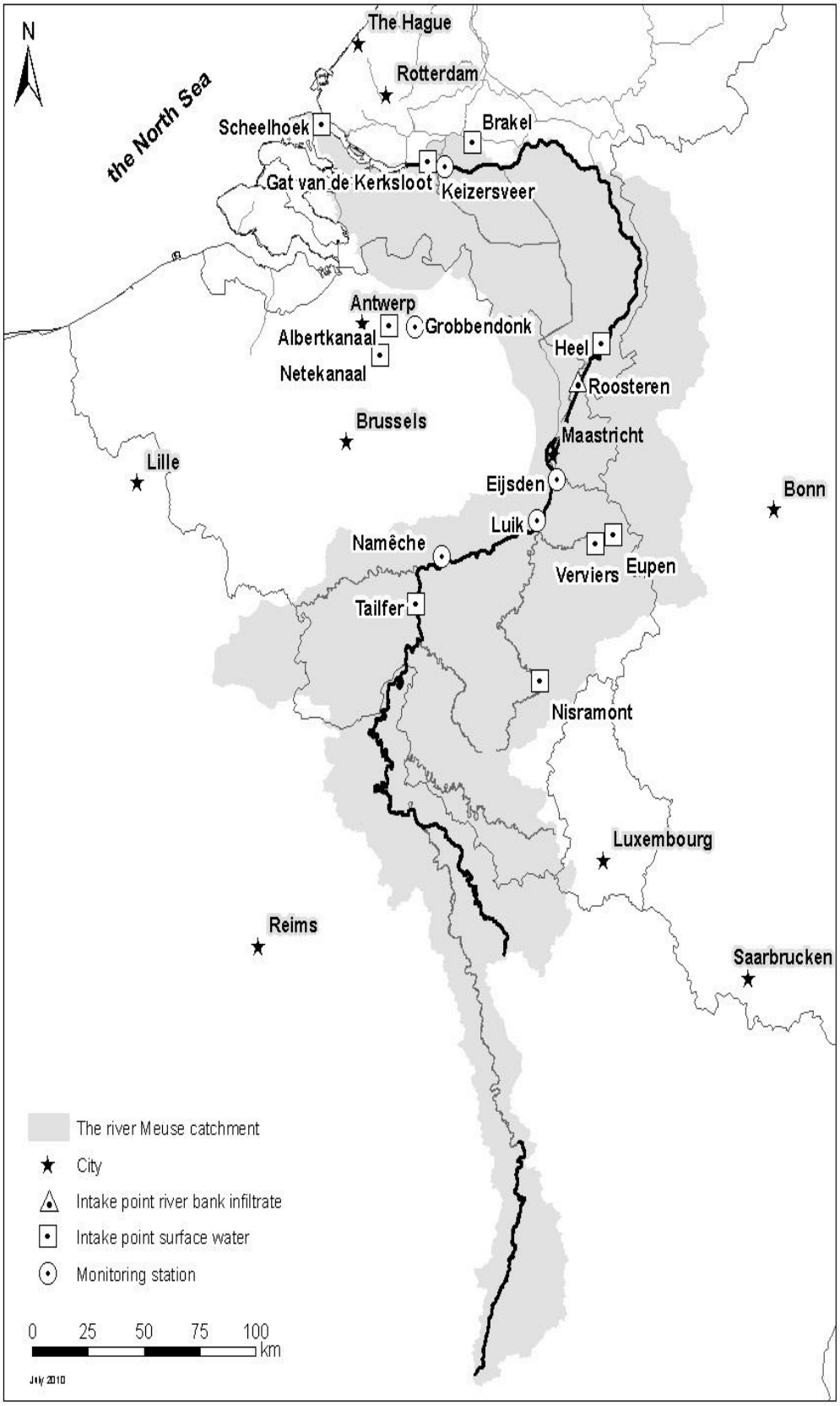

Figure 1. *The Meuse catchment area with the RIWA monitoring stations and intake points.*

To calculate the benchmark quotient (BQ) as an estimate of toxicity, the provisional guideline value (pGLV) was used. Most pGLVs were taken or calculated from the following sources:

- Guidelines for Drinking-water Quality, Fourth Edition (World Health Organization, 2011).
- Toxicological relevance of emerging contaminants for drinking water quality (Schriks et al., 2010).
- Geneesmiddelen in drinkwater en drinkwaterbronnen. Resultaten van het meetprogramma 2005/2006 RIVM (Versteegh et al., 2007).

The pGLVs that were not found in the aforementioned articles were found in specialized risk assessment reports of the specific chemical (see Appendix 4).

A literature study was carried out to select candidates for the list of drinking water relevant compounds. Publicly available journals found through Google Scholar and a search of ScienceDirect, were used as sources. Keywords that were used included "emerging compounds", "emerging substances", "emerging pollutants". Reports published by KWR Watercycle Research Institute (KWR), that performs joint research studies for the Dutch drinking water companies (bedrijfstakonderzoek - BTO), and RIWA-Meuse were also used. Toxicological information about pesticides was obtained from the site of the Dutch Board for the Authorization of Plant Protection Products and Biocides (CTGB). Extra attention was given to publications from 2011 or later, since the previous RIWA Meuse report was written before this time.

3. Criteria for ranking compounds which are relevant for drinking water production

3.1 RIWA lists of compounds

RIWA Meuse introduced a system of lists of drinking water relevant compounds in the river Meuse in 2007. The compounds on the lists should be measured in a common monitoring campaign of all drinking water companies along the Meuse. In 2011 the methodology was updated which resulted in three different lists of drinking water relevant compounds (Fischer et al., 2011). The three lists, in order of decreasing measuring frequency and importance to the drinking water companies, were as follows:

- List C: The list of drinking water relevant compounds. Compounds on this list were measured every four weeks (13 times a year). Compounds were placed on this list when they had been measured for several years, and found to be relevant enough through a ranking method (see Appendix 2).
- List B: The list of potentially drinking water relevant compounds. Compounds on this list were measured every four weeks (13 times a year). Compounds were placed on this list if they had been measured for 2 years and either belonged to a group that negatively influences the public perception of drinking water e.g. pharmaceuticals or pesticides, or if measured concentrations broke the odor/taste threshold, or were higher than 10% of the provisional guideline value (a benchmark quotient of 0.1). The compounds on this list were further monitored to evaluate if they were relevant to the production of drinking water, before being put on list C. This list was added in 2011 as an intermediate stage to provide emerging compounds of concern equal opportunity to be detected as compounds on list C.
- List A: The list of future potentially drinking water relevant compounds. Compounds on this list were measured every three months (4 times a year). Compounds were placed on this list if they had been found to be potentially relevant either through literature research, monitoring data of surface or drinking water, or if their usage was expected to increase in the near future.

More detailed descriptions of the compound lists and their respective criteria from 2011 can be found in Appendix 2 and Appendix 3.

3.2 Issues and proposed solutions for the methodology

Logically, the experience with the use of the methodology and the compound lists of 2011 brought some new insights and some issues were disclosed that could be improved. These issues are mentioned in this chapter, along with the proposed changes to the methodology.

3.2.1 General issues

The following issues are encountered with the methodology set in 2011:

- The names of the lists, A, B, and C, were in the order a compound moves along the flow scheme: new, potentially relevant compounds are put on list A, and if more strict criteria were met, compounds could end up on list C with relevant compounds. However, in practice, this was confusing, since list A was often intuitively expected to contain the most important compounds. The names of the list were therefore changed to an order in which the list of relevant compounds is first in sequence. The option whereby the names for list A and list C are swapped was rejected because this could cause confusion in the future if "list A" can refer to either the old list A, or the new list A. To prevent this, the new lists are ordered numerically, with 1 being the list of relevant compounds (formerly list C).
- The term "observed" is replaced by the more precise term "detected".
- The Danube-Meuse-Rhine (DMR) memorandum is replaced by the European River Memorandum (ERM) (IAWR et al., 2013).

3.2.2 List 1 (former list C) – Relevant drinking water compounds

In list 1 the following changes are made:

- The exact degree of compound removal through water treatment mentioned in the criteria is unclear. Since the scoring system also takes expected removal through water treatment in consideration, the criterion is considered redundant and is therefore removed.
- The time interval described in the criteria refers to specific years. This is changed to relative years instead. This prevents the criteria of the lists from needing updating of the years every time the compound lists are re-ranked.
- The percentage of minimum detections out of all measurements is changed from 8% to 7% to avoid problems with rounding. The equivalent of one detection out of 13 measurements is 1/13 = 7.69% which, when not rounded, is below 8%. At a percentage of 7% a frequency of 1 out of 13 will still be above this minimum regardless of rounding.
- The last criterion for list 1 states that the total score of a compound has to be 10 or higher. One of the elements that determine the total score of a compound is removal by water treatment. A compound receives a score between 1 and 3 for the parameters polarity, volatility and biodegradability. Since it is not desirable to add compounds to the list of relevant drinking water compounds that are easily removed, additionally it is added that the minimal score for removal should be 4.

3.2.3 Removal of list B

List B is removed from the prioritization methodology for the following reasons:

- In practice it turned out that it is easier to have two compound lists: one with relevant compounds and one with candidate compounds. The in-between list B was more difficult to explain to other institutes and often confusing.
- Applying the ranking methodology from 2011 would result in a list B with less than 5 compounds, which also favored the decision to transfer these compounds to the list of candidate compounds.
- Most criteria are also considered in list 1 which often makes it possible to directly evaluate compounds based on the criteria in list 1 and skip the criteria for list B:
    - The compound having a benchmark quotient above 0.1, and the compound having a negative effect on public perception is indirectly included in the criteria of list 1 through the scoring system.
    - Whether the compound is easily removed, and if the compound is harmful for the public perception of drinking water is also included through the scoring system.
    - The criterion for removal during treatment is repeated almost exactly in the criteria of list 1.

3.2.4 List 2 (former List A) – Candidate compounds

A criterion is added which specifies that the measuring technique has to have a sufficiently low limit of quantification to accurately monitor if the compound is below or above target values.

3.2.5 Addition of list 3 – No longer drinking water relevant compounds

The evaluation of the compound lists from 2011 resulted in a list of compounds that based on the new monitoring data did no longer fulfill the criteria. To make sure that the information with regard to the evaluation of these compounds (e.g. the pGLV-values and the reason why the compound did not make it to list 1) will be secured, a new list was created for sufficiently evaluated compounds that do not or no longer fulfill the criteria of list 1 or list 2.

Drinking water companies can decide if they want to continue measuring the compounds on this list, for reasons of their own.

3.3 New criteria of the lists of compounds

In summary, based on the above mentioned changes three new lists are established:

- List 1 – Drinking water relevant compounds
- List 2 – Candidate compounds
- List 3 – No longer drinking water relevant compounds

The updated criteria for each of the new lists are shown below. Red text indicates a change from the criteria in 2011. A flow scheme demonstrating how the criteria should be applied is shown in Figure 2.

3.3.1 List 1: Drinking water relevant compounds

The main list of relevant compounds, list 1, is shown below. This list was based on the criteria of the list of relevant compounds of 2011 (see Appendix 2).

Criteria for List 1: Drinking water relevant compounds

1. The compound was detected at two or more RIWA Meuse monitoring stations or intake points in the last 5 years (for a minimum of two years*), with a frequency of at least 7% of the measurements*.

   and
2. The compound was found to exceed ERM target values or the Drinking Water Standards from the Dutch Drinking Water Regulation on at least two different RIWA Meuse monitoring stations or intake points in the past 5 years (taking into account possible removal by conventional treatment), with a frequency of at least 1% of the measurements.

   and
3. The compound was found to exceed the drinking water standard or the ERM target value used by the drinking water companies, at least once in the past 3 years

   and
4. The total score of the compound has to be 10 or higher, of which at least 4 points are awarded by compound removal (sum of polarity, volatility, and biodegradability points).

If the benchmark quotient of the compound is 1 or higher, the compound is considered drinking water relevant and criteria 2, 3, and 4 can be neglected.

* If the compound is monitored more than 13 times per year, it has to be detected at two or more RIWA Meuse monitoring stations with a frequency of at least 7% of the measurements per year. This criterion is equivalent to the criterion requiring that the compound with a monitoring frequency of 13 times a year, is detected at least once a year.

3.3.2 List 2: Candidate compounds

The list of candidate compounds, list 2, is shown below. This list was based on the criteria of the list of potentially relevant compounds of 2011 (see Appendix 2).

Criteria for List 2: Candidate compounds

1. The compound is present in the river Meuse at concentrations well above the ERM target value
   or
2. The concentration of the compound is expected to increase due to increased use in the catchment area in the near future (e.g. due to a change in usage of pesticides) (based on expert judgement)
   or
3. The compound has undesirable properties for the production of drinking water and is expected to be present in the river Meuse (based on research),
   and
4. The compound can be monitored with an affordable measuring technique with a reasonable limit of detection

3.3.3 List 3: No longer drinking water relevant compounds

The newly added list 3 is shown below:

Criteria for List 3: No longer drinking water relevant compounds

Former list 1 and 2 compounds which have not been found to fulfill the criteria of list 1 in the past 5 years.

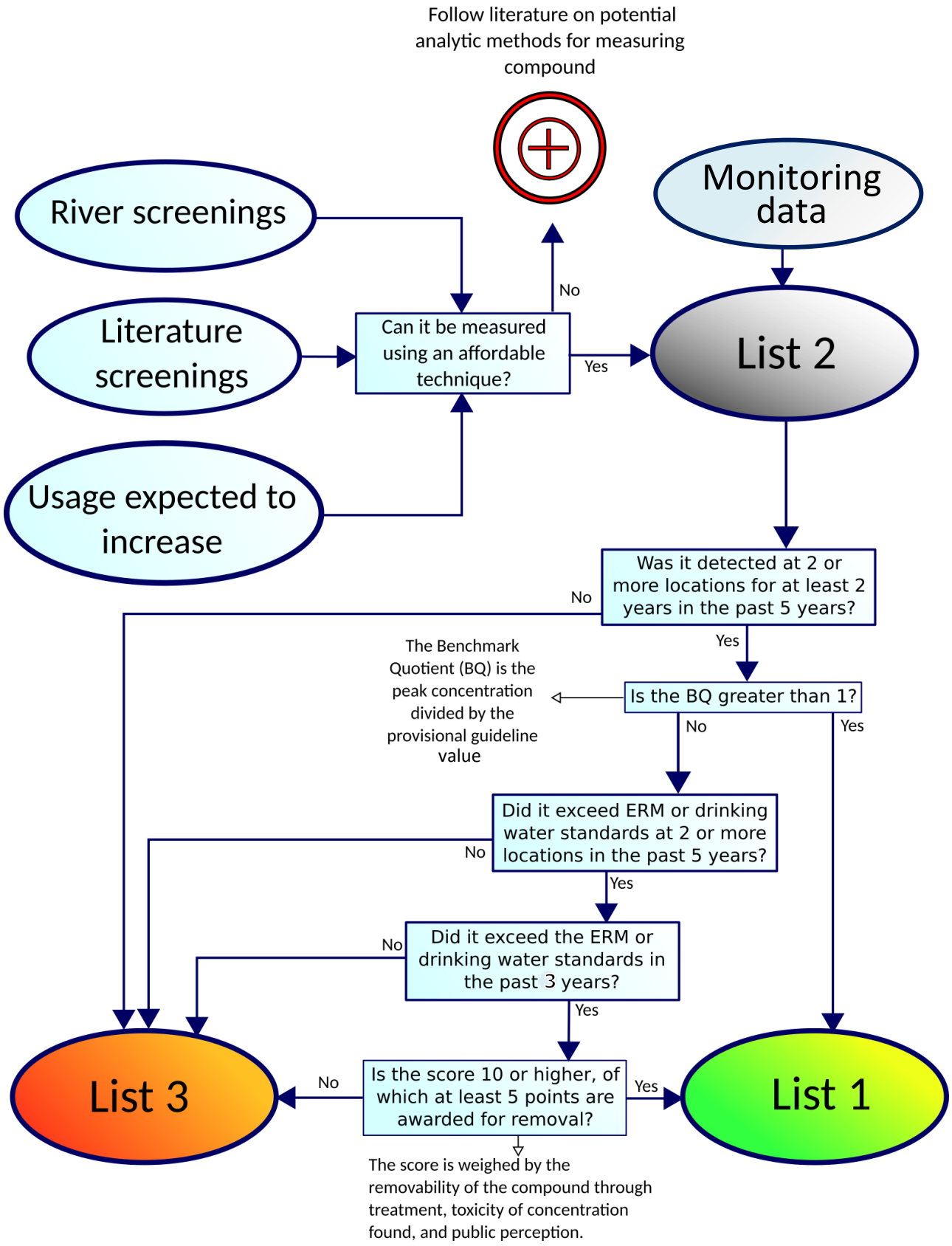

Figure 2. *Schematic overview of the updated criteria.*

3.4 Ranking methodology

3.4.1 Measuring and placement of compounds on the lists

Some compounds are not part of any lists of the RIWA Meuse monitoring programme, but were measured for 5 years or more at monitoring locations along the Meuse. When enough monitoring data were present to determine if these compounds fulfill the criteria of list 1, they were evaluated in this project. In case they passed the criteria, they were added directly to list 1. If not, they remain unlisted.

List 1: Drinking water relevant compounds — monitored 13 times a year for 5 years:
If a compound is placed on the list of drinking water relevant compounds (list 1) it will stay there for at least five years. When it has been measured the required 13 times per year for each of the 5 years, but is no longer found to fulfill the criteria of list 1, it will be moved to the list with no longer drinking water relevant compounds (list 3). If the compound does meet the criteria of list 1, it will remain on list 1.

List 2: Candidates for the list of drinking water relevant compounds — monitored 13 times a year for 1 year:
If a compound is placed on the list of candidate compounds that are potentially relevant for drinking water (list 2), it will be monitored 13 times during one year. After this year, it will be evaluated if the compound is likely to fulfill the criteria of list 1. If it is clear that the compound is not going to fulfill the criteria of list 1, it is taken off list 2, and it will be moved to the list with no longer drinking water relevant compounds (list 3). An exception to this rule is the situation where the use of the compound is expected to increase, and it is assumed that it will pass the criteria of list 1 in the near future due to increased use: in this case the compound will be kept on list 2. If it is not possible to evaluate if the criteria of list 1 will be met, the compound stays on list 2. If the criteria of list 1 will clearly be met, the compound is moved to list 1.

List 3: No longer drinking water relevant compounds — need for monitoring decided by drinking water companies:
Compounds on this list may be monitored at a frequency that can be decided by the drinking water companies individually. Reasons to continue measuring compounds are for example a legal obligation or the compound receives negative attention in the news, and drinking water companies want to be able to answer potential questions from users.

3.4.2 Scoring of drinking water relevant compounds

To rank compounds in order of increasing relevance for the drinking water function of the river Meuse, the compounds were scored based on following properties:

- Removal by water treatment (polarity, volatility, biodegradability).
- Toxicity (benchmark quotient).
- Odor/taste threshold.
- Public perception.

The scoring system is described in Appendix 1, and explained in detail in the 2011 RIWA Meuse report (Fischer et al., 2011).

For the calculation of a benchmark quotient the maximum concentration in the surface water is compared to a provisional drinking water guideline value that is based on toxicity data. Sometimes, the monitoring data contains outliers, meaning that the maximum concentration can be considerably higher than the 90th percentile concentration. To see if using a percentile concentration instead of a maximum concentration for the calculation of the benchmark quotient and odor/taste threshold would have a big impact the benchmark quotient, the differences were compared. It was found that using a 90th percentile concentration instead of a maximum concentration to calculate the benchmark quotient did not affect the placement of any of the compounds. It was therefore decided not to change the methodology and use the maximum concentration for the scoring of the compound.

3.4.3 Coupling of metabolites with parent compounds

Some compounds are highly resistant to degradation and remain in the environment for years without breaking down. These compounds typically remain in the water cycle unless they are removed by water treatment (Jones and de Voogt, 1999). Other compounds may be broken down in the environment through a variety of pathways, such as hydrolysis, photolysis, or enzymatic degradation by microbiological organisms (Alfano et al., 2000; Leahy and Colwell, 1990; Mabey and Mill, 1978). For some compounds the resulting breakdown products are toxic, in some cases even more toxic than their parent compound (Benitez et al., 2013).

In 2011 the pesticide chloridazon was placed on the list of drinking water relevant compounds; list C. Further evaluations showed that desphenyl-chloridazon, a metabolite of chloridazon, was detected at high concentrations at three of the monitoring locations. It was decided to also add desphenyl-chloridazon to list C, despite it did not fulfil the criteria for this list (because of a lack of monitoring data). This raised the question whether it is desirable to couple parent compounds and their metabolites together during the re-ranking of the lists. Further examples of compounds that can potentially be coupled to their metabolites are found in literature and previous measuring results from RIWA:

- Amino-methyl-phosphonic acid (AMPA) is a metabolite of glyphosate, a common herbicide. AMPA has been observed in the river Meuse at concentrations well above ERM target values since 2004 (RIWA-Meuse, 2013). It should be noted that AMPA is also a metabolite of other phosphonates. In 2010 large concentrations of AMPA were discovered in a branch of the river Meuse, caused by degradation of cooling water phosphonates originating from nearby industrial plants (RIWA-Meuse, 2013).
- Metformin, an anti-diabetic medication, was detected at high concentrations (0.3 µg/L) between 2010 and 2011 in the river Meuse and was added to the list of future potentially drinking water relevant compounds in 2011 (Fischer et al., 2011). Guanylurea, a metabolite of metformin, is formed during the treatment of metformin containing waste water (Scheurer et al., 2009). Guanylurea was added to

measurement campaigns since guanylurea concentrations exceeding ERM target values were observed in 2013, during a large scale study of pharmaceuticals in the water cycle in the Dutch province Limburg (RIWA- Meuse, 2013).

- Carbamazepine, an anticonvulsant medication, has been found in wastewater effluent in concentrations up to 0.17 µg/L. Its metabolite carbamazepine oxide has been found in the same effluent in much higher concentrations of up to 2.3 µg/L (Huerta-Fontela et al., 2010).
- N,N-Dimethylsulfamide (DMS) is a metabolite of the fungicide tolylfluanid. Tolylfluanid is not detected in the river Meuse, but DMS has been detected occasionally (RIWA- Meuse, 2013).

If a compound degrades quickly, it is difficult to observe trends of usage or appearance from measuring the concentration of the parent compound alone. Measuring metabolites along with their parent compound allows more accurate monitoring of the use and appearance of some compounds. In addition, some metabolites are equally or more toxic than their parent compound (Cleuvers, 2003; 2004). This means that if the parent compound degrades, this does not guarantee a lower toxicity. Some metabolites and transformation products of pharmaceuticals have reached the status of compounds of emerging concern (Michael et al., 2014). In the Netherlands, Belgium and France, the same regulatory standards apply for as well the parent compounds as the metabolites of pesticides and biocides. In the Netherlands, an exception is made for metabolites that are considered non-relevant: they have a higher regulatory standard of 1 µg/L instead of 0.1 µg/L (RIWA Meuse 2013). The European River Memorandum (ERM) also maintains the same target values for metabolites as they do for their parent compounds.

It could also help to have both the monitoring data of the parent compound and its relevant metabolite available to be able to demonstrate that the use of a certain parent compound causes problems when it degrades in a persistent metabolite.

Currently metabolites are not coupled with their parent compound when the lists are re-ranked. It is suggested that this should be changed. It is proposed that, when the lists are re-ranked, all compounds have to be checked, scored, and ranked independently. Then, if any compounds on list 1 have metabolites or parent compounds that are on list 2, these metabolites or parent compounds should be placed on list 1 as well.

Since compounds can have countless metabolites, it is not realistic to try to discover and measure every single metabolite. Metabolites or parent compounds should only be monitored alongside their respective metabolite or parent compound, if they have been found to be present in the Meuse through monitoring data, or if literature indicates that these compounds may be present in surface or drinking water. In practice, this means that only metabolites and parent compounds present on list 2, are eligible for coupling with list 1 metabolites or parent compounds.

4. Proposed drinking water relevant and candidate relevant compounds

The lists of drinking water relevant compounds (list 1) was re-ranked using its updated criteria. For this purpose, monitoring data from the period 2010-2014 were used. The new list of no longer drinking water relevant compounds (list 3) consists of all compounds that were tested on the criteria of list 1, but did not meet them. Finally, the list of candidate drinking water relevant compounds (list 2) was proposed based on literature study and measurement data.

4.1 List 1—Drinking water relevant compounds

Not only compounds from the former RIWA lists were tested on the criteria of the list 1, but all compounds that were measured for five years under the precondition that the necessary parameters were available to calculate the score. This resulted in the inclusion of six new compounds in list 1 that were not on any RIWA list before. An example is paroxetine. Although this compound was measured at only four monitoring locations, based on the measurement data from these locations, the compound fulfilled the criteria for list 1.

From the compounds from the former list B that were tested based on the criteria of list 1, fluoride and the ER-CALUX had a benchmark quotient (BQ) higher than 1 (criterion 1). These compound also fulfill the criteria of list 1 when the criterion of the BQ is disregarded. From the list 1 (C) compounds, none was found to have a BQ of 1 or higher. From the new compounds, guanylurea was found to have a BQ above 1. For this compound a pGLV could not be determined and the toxicological threshold concentration (TTC) was used instead. The TTC principle is based on toxicity predictions using low thresholds, to ensure that toxicological risk is small. Guanylurea will be reviewed in the future when an acceptable/tolerable daily intake value can be derived from toxicological data.

One of the compounds on list 1, glyphosate, has received bad publicity in the media after the World Health Organization (WHO) published a report on glyphosate in March 2015, changing the status of glyphosate to "possibly carcinogenic". Concentrations detected in surface water were frequently above 0.1 µg/L (C_{max} of 0.66 µg/L with a BQ of 0.001) and it is recommended to pay extra attention to this compound and review the provisional guideline value (pGLV) of this compound in the future should new information become available.

Table 2. *Proposed compounds relevant to the drinking water function of the river Meuse (list 1), scored based on removal by water treatment, toxicity, odor/taste threshold and public perception.*

#	Compound	CAS #	Category	Score[1]	Previous list
1a	Guanylurea (metabolite metformin)	141-83-3	Pharmaceutical	29	Unlisted
1b	Metformin	657-24-9	Pharmaceutical	17 (29)	A
2	ER-Calux (expressed in 17ß-estradiol equivalents)	-	Hormone disturbing compounds	28	B
3	DIPE (di-isopropyl Ether)[2]	108-20-3	Industrial Substance	19	C
4	NTA	139-13-9	Industrial Substance	19	Unlisted
5	Fluoride	16984-48-8	Industrial Substance	>18	B
6	Benzo(a)pyrene	50-32-8	Polyaromatic hydrocarbon	18	C
7	DEHP	117-81-7	Plasticizers	17	A
8	Isoproturon	34123-59-6	Pesticide	16	C
9	Paroxetine	61869-08-7	Pharmaceutical	16	Unlisted
10	Terbuthylazine	5915-41-3	Pesticide	16	Unlisted
11	Ibuprofen	15687-27-1	Pharmaceutical	15	B
12	DTPA	67-43-6	Industrial Substance	13	Unlisted
13	EDTA	1964-02-08	Industrial Substance	13	C
14	Iohexol	66108-95-0	Radiocontrast agent	12	B
15	Iomeprol	78649-41-9	Radiocontrast agent	12	B
16	Iopamidol	60166-93-0	Radiocontrast agent	12	B
17	Iopromide	73334-07-3	Radiocontrast agent	12	B
18	Acetone[2]	67-64-1	Industrial Substance	11	Unlisted
19	Amidotrizoic acid	117-96-4	Radiocontrast agent	11	B
20a	Glyphosate	1071-83-6	Pesticide	12	C
20b	AMPA (metabolite glyphosate)	1066-51-9	Pesticide	11 (12)	B
21	Desphenyl chloridazon (metabolite chloridazon)	6339-19-1	Pesticide	11	C
22	DMS (metabolite tolylfluanide)	3984-14-3	Pesticide	11	A
23	DEET	134-62-3	Biocide	10	B
24	Metoprolol	37350-58-6	Pharmaceutical	10	C
25	Nicosulfuron	111991-09-4	Pesticide	10	C
26	Sotalol	3930-20-9	Pharmaceutical	10	B

[1] The score of compounds was calculated using the scoring system described in Appendix 1. See Appendix 4 and Appendix 5 for details. Metabolites are grouped together: if the score of the parent compound or metabolite is higher, the score of the highest compound is given in between brackets

[2] DIPE and acetone regularly occur in high concentrations in the Meuse. They have a clear emitting source and it is proposed to monitor them only at the monitoring stations downstream from this source.

4.2 List 2—Candidate drinking water relevant compounds

All compounds that were previously on list A or B could be tested on the criteria of list 1. Based on the outcome they were either placed on list 1 or 3. An exception was made for urotropine. This compound is detected in the Meuse in concentrations above 1 µg/L. Since the compound is not toxic in these concentrations with a calculated BQ of 0.008, the total score of urotropine is below 10. However, on the EWM-meeting of 11 June 2015, the EWM members decided to keep urotropine on list 2 since the compound was not measured at all monitoring stations and the concentrations exceed the signaling parameter of 1 µg/L.

Bisphenol A used to be present on the list of relevance substances, but was removed in 2011 because the limit of detection of the available analytical methods was high and it was argued that the compound could better be followed via the monitoring of estrogenic activity. At the moment new techniques are available at the drinking water laboratories which can measure bisphenol A at lower levels (~5 ng/L) and it is proposed to replace this compound on list 2.

Furthermore, a literature study on emerging compounds was carried out and the screening data from HWL and Water-link was evaluated to check for potentially relevant compounds to the drinking water function of the river Meuse (see chapter 4.4). The complete list of proposed compounds can be found in Table 5 and 6 in Chapter 4.4 and 4.6. All compounds for which an analytical method is available were placed on list 2. List 2 is shown in Table 3.
Since this list contains many compounds, it is proposed to subdivide the list in parts and focus on one part in the yearly monitoring programme. After one year the compounds that have been monitored can be evaluated as described in chapter 3.4.1, and in the next year the focus in the monitoring programme can be placed on the second part of the compounds from list 2. In Table 3 the orange color indicates if the compounds will be part of the monitoring programme in 2016.

Table 3. *Candidate drinking water relevant compounds potentially relevant to the drinking water function of the river Meuse (list 2).*

Compound	CAS number	Application	Lab with method[2]	Source[3]
Industrial compounds				
Benzylalcohol	100-51-6	Solvent	TZW	SCR
Aniline	62-53-3	Industrial compound	TZW	SCR
Melamine	108-78-1	Resin	KWR, TZW	LIT
Pyrazole[1]	288-13-1	Industrial compound	ALZ, HWL, KWR	EMI
Tert-butyl alcohol (metabolite MTBE)	75-65-0	Fuel component	TZW	LIT
Urotropine	100-97-0	Industrial compound	KWR, TZW	other
Hormone disrupting compounds				
Anti-AR CALUX	-	Anti-androgenic activity	BDS	LIT

Compound	CAS number	Application	Lab with method[2]	Source[3]
GR-CALUX	-	Glucocorticoid activity	BDS	LIT
Bisfenol A	80-05-7	Industrial compound	HWL, KWR	other
Pesticides/biocides				
3,5,6-TCP (chlorpyrifos + triclopyr metabolite)	6515-38-4	Herbicide	TZW	LIT
Metazachlor ethane sulfonic acid	171118-09-5	Herbicide	KWR, TZW	LIT
Metazachlor oxalic acid	1231244-60-2	Herbicide	KWR, TZW	LIT
Metolachlor ethane sulfonic acid	172960-62-2	Herbicide	KWR, TZW	LIT
Oxadiazon	19666-30-9	Herbicide	KWR	SCR
Pharmaceuticals				
4-FAA (metabolite metamizol)	1672-58-8	Analgesic	KWR, TZW	LIT
Propyphenazone	479-92-5	Analgesic	TZW	LIT
Tramadol	27203-92-5	Analgesic	KWR, SWDE TZW	LIT/SCR
4-AAA (metabolite metamizol)	83-15-8	Analgesic	KWR, TZW	LIT
Amoxicillin	26787-78-0	Antibiotic	TZW	LIT
Ciprofloxacin	85721-33-1	Antibiotic	TZW	LIT
Clarithromycin	81103-11-9	Antibiotic	SWDE, TZW	LIT
Clindamycin	18323-44-9	Antibiotic	TZW	LIT
Erythromycin	114-07-8	Antibiotic	TZW	LIT
Roxithromycin	80214-83-1	Antibiotic	TZW	LIT
Gabapentin	60142-96-3	Anticonvulsant	KWR, TZW	LIT/SCR
Lamotrigine	84057-84-1	Anticonvulsant	KWR, TZW	SCR
Citalopram	59729-33-8	Antidepressant	SWDE, TZW	LIT
O-Desmethylvenlafaxine	93413-62-8	Antidepressant	TZW	LIT
Venlafaxine	93413-69-5	Antidepressant	SWDE, TZW	LIT/SCR
Fluconazole	86386-73-4	Antifungal medication	TZW	LIT
Irbesartan	138402-11-6	Antihypertensive	SWDE, TZW	LIT
Telmisartan	144701-48-4	Antihypertensive	TZW	SCR
Valsartan	137862-53-4	Antihypertensive	TZW	SCR
Amisulpride	71675-85-9	Anti-psychotic	TZW	SCR

[1] Pyrazole has a clear emitting source and it is proposed to monitor the compound only at the monitoring stations downstream from this source.
[2] ALZ = Aqualab Zuid, BDS = Biodetection Systems, HWL = Het Waterlaboratorium, KWR = KWR Watercycle Research Institute, SWDE = La société wallone des eaux, TZW = Technologie Zentrum Wasser
[3] Refers to the source in which the compound was found as a potential candidate: literature (LIT), screening (SCR) or incidental emissions (EMI)

4.3 List 3— No longer drinking water relevant compounds

Compounds that were previously on list A, B, or C, and have been measured for 5 or more years, and have the necessary parameters available to calculate the score, but were not found to exceed the ERM target values often enough or were not detected often enough (see chapter 3.3.1) were moved to the list of evaluated no longer drinking water relevant compounds (list 3). The proposed list 3 is shown in Table 4.

Table 4. *Proposed compounds no longer relevant to the drinking water function of the river Meuse (list 3), scored based on removal by water treatment, toxicity, odor/taste threshold and public perception.*

Compound	CAS #	Category	Previous list	Reason for removal
2,4-D (2,4-dichlorophenoxyacetic Acid)	94-75-7	Pesticide	C	ERM rate below 1%
4-n-Nonyl phenol	104-40-5	Industrial substance	A	Detection rate low
Acesulfame-K	55589-62-3	Artificial sweeteners	A	Score below 10
AHTN (6-acetyl-1,1,2,4,4,7-hexamethyltetraline)	1506-02-01	Fragrances and musks	A	Detection rate low
Aspirin (acetylsalicylic acid)	50-78-2	Pharmaceutical	B	Detection rate low
BAM (2,6-dichlorobenzamide)	2008-58-4	Pesticide	B	ERM rate below 1%
Barbital	57-44-3	Drugs of abuse	A	Detection rate low
BBP (butylbenzylphtalate)	85-68-7	Plasticizers	A	Detection rate low
Benzotriazole	95-14-7	Industrial substance	A	ERM rate low
BPS (4,4'-sulfonyldiphenol)	80-09-1	Industrial substance	A	ERM rate below 1%
Carbamazepine	298-46-4	Pharmaceutical	C	Below ERM last 2 years
Carbendazim	10605-21-7	Pesticide	C	Detection rate low
Chloridazon	1698-60-8	Pesticide	C	Below ERM last 2 years
Chlorotoluron	15545-48-9	Herbicide	C	ERM rate below 1%
DBP (dibutyl phthalate)	84-74-2	Plasticizers	A	Detection rate low
DEP (diethyl phthalate)	84-66-2	Plasticizers	A	Detection rate low
DIBP (di-(2-methyl-propyl)phthalate)	84-69-5	Industrial substance	A	Detection rate low
Diclofenac	15307-86-5	Pharmaceutical	C	ERM rate below 1%
Diglyme (bis(2-methoxyethyl)ether)	111-96-6	Industrial substance	B	ERM rate below 1%
Dimethenamid	87674-68-8	Herbicide	B	ERM rate below 1%
Diuron (DMCU)	330-54-1	Herbicide	C	Detection rate low
DMSA (N,N-dimethylaminosulfanilide)	4710-17-2	Pesticide	A	ERM rate below 1%
Estrone	53-16-7	Hormones	B	Detection rate low
ETBE (ethyl-tertiairy-butyl-ether)	637-92-3	Industrial substance	B	Detection rate low
Galaxolide (HHCB)	1222-05-5	Fragrances and musks	A	Detection rate low
Lincomycin	154-21-2	Pharmaceutical	B	Detection rate low
MCPA (4-chloro-2-methylphenoxyacetic Acid)	94-74-6	Pesticide	C	ERM rate below 1%
Mecoprop (MCPP)	93-65-2	Pesticide	C	ERM rate below 1%

Compound	CAS #	Category	Previous list	Reason for removal
Metazachlor	67129-08-2	Herbicide	B	ERM rate below 1%
Methyl-desfenylchloridazon	17254-80-7	Pesticide	C	Detection rate low
Metolachlor	51218-45-2	Pesticide	C	ERM rate below 1%
MTBE (methyl-tert-butylether)	1634-04-04	Industrial substance	C	Detection rate low
Musk (ketone)	81-14-1	Fragrances and musks	A	Detection rate low
Musk (xylene)	81-15-2	Fragrances and musks	A	Detection rate low
Naproxen	22204-53-1	Pharmaceutical	B	Detection rate low
N-butylbenzenesulphonamide	3622-84-2	Plasticizers	A	Detection rate low
NDMA (nitrosodimethylamine)	62-75-9	Industrial substance	A	Detection rate low
Pentobarbital	76-74-4	Drugs of abuse	A	Detection rate low
PFBA (perfluorobutanoic acid)	375-22-4	Perfluorinated compound	A	Detection rate low
PFBS (perfluorobutane sulfonate)	29420-49-3	Perfluorinated compound	A	Detection rate low
PFHxS (perfluorohexane sulfonate)	432-50-7	Industrial substance	A	Detection rate low
PFOA (perfluorooctanoic acid)	335-67-1	Perfluorinated compound	A	Detection rate low
PFOS (perfluorooctanoic sulfonate)	1763-23-1	Perfluorinated compound	A	ERM rate below 1%
Phenazone	60-80-0	Pharmaceutical	B	Detection rate low
Phenobarbital	50-06-6	Drugs of abuse	A	Detection rate low
Salicylic Acid	69-72-7	Pharmaceutical	B	Detection rate low
Sucralose	56038-13-2	Artificial sweeteners	A	Score below 10
Sulfamethoxazole	723-46-6	Pharmaceutical	B	Detection rate low
Surfynol 104 (2,4,7,9-tetramethyl-5-decyne-4,7-diol)	126-86-3	Industrial substance	A	Detection rate low
TBP (tributylphosphate)	126-73-8	Industrial substance	C	ERM rate below 1%
TCEP (tris(2-chloroethyl) phosphate)	115-96-8	Industrial substance	A	Detection rate low
TCPP (tri-(2-chloroisopropyl) phosphate)	13674-84-5	Industrial substance	A	Detection rate low
Tolyltriazole (5-methyl-1-H-benzotriazole)	29385-43-1	Industrial substance	A	Detection rate low

4.4 Literature study on candidates for the list of drinking water relevant compounds

In Chapter 4.4 a descriptive review is given of the literature search that was performed to investigate candidates for the list of drinking water relevant compounds. Emerging compounds of possible concern were divided into categories depending on their origin and use. To decide if a compound should be included in list 2 the reported concentrations played an important role: compounds that were detected above the ERM target value of 0,1 µg/L were considered. Furthermore, aspects like toxicology and expected use in the catchment area of the river Meuse were also taken into account (partly based on expert judgement). The bold depictured compounds in the paragraphs below are new candidates for list 2, if an affordable measuring technique is available. A summary of these compounds is given in Table 5.
Besides literature data, the screening results from HWL and Water-link were used for the evaluation of relevant compounds for the drinking water production of the Meuse (Chapter 4.6). A summary of these compounds is given in Table 6. The final selection of compounds is given in Table 3.

In a BTO project, a large scale liquid chromatography-high resolution mass spectrometry (LC-HRMS) suspect screening was performed to prioritize 5200 authorized chemicals based on 151 water samples (waste water, surface water, groundwater and drinking water) (Sjerps et al. 2015). They found 243 suspects, most of them with an unconfirmed identity. A further evaluation of the suspects (identification and risk assessment) will be performed in projects under the BTO framework and it is recommended to follow the results of these projects in future evaluations of the relevant compounds lists of the RIWA Meuse.

4.4.1 Pharmaceuticals

General

Pharmaceuticals are a broad category of pollutants. A part of the pharmaceuticals is excreted unchanged in urine and feces and may reach surface water if they are not removed during waste water treatment. While concentrations of pharmaceuticals found in drinking water are generally low enough to have no significant effects on health, the public concern of pharmaceuticals in drinking water is high (Houtman et al., 2013). Drinking water companies are already monitoring a selection of pharmaceuticals in their sources and drinking water. In the monitoring program of Dunea over 40 pharmaceuticals are monitored in the surface water of the Meuse. The only pharmaceutical that is regularly measured above the ERM target value of 0.1 µg/L is metformin. Tthe compounds paroxetine, fenofibrate and theophylline are sporadically detected in concentrations above 0.1 µg/L, but do not comply to the criteria of list 1.

Since 2013 **guanylurea** is also monitored in the Meuse. Guanylurea is the most important transformation product of metformin, an oral antidiabetic pharmaceutical. Metformin was placed on list A in 2011 because it was found in concentrations above 1 µg/L in the Meuse. Measured concentrations of guanylurea in the Meuse are well above the 1 µg/L and are exceeding both the ERM target value and the "signaling parameter

(signaleringsparameter)" of 1 µg/L for anthropogenic substances in the Dutch Drinking Water Regulation (2011). No toxicity data are available for guanylurea. It is recommended to add guanylurea to the lists of relevant substances in the Meuse.

Data on European surface water concentrations found in literature show the presence of the following pharmaceuticals: **gabapentin** at concentrations up to 1.87 µg/L, paracetamol at up to concentrations 1.38 µg/L, theophylline at concentrations up to 0.55 µg/L, and the antibiotics **amoxicillin, nortramadol, erythromycin**, and **cimetidine** at up to 0.24 µg/L (reviewed in Petrie et al, 2015). In a Swedish study the following compounds were reported that exceeded surface water concentrations of 0.1 µg/L, namely: bisoprolol, **ciprofloxacin, citalopram, clarithromycine, clindamycin, codeine,** diclofenac, **fexofenadine, flecainide, fluconazole,** ibuprofen**, irbesartan, mirtazapine,** paracetamol, **roxithromycin,** trimethoprim, **venlafaxine** (Fick et al., 2011). The compounds that are not depictured in bold are already monitored in the Meuse, and were therefore evaluated in this project. They do however not fulfill the criteria of list 1. Some of the compounds belong to the group of antibiotics which were included in the new watch list of 2015 for substances that should be monitored European-Union wide pursuant to Directive 2008/105/EC on environmental quality standards in the field of water policy[2].

In Sjerps et al. 2015, a list of suspects that were detected in different water matrices was compared with literature data. The following identified pharmaceuticals were found in the screening study of KWR as well as in monitoring studies described in literature: oxazepam, phenazone, **propyphenazone** and **tramadol**. The last two compounds are not yet monitored in the Meuse and could be potentially relevant for the drinking water production.

In a study on transformation products of pharmaceuticals in Dutch surface waters, compounds with maximum concentrations above 0.1 µg/L were: **4-acetylaminoantipyrine** (4-AAA) at 0.17 µg/L, **4-formylaminopyrine** (4-FAA) at 0.16 µg/L (transformation products of the analgesic metamizol), and **O-desmethylvenlafaxine** at 0.11 µg/L L (transformation product of the antidepressant venlafaxine) (de Jongh et al., 2012).

In the joint RIWA monitoring programme of drinking water companies along the river Rhine, gabapentin and hydrochlorothiazide were detected in concentrations above 0.1 µg/L. Also, the transformation products 4-AAA and 4-FAA were detected in concentrations above 0.1 µg/L.

All pharmaceuticals that exceed a concentration of 0.1 µg/L are potentially relevant for the drinking water production. All of the above mentioned compounds, except for the ones that are already monitored in the Meuse and evaluated, are therefore possible candidates for list 2.

[2] http://eur-lex.europa.eu/legal-content/NL/TXT/PDF/?uri=OJ:JOL_2015_078_R_0008&from=EN

Cytostatic agents

Cytostatic agents are compounds that halt and prevent cell growth and are used e.g. in cancer treatment. HWL measures several cytostatics, cyclophosphamide and iphosphamide, neither of which were detected in the abstracted surface water and drinking water in 2013. Tamoxifen, a drug that suppresses breast cancer, was found in surface water at concentrations of 0.21 µg/L (Petrie et al., 2015). In the Netherlands and Belgium, tamoxifen is subscribed in very low amounts, so it is not expected to be present in high concentrations in the surface water of the Meuse.

Veterinary medicines

Veterinary medicines are not the subject of research as often as humane drugs, even though the usage rates of both are approximately the same. Most of the veterinary drugs used are antibiotics (~90%) (Velzeboer et al., 2014). Other types of veterinary drugs are antiparasitics and antifungals. Example of commonly used antibiotics and antiparasitics that degrade relatively fast are penicillin based antibiotics. Examples of compounds that degrade relatively slow are tetracyclines, ivermectin, and (fluoro)quinolones. An important difference between humane and veterinary drugs is their source and environmental pathway; humane drugs generally enter through waste water, and make their way through waste water treatment plants to the surface water, while veterinary drugs enter the surface and ground water through manure (Velzeboer et al., 2014).

Ivermectin is an antiparasitic medicine administered to horses. It has also been found to be toxic to insects and marine wildlife. It degrades slowly in the environment. Ivermectin was detected in Dutch surface water (Sjerps et al., 2013) and in Dutch river water samples ivermectin concentrations below 0.1 µg/L were detected (Lahr et al., 2014). Potential ivermectin uptake through drinking water with a maximum concentration of 0.1 µg/L is low compared to medical doses given, however chronic toxic effects for *D. magna* at ivermectin concentrations as low as 0.001 ng/L have been reported (Garric et al., 2007).

Although ivermectin is mainly a concern because of environmental health effects and not for its human health effects, it is proposed to add ivermectin to list 2 because of its presence in surface water, and the negative publicity in the media.

MRI contrast agents

Magnetic Resonance Imaging (MRI) is a relatively new technique used in medical diagnosis of the human body. Some MRI scanning methods inject the patient with contrast agents which improve the quality of the generated image. Though the use of MRI contrast agents has increased in the recent years, the long term effects of MRI contrast agents remain largely unknown (Kools et al., 2013).

A study by RIWA showed that the increase of usage of MRI scanners in healthcare has led to a higher occurrence of MRI contrast agents in the environment (Kools et al., 2013). Gadoteric acid and gadobutrol are the most frequently used MRI contrast agents in the Netherlands with over 579 kg and 188 kg sold respectively in 2011 (Kools et al., 2013). Gadolinium-based MRI contrast agents have been detected in different water

sources, including the river Rhine, but a widespread analysis of MRI contrast agents in the Netherlands has yet to be carried out. The concentrations of MRI contrast agents in the river Rhine were found to be below target values and concentrations which have adverse effects. Because concentrations of MRI contrast agents found so far have been below the ERM target value, no MRI contrast agents are proposed to be added to list 2. It is suggested to review MRI contrast agents in the future, as improved analytical methods may reveal more potentially relevant MRI contrast agents.

Illegal and unregulated drugs

The use of illegal and unregulated drugs can be monitored through waste water analysis. Illegal and unregulated drugs have received bad publicity recently after news reports stated that drinking water may contain miniscule amounts of drugs, e.g. waste water from Amsterdam. In a study published by van der Aa et al. (2013), several illegal and unregulated drugs were investigated in surface water, raw water, and waste water (water that has undergone a conventional drinking water pre-treatment) in the Netherlands. In the surface water methamphetamine, oxazepam, temazepam, cocaine, benzoylegonine, morphine, codeine, and methadon were found at concentrations well below 0.1 µg/L. In the raw water only three kinds of drugs were found: oxazepam, temazepam and benzoylegonine at concentrations below 0.01 µg/L, and in the drinking water only benzoylegonine was found in concentrations below the detection limit of 1 ng/L. The risk on public health of these compounds through consumption of drinking water was found to be negligible. A study in the UK found codeine at concentrations up to 0.347 µg/L in surface waters (Petrie et al., 2015), however the study observed codeine at concentrations below 0.023 µg/L in the Netherlands.

Because concentrations of illegal and unregulated drugs found in the Netherlands are low, it is not proposed to add any illegal or unregulated drugs to list 2.

4.4.2 Plant protection products and biocides

Pesticides

Pesticides can be divided into subcategories such as herbicides, insecticides, fungicides, etc. They are used in agriculture to improve crop yield, but are also available to consumers to a limited degree. Pesticides are marked as having a negative effect on the public perception of drinking water. A large amount of pesticides is monitored by the drinking water companies because of a regulatory obligation. The use of pesticides changes through the years. New pesticides are being allowed on the market, other pesticides are banned. The EWM analysed in 2011 which pesticides and biocides were allowed in 2011 based on data from the CTGB. From this analysis it followed that there are 341 active substances allowed as pesticide, from which 171 were not present in the RIWABASE (EWM document 13-21). RIVM ranked the pesticides based on expected emission and the following compounds were found to have a high emission and are not monitored yet: chloromequat, diquat dibromide, mandipropamid, prothioconazole, metaldehyde, cyazofamide, hymexazole, karvon D, trinexapac ethyl,

fludioxonil, benthiavalicarb isopropyl, fenpropidin, fosetyl, mailine hydrazide and bifenox. At the moment a BTO research project is conducted by KWR in which an inventarisation is made of the possible drinking water relevant pesticides and metabolites. This project will be completed in 2016 and it is recommended to use the results from this project to select the pesticides which could be relevant for the drinking water function of the river Meuse and include them in List 2 in the next evaluation.

In the last years in literature, the focus is more often on the metabolites of pesticides. In a study on pesticide metabolites in surface waters, compounds were ranked according to their concentration in surface waters in Germany and the relevance for drinking water was indicated (Reemtsma et al., 2013). Based on concentrations, the highest ranked metabolites were: metazachlor ethane sulfonic acid (ESA) (with the highest median concentratrion of almost 0.1 µg/L, terbuthylazine-2-hydroxy MT13 (with the highest frequency of detection and a median concentration of 0.04 µg/L), dichlorphos metabolite 2,2-dichloroacetic acid, chlorothalonil metabolite R417888, dimethachlor metabolite CGA369873, trisulfuron metabolite BH 635-2, tritallate metabolite TCPSA, metazachlor metabolite BH 479-12, and the captan metabolite THPAM. Based on drinking water relevance the highest ranked metabolites were terbuthylazine-2-hydroxy MT13, chlorpyrifos metabolite + triclopyr metabolite 3,5,6-TCP (share same parent compound) and the thiamethoxam metabolite clothianidin. Clothianidin is measured in the regular monitoring programme of Dunea, and did not exceed the limit of quantification of 0.02 µg/L in the intake water of Brakel in 2014.

In the joint RIWA monitoring programme of drinking water companies along the river Rhine, the metabolites of metazachlor; metazachlor oxalic acid, metazachlor ethane sulfonic acid, and the metabolite of metolachlor; metolachlor ethane sulfonic acid were detected in concentrations above 0.1 µg/L.

It is proposed to add **3,5,6-TCP** to list 2 because of its potential relevance to drinking water. **Metazachlor oxalic acid**, **metazachlor ethane sulfonic acid**, and **metolachlor ethane sulfonic acid** are proposed to be added to list 2 because they are detected in concentrations above 0.1 µg/L.

Biocides

Biocides are compound aimed to repel or kill harmful organisms. KWR has examined samples of surface water for nine different biocides (Vughs et al., 2014). The only biocide that was detected was irgarol, an algicide, in the intake water near Brakel at a concentration of 6 ng/L, well below the signal value of 0.1 µg/L. Because biocide concentrations reported are well below the signal value of 0.1 µg/L, it is not proposed to add them to list 2.

Neonicotinoids

Neonicotinoids are insecticides with a structure similar to nicotine. They have become controversial in the recent years due to their suspected detrimental environmental effects. An important neonicotinoid is imidacloprid, an insecticide used in agriculture. It currently is the most commonly used insecticide in the world, and is used in a variety of locations, such as open and closed cultivation, and domestic use. Imidacloprid degrades slowly in the

environment and is expected to accumulate upon heavy usage. It is toxic for aquatic lifeforms and has received negative publicity due to linkage with recent honey bee colony collapses. In April 2013 the EU restricted the use of imidacloprid, and the Netherlands has restricted permission for the use of imidacloprid containing products use since September 30th 2013 (Verhagen et al., 2014). In the past five years imidacloprid was occasionally found in the intake water near Brakel in concentrations up to 0.03 µg/L, though it was not found in drinking water (with a detection limit of 0.01 µg/L). Despite its negative public perception, it is not proposed to add imidacloprid to list 2 due to low reported concentrations in surface and drinking water. Neonicotinoids were included in the new watch list of 2015[3]. In this way, neonicotinoids will continue to be monitored.

4.4.3 Industrial compounds

General

Industrial compounds consist of a variety of substances, used in industries but also in households. It is very difficult to gain an exact insight in which industrial compounds could be relevant for the drinking water function of the Meuse. Chemicals are regulated in the REACH legislation (Registration, Evaluation and Authorisation of CHemicals, 1907/2006), but it concerns so many compounds that ranking them to relevance is not possible within the scope of this project. Besides, there are also many industrial compounds which are used as intermediate products and for these compounds the evaluation dossiers contain only limited information as became clear in the case of the pyrazole incident (see Chapter 4.5).

In analytical screenings that are being performed in the river Meuse, industrial compounds still remain a notable part of the total amount of compounds, making it evident that this group of compounds remains a concern for the drinking water companies. Very recently a BTO report was published which describes the results of a project in which new chemical threats to the sources of drinking water are explored (Sjerps et al. 2015). In this research 215 compounds signaled attention, including several industrial compounds.

(Quantitative) concentration ranges in rivers are not known for the industrial compounds from the screenings and BTO research and including them all in list 2 would make the list very extended. Since the next step in the BTO research is to prioritize the signaled chemicals, it is recommended to follow this research and use the results of the prioritization as input for list 2 during the next evaluation.

In literature, a study on the presence of industrial substances from treated chemical plant waste was found. In this study several compounds were discovered with notable concentration ranges in wastewater: **dichlorobenzene** (0.8-14 µg/L), **tributylamine** (5.1 µg/L), **3'-(trifluoromethyl)acetophenone** (0.3-5

[3] http://eur-lex.europa.eu/legal-content/NL/TXT/PDF/?uri=OJ:JOL_2015_078_R_0008&from=EN

µg/L), **dichloroaniline** (0.2-4.8 µg/L) and **tetramethylbutanedinitrile** (0.5-1.4 µg/L) (Botalova et al., 2011). It is recommended to add these compounds to list 2.

In the GCMS-screening of HWL, **tetra-acetyl-ethylene-diamine** (**TAED**) and **tri-phenyl-phosphine oxide** (**TPPO**) are often detected in the surface water of the Meuse. TAED is used a bleaching agent in households and industries. It was found during a screening analysis of the river Meuse near Keizersveer, and also during a screening analysis of HWL. It was found once at a indicative concentration of 2 µg/L in the intake water of Brakel. The no observed adverse effect level (NOAEL) of TAED has been reported as 90 mg/kg/day by HERA (2002). On basis of this NOAEL it can be said that the effects on humane health by TAED intake through drinking water are small (Slootweg and de Boer-Breedeveld, 2014). TPPO is a reagent used for industrial crystallization processes, and is also used as a solvent. It is recommended to include TAED and TPPO in list 2.
In the joint RIWA monitoring programme of drinking water companies along the river Rhine, the resin **melamine** was detected in concentrations above 0.1 µg/L.

RIVM drew attention to the compound **tert-butyl alcohol** (TBA), a degradation product of MTBE. This compound is mentioned as a problematic compound for the extraction of groundwater for the production of drinking water (personal communication M. van der Aa (RIVM)). Since MTBE is also detected in the surface water of the Meuse, TBA could be a relevant compound.

It is proposed to add melamine and TBA to list 2, the other compounds either have concentrations well below toxic levels, or are only occasionally reported at high concentrations.

Flame retardants

Flame retardants are substances used to repel or slow down fire. The EU project ENFIRO has done research on halogen free flame retardants, including flame retardants based on organophosphates, metals, organic nitrogen, and nanoclay. The goal was to develop flame retardants with less toxicity than previously used flame retardants (Waaijers et al., 2013). In Spain the flame retardants tris(chloropropyl)phosphate (TCPP), tri-n-butylphosphate (TnBP), di-n-butyl phosphate (DnBP), and di-(2-ethylhexyl)phosphoric acid (DEHPA) were found at concentrations above 0.1 µg/L in surface water. In waste water effluent the compounds tris(2-chloroethyl) phosphate (TCEP), tri-iso-butyl phosphate (TiBP), tris-(butoxyethyl)-phosphate (TBEP), diphenyl phosphate (DPhP) were found at concentrations above 0.1 µg/L. Removal by water treatment was reported to be lower for the compounds TCEP, TCPP, and TBEP than for TiBP and TnBP (Rodil et al., 2012). An EU-wide wastewater treatment study found the tris-phosphates TBEP, tributylphosphate (TBP), tris[2-chloro-1-(chloromethyl)ethyl]phosphate (TDCP), and TiBP at median concentrations above 0.1 µg/L in waste water (Loos et al., 2013). The phosphate flame retardants that are regularly detected with the screening techniques used for the monitoring of the Meuse are: TiBP, TCEP, TCPP, TBP, TBEP, triethylphosphate (TEP), and tri(1,3-dichloro-2-propanol)phosphate (TDCPP). In 2011, three phosphates were placed on the list of (potentially) relevant compounds, namely TBP (list 1), TCEP (list 2) and TCPP (list 3). None of them met the criteria to stay on the list. Since these three phosphates are the most frequently detected ones, in the highest concentrations, it is not

proposed to include the other phosphates on list 2. It is recommended to follow the occurrence of phosphates in the Meuse via screening techniques and evaluate them during the next update of the lists.

Ionic liquids

Ionic liquids are salts with a melting point of 100 ºC or below. They are commonly used as solvent, lubricant, fuel, and as active compound for pharmaceutical products. The most commonly used heterocyclic cationic liquids are imidazolium, pyridinium, pyrrolidinium, morpholinium, piperidium, and quinolium. Functional groups of ammonium, phosphonium, and sulphonium are also used. Ionic salts are often used as a replacement for organic solvents, as ionic salts are less volatile. They have a low viscosity, a higher density compared to organic solvents, are thermally and electrochemically stable, and are hygroscopic and therefore generally highly soluble in water. Ionic salts can act as solvent for organic, inorganic, polar, and non-polar compounds, and polymers, depending on their composition (Velzeboer et al. 2014).

The category of ionic liquids is broad, due to the varying compounds and compositions among it. This makes it difficult to assess the risks of the compounds in this category in general. It is expected that ionic liquids end up in drinking water due to their high solubility in water. While ionic liquids are considered to be environmentally friendly because of their low vapor pressure, this makes them more of a threat to aquatic and terrestrial biosystems since these liquids do not vaporize quickly. The anionic components of the liquids do not possess significant toxic properties, except for perfluoro-anions. The cationic components of piridinium are more environmentally friendly than imidazolium. Current research of the effects of ionic liquids on the ecology is limited (Pham et al., 2010).

It is not proposed to add ionic liquids to list 2 since available data is limited. It is recommended to review ionic liquids in the future, when more information about their toxicity, biodegradability, and presence in surface and drinking water is available, and when better analytic techniques for measuring ionic liquids have been developed.

Perfluorinated compounds

Perfluorinated compounds are organic molecules containing C – F bonds. They have many applications, such as cooking utensils, surfactants, and refrigerants. They are stable compounds and have the tendency to bioaccumulate. The perfluorinated compounds perfluorooctanesulfonate (PFOS) and perfluorooctanoic acid (PFOA) were some of the most commonly used perfluorinated compounds. Since the use of these compounds was restricted by EU regulation, they were replaced by other perfluorinated compounds that have become more common recently. Examples of compounds that have replaced PFOS and PFOA are: perfluorobutanesulfonic acid (PFBS), perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), and perfluorohexanoic acid (PFHxA), which are difficult to remove during water treatment.

Most of the perfluorinated compounds mentioned were included in the RIWA compound lists of 2011, but they no longer fulfill the criteria for list 1. PFPeA is not part of the lists, but considering the low concentrations found

of other perfluorinated compounds, it is not expected that PFPeA is potentially relevant. It is therefore not proposed to add any perfluorinated compounds to list 2.

Quaternary ammonium compounds

Quaternary ammonium compounds form a group of industrial chemical compounds which are used in many different applications such as: biocides, disinfectants, fabric softeners, herbicides, antifouling, personal care products and wood preservatives. As such, they are among the most relevant organic pollutants of anthropogenic origin with the potential to enter the environment (Vughs and Kolkman, 2014).
Tetrapropylammonium (TPA) was detected in concentrations of up to 3 µg/L in the river Rhine in 2013. This prompted a second study, which showed that TPA was found in surface water and drinking water at much lower concentrations between 0.01-0.1 µg/L, although it is poorly removed by water treatment. In the same study, 26 additional quaternary ammonium compounds were measured, but none were detected in a concentration above 0.05 µg/L (which is below the ERM target value) (Vughs and Kolkman, 2014). It is therefore not proposed to add quaternary ammonium compounds to list 2.

Siloxanes

Siloxanes are organic compounds containing silicium, oxygen, carbon, and hydrogen. Siloxanes are versatile compounds used in for example sealants, food, lubricants, toys, and shampoos. Cyclic and linear siloxanes are resistant to degradation and thus accumulate easily in the environment. Examples of linear siloxanes are: hexamethyldisiloxane (HMDSO), octamethyltrisiloxane, and decamethyltrisiloxane. Examples of cyclic siloxanes are: octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), dodecamethylcyclohexasiloxane (D6), and tetradecamethylcycloheptasiloxane (D7) (Velzeboer et al. 2014). D4, D5, and D6 are bioaccumalative with a bioconcentration factor between 2000 and 12400. HDMSO is not bioaccumulative, but it is volatile. The high octanol-water partition coefficients K_{ow} of D4, D5, D6, and HDMS, which are between 3.7 and 4.9, lead to the assumption that these compounds are easily removed during treatment. Furthermore, siloxanes are poorly soluble in water, meaning that human uptake of siloxanes through drinking water will be limited. Octamethyltetrasiloxane, a compound used in cosmetics, has been mentioned as being frequently used, toxic, bioaccumulative, and negatively affecting reproduction (Smit and Wuijts, 2012). There is however not enough evidence for it being potentially relevant for the production of drinking water.

It is not proposed to add any siloxanes to list 2 due to their limited data available and presumably easy removal by water treatment. It is recommended to review siloxanes in the future, when more data is available.

UV filters

UV filters are used in sunscreens, cosmetics, and personal care products. The interest in the presence of UV filters rose after they were found in drinking water through screenings. UV filters can be divided in organic UV filters, which absorb UV radiation, and inorganic UV filters (TiO_2, ZnO), which reflect and disperse UV radiation. Examples of organic UV filters are benzophenone-3 (BP-3), 2-ethylhexyl 4-(dimethylamino)benzoate (OD-PABA),

4-methyl-benzylidene camphor (4-MBC), ethyl-hexyl methoxy-cinnamate (EHMC), octocrylene (OC), iso-amyl-methoxy-cinnamate (IAMC), and phenyl-benzimidazole sulfonic acid (PBSA). Most of these compounds are lipophilic and poorly soluble in water (Richardson and Ternes, 2011).

In Spain, the UV-filter PBSA was detected at concentrations of up to 0.9 µg/L in waste water effluent, though surface and drinking water concentrations were well below 0.1 µg/L. Benzophenone-4 (BP-4) was found at concentrations of over 1 µg/L in waste water effluent and over 0.1 µg/L in surface water. However, concentrations in the Meuse are expected to be lower since the use of UV-filters in colder countries like Belgium and the Netherlands is lower than in Spain. 4-Methylbenzylidene camphor (4-MBC) and benzophenone-3 (BP-3) were found in waste water at concentrations below 90 ng/L (Rodil et al., 2012). It is proposed to follow the trends of UV-filters through screenings, but not toadd any UV-filters to list 2 for now.

4.4.4 Other categories

Artificial sweeteners

Artificial sweeteners replace natural sweeteners in some products, for example because of their lower caloric content, or stronger taste. They are generally not metabolized inside the human body and are secreted unchanged. Besides acesulfame and sucralose, two other artificial sweeteners, saccharine and cyclamate have been reported to be present in surface water, but are well removed during water treatment (Slootweg and Speksnijder, 2014). None of the four artificial sweeteners fulfill the criteria for list 1. No other potentially relevant artificial sweeteners have been found.

Carboxylates/carbonic acids

Carboxylates or carbonic acids are organic acids containing a $COOH/COO^-$ rest group. Carbonic acids generally are weak acids, with some exceptions such as benzoic acid. Carbonic acids contain long aliphatic tails which are also called fatty acids.

Water treatment using oxidation by e.g. ozone treatment can form carbonic acids from other organic compounds present in the water. Carbonic acids pose little threat to human consumption; however they are consumed as substrate by microbial organisms, leading to accumulation of biomass and biofilms of pipelines. This makes the analysis of carbonic acids in water important. In a joint research project of the Dutch drinking water companies, carbonic acids were analyzed. Six organic acids were detected: lactate (lactic acid), acetate (sodium acetate), formate (formic acid), malate (malic acid), oxalate (sodium oxalate), citrate (citric acid) (Velzeboer, 2014).

Since carbonic acids do not pose a direct risk to human health, it is not proposed to add them to list 2. Indirect risks have not been established yet.

Hormones and hormone disturbing compounds

Compounds with hormonal activity in water streams have gotten a lot of attention in the past decades. Hormones and hormone disturbing compounds have a negative effect on the public perception of drinking water. Since compounds with hormonal activity can exert effects at very low concentrations, it is a challenge to perform analytical measurements with a relevant limit of detection. A good alternative is the use of bioassays. A bioassay involves the use of live animal or plant (*in vivo*) or tissue or cell (*in vitro*) to determine the biological activity of a substance. The advantage is that bioassays are suitable to assess effects of complex mixtures and that they can measure effects with low limits of detection (e.g. 7 pg/L for the ER-CALUX that measures estrogenic activity).

For estrogenic activity, the ER-CALUX is already included in list 1 of the RIWA Meuse monitoring programme. Recent investigations for Dutch drinking water companies show that anti-androgenic activity and glucocorticoid activity are also relevant parameters for surface waters (unpublished data). It is therefore proposed to add the bioassays **GR-CALUX** (glucocorticoid activity) and **anti-AR CALUX** (anti-androgenic activity) to list 2. For the GR-CALUX, KWR established a trigger value of 21 ng/L dexamethasone-equivalents (Brand et al. 2013) which could be used instead of the ERM target value (as is also done for the ER-CALUX).

Microplastics

Microplastics are plastic particles with a diameter of 0.06 – 5 mm (Besseling et al., 2014). Although not much is known about the exact sources of microplastics, plastic objects and waste discarded in marine environments are thought to be the biggest contributors to microplastics (Derraik, 2002), but microplastics also originate from household products such as synthetic clothing or cosmetics whose microplastic particles leave through waste water streams (Andrady, 2011; Fendall and Sewell, 2009).

Microplastics are highly resistant to degradation and are estimated to persist in the environment for possibly hundreds of years or more (Barnes et al., 2009). They have been reported to have adverse effects on marine wildlife (Cole et al., 2011). However, the effects of microplastics on human have not been sufficiently studied yet. Microplastics have been reported to affect molecular and cellular pathways, and compounds bound to microplastics can be released once microplastics are taken up, causing toxicity (Avio et al., 2015).

In a study conducted by Wageningen Universiteit on microplastics in Dutch fresh water streams, five locations in the Netherlands were sampled and filtered for small particles (Andrady, 2011). Particles with a size between 50 µm – 1 mm were separated from the water. The microplastic particle concentration from the measured samples was found to be approximately 0.08 ± 0.05 particles per m^3 of water. Effluent from sewage treatment was found to make a prominent contribution to microplastic pollution, containing 17 to 68 times as much microplastic as samples from other locations. It is however lower than previously found microplastic concentration in sewage treatment effluent of 10,000 – 20,000 particles per m^3 (Leslie et al., 2012). It is possible that the fiber-like dimensions of the microplastics (long but thin) allow them to pass through the net more easily then intended, meaning that used sampling methods may underestimate the true microplastic concentration in the water. Due to the varying shapes and sizes of microplastics, it can be difficult to tell them apart from other non-plastic fibers

and particles. Recently, Urgert (2015) carried out a study in on the abundance and composition of microplastics in the Dutch parts of the European rivers Meuse and Rhine. Using aman and Fourier Transform spectroscopy in combination with Principal Component Analysis (PCA) the composition of particles was identified. Both in the Meuse and Rhine, films, scrubs and the majority of the miscellaneous microplastics were identified as polyethylene. No spherules were found in the Meuse. For the size range of 0.125-5 mm, average concentrations of 0.14 mg or 9.7 microplastics per m^3 were calculated for the Meuse. This is an under limit as microplastics can become lost during processing or overlooked (Urgert 2015).

The relevance of microplastics for the production of drinking water and health are not fully understood. It is also not known if microplastics are removed from the drinking water in water treatment steps. Detection of microplastics in water is difficult due to the large variety of shapes and compositions, and such microplastics are currently not regularly measured in drinking water.

Microplastics are under focus in the BTO-research. It is recommended to follow the progress in the BTO research and add the microplastics to list 2 when the research demonstrates their relevance and better analytical techniques are developed.

Musks

Musks are fragrant compounds used in perfumes, detergents, and cosmetics. Because of their nature, musks tend to be very volatile. Using the GC-MS screening method of HWL, xylene, ketone, galaxolide, and AHTN, were detected. These musks were previously added to the lists. Musks in general are well removed during water treatment. No other musks that are potentially relevant have been found. It is therefore not proposed to add any musk to list 2.

Nanoparticles

Nanoparticles are a group of compounds potentially relevant for the quality of surface and drinking water. The Dutch ministry of infrastructure specifically mentioned nanoparticles in their latest regulation (Ministerie van Infrastructuur en Milieu, 2014). Nanoparticles are under attention in the Joint Research studies for the Dutch drinking water companies (BTO) performed by KWR and developments for methods for analysis are currently in progress. KWR is also involved in the European research project "NANONEXT" (e.g. Baüerlein and Lammertse, 2015).

It is recommended to await the results from the BTO research before adding nanoparticles to list 2.

Table 5. *Compounds that are reported in literature in surface waters in Europe in concentrations that are potentially relevant for the drinking water production.*

Compound	CAS	Category	Target analysis available?	Reference
Pharmaceuticals				
4-AAA (metabolite metamizol)	83-15-8	analgesic	Yes	de Jongh et al., 2012; RIWA Rhine[1]
4-FAA (metabolite metamizol)	1672-58-8	analgesic	Yes	de Jongh et al., 2012; RIWA Rhine[1]
Amoxicillin	26787-78-0	antibiotic	Yes	Petrie et al, 2015
Cimetidine	51481-61-9	H_2-receptor antagonist	?	Petrie et al, 2015
Ciprofloxacin	85721-33-1	antibiotic	Yes	Fick et al., 2011
Citalopram	59729-33-8	antidepressant	Yes	Fick et al., 2011
Clarithromycin	81103-11-9	antibiotic	Yes	Fick et al., 2011
Clindamycin	18323-44-9	antibiotic	Yes	Fick et al., 2011
Codein	76-57-3	analgesic	?	Fick et al., 2011
Erythromycin	114-07-8	antibiotic	Yes	Petrie et al, 2015
Fexofenadine	83799-24-0	antihistamine	?	Fick et al., 2011
Flecainide	54143-55-4	antiarrhythmic agent	?	Fick et al., 2011; screening data
Fluconazole	86386-73-4	antifungal medication	Yes	Fick et al., 2011
Gabapentin	60142-96-3	anticonvulsant/ analgesic	Yes	Petrie et al, 2015; RIWA Rhine[1]
Irbesartan	138402-11-6	angiotensin II receptor antagonist	Yes	Fick et al., 2011
Ivermectin	70288-86-7	veterinary parasiticide	?	Sjerps et al., 2013; Lahr et al., 2014
Mirtazapine	61337-67-5 (85650-52-8)	antidepressant	?	Fick et al., 2011
Nortramadol (metabolite tramadol)	75377-45-6	analgesic	?	Petrie et al, 2015
O-Desmethylvenlafaxine	93413-62-8	antidepressant	Yes	de Jongh et al., 2012
Propyphenazone	479-92-5	analgesic	Yes	Sjerps et al. 2015
Roxithromycin	80214-83-1	antibiotic	Yes	Fick et al., 2011
Tramadol	27203-92-5	analgesic	Yes	Sjerps et al. 2015
Venlafaxine	93413-69-5	antidepressant	Yes	Fick et al., 2011
Industrial compounds				
Dichloroaniline	-	industrial compound	?	Botalova et al., 2011
Dichlorobenzene	-	industrial compound	?	Botalova et al., 2011
Melamine	108-78-1	resin	Yes	RIWA Rhine[1]
Tert-butyl alcohol (metabolite MTBE)	75-65-0	fuel component	Yes	RIVM
Tetramethylbutanedinitrile	3333-52-6	industrial compound	?	Botalova et al., 2011
Tributylamine	102-82-9	industrial compound	?	Botalova et al., 2011
3'-(Trifluoromethyl)acetophenone	349-76-8	industrial compound	?	Botalova et al., 2011
Pesticides and biocides				
3,5,6-TCP (chlorpyrifos + triclopyr met)	6515-38-4	herbicide	Yes	Reemtsma et al., 2013
Metazachlor oxalic acid	1231244-60-2	herbicide	Yes	RIWA Rhine[1]
Metazachlor ethane sulfonic acid	172960-62-2	herbicide	Yes	RIWA Rhine[1]
Metolachlor ethane sulfonic acid	171118-09-5	herbicide	Yes	RIWA Rhine[1]
Hormone disturbing compounds				
GR-CALUX	-	Glucocorticoid activity	Yes	KWR data (unpublished)
Anti-AR CALUX	-	Anti-androgenic activity	Yes	KWR data (unpublished)

[1] RIWA Rhine monitoring programme of 2014

4.5 Incidental emissions

A few compounds are detected every now and then in very high concentrations in the Meuse. This concerns e.g. acetone and di-isopropylether (DIPE) which are responsible for a significant part of the alarms at Eijsden. Rijkswaterstaat and the *Service Public de Wallonie* (SPW) have found that the industrial emissions of these compounds take place at the same location in the Walloon part of the river basin. At the moment, an obligation to investigate the origin of acetone, is included in the discharge permit. However, the steps to reduce the discharge of acetone and DIPE are not yet taken (RIWA 2015). DIPE was already placed on the list of relevant compounds and it is recommended to keep it on list 1 (also based on the criteria). Acetone was not yet on any list, but enough monitoring data were available to evaluate the compound and based on the criteria, the compound should also be placed on list 1. The monitoring of these compounds could be limited to the monitoring stations downstream of the known emission location.

In July 2015, an unknown compound was detected via screening of Meuse water near Heel. This compound was identified by KWR as the polar compound pyrazole, which is applied as an intermediate in the production of i.a. acrylonitrile (memo KWR to WML, 24 July 2015). Concentrations were found in up to 90-100 µg/L. Toxicity data is rare for this compound and KWR proposes to use the TTC-value of 0,01 µg/L for pyrazole since a provisional drinking water guideline cannot be determined. The lower TTC-value was chosen since QSAR-models indicate that the compound is possibly genotoxic. Since there is not enough monitoring data available to determine if pyrazole complies to the criteria of list 1, it is proposed to add pyrazole in the first instance to list 2 with candidate drinking water relevant compounds. Since pyrazole has a known emitting source, it is proposed to monitor them only at the monitoring stations downstream from this source, similar as for DIPE and acetone.

4.6 Screening data

In the last years, in addition to the target analyses, screening techniques are applied to monitor the surface water of the Meuse. With these screenings a broad range of chemicals are monitored. Instead of measuring pure standards, the mass spectra of compounds are compared with spectra in a library. If a match is found, the identity of the the compound can be determined. If a match is not found, the compound is indicated as "unknown". The results from the screenings performed along the Meuse are summarized in the RIWA Meuse report of 2013, 2014 and 2015 (RIWA Meuse, 2013, 2014, 2015). In this report, the most frequently detected compounds are displayed. The most often found compounds fall in the category of pharmaceuticals, industrial compounds and compounds used in consumer products. In Table 6, a list is given of the most frequently detected known compounds in 2014 that are not measured in target analyses or are not yet evaluated. These compounds qualify to be included on list 2, in case an affordable measuring technique is available.

Table 6. *Compounds detected in 2014 in the surface water of the Meuse with screening techniques. Compounds that are part of target analyses or have been evaluated earlier are not included*

Compound	CAS	Category	Target analysis available?
Pharmaceuticals			
3,4-Dimethoxyphenethylamine	120-20-7	neurotransmitter	?
3,4-Methylenedioxyethylamphetamine	82801-81-8	amphetamine	?
Amisulpride	71675-85-9	anti-psychotic	Yes
Butetamate	14007-64-8	bronchodilator	?
Celiprolol	57470-78-7	beta-blocker	?
Certomycin	56391-57-2	antibiotic	?
Cetobemidone	5965-49-1	painkiller	?
Ciclacilline	3485-14-1	antibiotic	?
Cyclopentamine	102-45-4	decongestant	?
Eprosartan	133040-01-4	antihypertensive	?
Etilefrine	709-55-7	antihypotensive	?
Flecaïnide	54143-55-4	anti-arritmicum	?
Gabapentin	60142-96-3	anticonvulsant	Yes
Lamotrigine	84057-84-1	anticonvulsant	Yes
Meperidine/ pethidine	57-42-1	(narcotic) analgesic	?
N-methyl-1-(1,3-benzodioxol-5-yl)-2-aminobutane	103818-46-8	amphetamine	?
Oxilofrine	365-26-4	antihypotensive	?
Sulpiride	15676-16-1	anti-psychotic	?
Telmisartan	144701-48-4	antihypertensive	Yes
Thymopentin	177966-81-3	immunostimulant	?
Tramadol	27203-92-5	painkiller	Yes
Valsartan	137862-53-4	antihypertensive	Yes
Venlafaxine	93413-69-5	serotonin inhibitor	Yes
Industrial compounds			
4,4-Sulfonyldifenol	80-09-1	Industrial compound	?
Aniline	62-53-3	Industrial compound	Yes
Benzylalcohol	100-51-6	Solvent	Yes
Capric acid	334-48-5	Industrial compound	?
Propiophenone	93-55-0	Industrial compound	?
Tetra-acetyl-ethylene-diamine (TAED)	1054305-70-4	Industrial compound	?
Tri-phenyl-phosphine oxide (TPPO)	791-28-6	Industrial compound	?
Pesticides and biocides			
Dettol (chloroxylenol)	88-04-0	Biocide	?
Oxadiazon	19666-30-9	Herbicide	Yes
Unknown application			
1,2,3-Propanetriol, 1-nitrate	?	?	?
1,2-Ethanediol, dinitrate	?	?	?
3-Hexanone-2.5-dimethyl-4-nitro	?	?	?
5-Methyl-1-hexene	?	?	?
Cyclotetradecane	?	?	?

5. Conclusions

The criteria set by Fischer et al. (2011) for selecting the relevant and potentially relevant compounds were reviewed. This resulted in an adapted set of criteria. The most notable changes were the renaming of the lists from C, B, A into 1, 2 and 3, the deletion of list B (list of potentially relevant compounds) and the introduction of list 3. This resulted in a system with the following three lists:

- List 1: Drinking water relevant compounds
- List 2: Candidate drinking water relevant compounds
- List 3: No longer drinking water relevant compounds

A scoring system for the list of relevant compounds was used. The compounds were scored based on removal by water treatment, toxicity, odor/taste threshold and public perception. The compounds on the former list A, B and C were re-ranked using the new criteria. Based on a literature study on emerging organic compounds in surface waters in Europe, and screening data from the drinking water laboratories, new potentially relevant compounds were proposed as candidates for the list of drinking water relevant compounds and placed on list 2. The resulting lists are given in Table 7.

Table 7. *The three lists of compounds as finally proposed in this report.*

List 1: Drinking water relevant compounds (inlcuding scores)					
Pharmaceuticals		**Pesticides**		**Industrial compounds**	
Ibuprofen	15	Desphenyl chloridazon	11	Acetone	11
Metformin + Guanylurea	29	DEET	10	DIPE	19
Metoprolol	10	DMS	11	DTPA	13
Paroxetine	16	Glyphosate + AMPA	11	EDTA	13
Sotalol	10	Isoproturon	16	Fluoride	>18
X-ray contrast agents		Nicosulfuron	11	NTA	19
Amidotrizoic acid	11	Terbuthylazine	16	**Plasticizer**	
Iohexol	12	**Polycyclic aromatic hydrocarbon**		DEHP	17
Iomeprol	12	Benzo(a)pyrene	18		
Iopamidol	12	**Hormone disturbing compounds**			
Iopromide	12	ER-Calux (bioassay for estrogenic activity)	27		

Table 7 continuation. *The three lists of compounds as finally proposed in this report.*

List 2: Candidate drinking water relevant compounds			
Pharmaceuticals[1]		**Industrial compounds**	**Hormone disrupting compounds**
4-FAA (metabolite metamizol)	Gabapentin	Benzylalcohol	Anti-AR CALUX
Propyphenazone	Lamotrigine	Aniline	GR-CALUX
Tramadol	Citalopram	Melamine	Bisfenol A
4-AAA (metabolite metamizol)	O-Desmethylvenlafaxine	Pyrazole[2]	**Pesticides/biocides**
Amoxicillin	Venlafaxine	Tert-butyl alcohol (metabolite MTBE)	3,5,6-TCP (chlorpyrifos + triclopyr metabolite)
Ciprofloxacin	Fluconazole	Urotropine	Metazachlor ethane sulfonic acid
Clarithromycin	Irbesartan		Metazachlor oxalic acid
Clindamycin	Telmisartan		Metolachlor ethane sulfonic acid
Erythromycin	Valsartan		Oxadiazon
Roxithromycin	Amisulpride		

[1] The compounds that are depicted in orange are selected for the RIWA monitoring programme of 2016
[2] Pyrazole has a clear emitting source and it is proposed to monitor the compound only at the monitoring stations downstream from this source.

List 3: No longer drinking water relevant compounds			
Pharmaceuticals	**Pesticides**	**Industrial compounds**	**Plasticizers**
Aspirin	2,4-D	4-n-Nonyl phenol	BBP
Carbamazepine	BAM	Diglyme	DBP
Diclofenac	Carbendazim	Benzotriazole	DEP
Galaxolide	Chloridazon	BPS	DIBP
Lincomycin	Chlorotoluron	DMSA	N-butylbenzenesulphonamide
Naproxen	Dimethenamid	ETBE	TBP
Phenazone	Diuron	MTBE	TCEP
Salicylic Acid	MCPA	NDMA	TCPP
Sulfamethoxazole	Mecoprop	Surfynol 104	**Perfluorinated compounds**
Drugs of abuse	Metazachlor	Tolyltriazole	PFBA
Barbital	Methyl-desfenylchloridazon	**Fragrances and musks**	PFBS
Pentobarbital	Metolachlor	AHTN	PFHxS
Phenobarbital	**Artificial sweeteners**	Musk (ketone)	PFOA
Hormones	Acesulfame-K	Musk (xylene)	PFOS
Estrone	Sucralose		

6. Recommendations

- It is recommended to use the proposed lists for the monitoring effort by the drinking water companies along the river Meuse.
- It is recommended to update the list of relevant compounds (list 1) on a two-yearly basis. It is also recommended to subdivide the list of candidates for the list of relevant compounds in several parts (list 2) and monitor each part during one year. After this year, it will be evaluated if the detected compounds of list 2 are eligible to enter list 1. In the next year, the focus in the monitoring programme can be placed on another part of list 2. In this way, the monitoring effort is spread over a few years and list 2 stays flexible and up to date.
- It is recommended to keep a list of "no longer drinking water relevant compounds" (list 3), to preserve compounds that drinking water companies feel should still be measured. Also it helps to have a clear view on compounds that may no longer be relevant to the drinking water function of the Meuse. These compounds can be removed or re-checked at a later time if there is suspicion that they have become more relevant, e.g. due to their increased use.
- It is proposed to check compounds of emerging concern for new analytical methods and toxicological data in the future, to help identify more compounds of emerging concern, and obtain sufficient information to determine if they should be placed on list 2.
- If analytical methods are not available for compounds that are identified as candidates for list 2, it is recommended to develop an analytical method for these compounds (or check the possibilities of adding these compounds to an existing method). In Appendix 6 is indicated which compounds it concerns.

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Appendix 1 — Calculation of compound score

The scoring system used was earlier described in (Fischer et al., 2011).

The list of compounds that are relevant to the drinking water function of the River Meuse are proposed to be scored, according to the following principles:

1. The main chemical properties that influence the removal by water treatment; polarity, volatility and removal by powdered activated carbon are ranked:

a) For polarity the log K_{ow} of the compound is used.

b) For volatility the vapor pressure of the compound is used.

c) For biodegradability of the compound the primary biodegradation model (BioWIN3, in EPI Suite 4.1) is used.

2. The toxicological benchmark quotient (BQ) is derived for each compound. BQ is the maximum concentration found in the river (C_{max} water) divided by the provisional toxicological drinking water guideline value (pGLV):

$$pGLV = \frac{TDI * m_{adult} * 10\%}{2L/day}$$

Where TDI is the tolerable daily intake in µg (kg body mass)$^{-1}$ day^{-1}, and m_{adult} is the average adult body mass in kg. For the calculations a m_{adult} of 60 kg is assumed.

Table 8. *Point attribution for polarity, volatility, biodegradability, and toxicity.*

Polarity		**Volatility**		**Biodegradability**		**Toxicity**	
Log K_{ow}	Score	Vapor pressure (mm Hg)	Score	BioWIN3	Score	BQ	Score
>6	0	>52.5	0	>4.75 – 5	0	<0.01	0
>3 - 6	1	>35 – 52.5	1	>3.25 – 4.75	1	0.01 – 0.1	6
0 – 3	2	17.5 – 35	2	2.25 – 3.25	2	>0.1 – 1	12
<0	3	<17.5	3	<2.25	3	>1	18

3. If the odor/taste threshold is breached by C_{max} water, 3 points are awarded.

4. If the compound belongs to one of the following categories: pharmaceutical, pesticide, hormone, or hormone disruptor, or is a metabolite of a compound from one of these categories, it is considered harmful to the public perception of the drinking water consumers and 3 points are awarded.

Appendix 2 — Criteria of the lists from 2011

Criteria and list of relevant compounds (list C)

The criteria for list C as set in 2011 are listed below.

Criteria C

If the BQ of the compound is greater than 1, criteria 1, 3, and 4 can be neglected.

1. The compound is hardly removed by basic treatment, i.e. physical treatment (coagulation, aeration, rapid sand filtration) and disinfection (by chlorination, ozone, or UV).
   and
2. The compound was observed on at least two RIWA Meuse monitoring stations or intake points in the period 2005 – 2010 (for a minimum of three years*).
   and
3. The compound was found to exceed DMR target values or the Drinking Water Standards on at least two RIWA Meuse monitoring stations or intake points at least once in the period 2005 – 2010 (taking into account possible removal by conventional treatment).
   and
4. The compound was found to exceed the drinking water standard or the DMR target value used by the drinking water companies, at least once in the period after 2008.
   and
5. The total score of the compound when prioritized has to be 10 or greater.

*If the compound is monitored >13 times per year, it has to be detected at two or more RIWA Meuse monitoring stations with a frequency of at least 8% of the measurements per year (this criterion is equivalent to the criterion requiring that the compound is detected at least once a year with a monitoring frequency of 13 times a year).

If a compound is considered relevant to the drinking water function of the river Meuse, it should be included in an additional monitoring campaign and should be measured at least 13 times a year by all members of RIWA Meuse at relevant monitoring stations and measuring points.

Criteria and list of potentially relevant compounds (list B)

The criteria for list B as set in 2011 are listed below.

Criteria B

1. The compound was observed at two or more RIWA Meuse monitoring stations during the last year.
   and
2. The compound is difficult to remove by basic treatment, i.e. physical treatment (coagulation, aeration, rapid sand filtration) and disinfection (by chlorination, ozone or UV).
   and
3. The compound has undesirable properties for the production of drinking water:
   - The benchmark quotient (BQ, see Appendix 1), if information is available is greater than 0.1.

   or
   - The compound has a low odor/taste threshold.

   or
   - The compound is considered harmful to the public perception of the drinking water quality.

When a compound is considered potentially relevant to the drinking water function of the river Meuse, it should be included in an additional monitoring campaign and should be measured at least 4 times a year by all members of RIWA Meuse at relevant monitoring stations and measuring points (Van den Berg, 2009).

Criteria and list of future potentially relevant compounds (list A)

The criteria for list A as set in 2011 are listed below.

Criteria A

- The compound is present in the river Meuse at concentrations well above the DMR target value.
  or
- The concentration of the compound is expected to increase due to increased use in the catchment area in the near future (e.g. due to a change in usage of pesticides).
  or
- The compound has undesirable properties for the production of drinking water and is expected to be present in the river Meuse (based on research).
  and
  - The compound can be monitored with an affordable measuring technique.

Appendix 3 — Compound lists from 2011

The list of relevant compounds from 2011 is shown in **Table 9**. The score was calculated as described in Appendix 1. The list of drinking water relevant compounds is shown in **Table 9**. The list of potentially drinking water relevant is shown in **Table 10** and the list of future compounds potentially relevant to the drinking water function future is shown in **Table 11**.

Table 9. *The 2011 list of compounds relevant to the drinking water function of the river Meuse.*

Compound	CAS #	Category	Score
Benzo(a)pyrene	50-32-8	PAK	24
Diuron	330-54-1	Pesticide	22
MCPA (4-chloro-2-methylphenoxyacetic Acid)	94-74-6	Pesticide	21
DIPE (Di-isopropyl Ether)	108-20-3	Industrial	19
EDTA	64-02-8	Industrial	19
2,4-D	94-75-7	Pesticide	16
Chlorotoluron	15545-48-9	Pesticide	16
Isoproturon	34123-59-6	Pesticide	16
s-Metolachloor	51218-45-2	Pesticide	16
Diclofenac	15307-86-5	Pharmaceutical	15
Mecoprop (MCPP)	93-65-2	Pesticide	15
MTBE (Methyl-tert-butylether)	1634-04-4	Industrial	13
Nicosulfuron	111991-09-4	Pesticide	11
TBP (Tributylphosphate)	126-73-8	Industrial	11
Glyphosate	1071-83-6	Pesticide	11
Carbamazepine	298-46-4	Pharmaceutical	10
Carbendazim	10605-21-7	Pesticide	10
Chloridazon	1698-60-8	Pesticide	10
Metoprolol	37350-58-6	Pharmaceutical	10

Table 10. *The 2011 List of compounds potentially relevant to the drinking water function of the river Meuse. Highlighted rows indicate a compound that was measured at less than three monitoring stations.*

Compound	CAS #	Category	Undesirable property
Acetylsalicylic acid	50-78-2	Pharmaceutical	Public perception
Caffeine	58-08-2		
Phenazone	60-80-0		
Iopromide	73334-07-3		
Lincomycin	154-21-2		
Naproxen	22204-53-1		
Sotalol	3930-20-9		
Sulfamethoxazole	723-46-6		
BAM	2008-58-4	Pesticide	Public perception
DEET	134-62-3		
Dimethenamid	87674-68-8		
DMSA (N,N-dimethylaminosulfanilide)	4710-17-2		
Metazachlor	67129-08-2		
Diglyme	111-96-6	Industrial compound	
ETBE (Ethyl-tertiairy-butyl-ether)	637-92-3		Odor and taste threshold of 1-2 µg/L
Fluoride	16984-48-8		
TCEP (tris(2-chloroethyl) phosphate)	115-96-8		
Estrogenic activity		Endocrine disrupting compound	BQ>0.1, public perception
p,p-sulfonyldiphenol	80-09-1	Suspected endocrine disrupting compound	Public perception

Table 11. *Compounds future potentially relevant to the drinking water function of the river Meuse from 2011.*

Compound	CAS #	Category	References
Pheno barbital	50-06-6	Drugs of abuse (tranquilizers)	Van der Aa, 2010
Pento barbital	76-74-4		Van der Aa, 2010
Barbital	57-44-3		Van der Aa, 2010
Sucralose	56038-13-2	Artificial sweeteners	Scheurer, 2009; Scheurer, 2010
Acesulpham	55589-62-3		Scheurer, 2009; Scheurer, 2010
Musk xylene	81-15-2	Fragrances and musks	Kumar, 2010; Lee, 2009
Musk ketone	81-14-1		Kumar, 2010; Lee, 2009
HHCB (galaxolide)	1222-05-5		Murray, 2010; Lee, 2009; monitoring HWL
6-Acetyl-1,1,2,4,4,7-hexamethyltetraline			Murray, 2010; Lee, 2009
PFOS (perfluorooctanoic sulfonate)	1763-23-1	Perfluorinated compounds	Murray, 2010; Nakayama, 2010; Houtman, 2010; monitoring HWL
PFOA (perfluorooctanoic acid)	335-67-1		Murray, 2010; Nakayama, 2010; Houtman, 2010; monitoring HWL
PFBA (perfluorobutanoic acid)	375-22-4		Nakayama, 2010; monitoring HWL
PFBS (perfluorobutanesulfonic acid)	29420-49-3		Monitoring HWL
PFHxS (perfluorohexane sulfonate)	432-50-7		Monitoring HWL
4-Nonylphenol	104-40-5	Industrial compounds	Murray, 2010; Calderon-Precidado, 2011
Benzotriazole	95-14-7		Murray, 2010
Tolytriazole	29385-43-1		Murray, 2010
NDMA (nitrosodimethylamine)	62-75-9		Houtman, 2010
Surfynol 104 (2,4,7,9-tetramethyl-5-decyne-4,7-diol)	126-86-3		Monitoring HWL
TCPP (tris(chloropropyl)phosphate)	13674-84-5		Monitoring HW
DEP (diethyl phthalate)	84-66-2	Plasticizers/ flame retardants	Murray, 2010; Houtman, 2010; monitoring HWL
DBP (dibutyl phthalate)	84-74-2		Murray, 2010; Houtman, 2010; monitoring HWL
DEHP (diethylhexyl phthalate)	117-81-7		Murray, 2010; Houtman, 2010; monitoring HWL
DIBP (diisobutylphthalate)	84-69-5		monitoring HWL
BBP (benzylbutylphthalate)	85-68-7		monitoring HWL
n-Butyl-benzenesulfonamide	3622-84-2		monitoring HWL
Metformin	657-24-9	Pharmaceuticals	monitoring HWL

Appendix 4 — Toxicological endpoints used to calculate Benchmark Quotient (BQ)

Table 12. *Toxicological endpoints and references for compounds relevant to the drinking water function of the river Meuse (List 1 of 2015). The numbers highlighted in purple are the numbers used to calculate the BQ.*

Compound	CAS #	NOAEL (mg/kg day)	ADI/TDI (µg/kg day)	(Provisional) drinking water limit (µg/L)	C_{max} 2010-2014 (µg/L)	BQ	Reference[1]
Guanylurea (metabolite metformin)	141-83-3	-	-	0.1	3.50	35.0	-
Metformin	657-24-9	-	62	217	2.80	0.013	Houtman et al. 2014
ER-Calux (expressed in 17ß-estradiol equivalents)[B]	-	-	-	0.0038	0.0096	2.526	Brand et al. 2013
DIPE (di-isopropyl Ether)	108-20-3	-	-	150	18.73	0.125	Puijker et al. 2011
NTA	139-13-9	-	10	200	86.2	0.431	WHO (2011)
Fluoride	16984-48-8	-	-	1500	1710	1.14	WHO (2011)
Benzo(a)pyrene	50-32-8	-	-	0.7	0.088	0.126	WHO (2011)
DEHP	117-81-7	-	-	8	6.10	0.763	WHO (2011)
Isoproturon	34123-59-6	3.00	3.0	9	0.21	0.023	WHO (2011)
Paroxetine	61869-08-7	-	2.9	10	0.51	0.051	Houtman et al. 2014
Terbuthylazine	5915-41-3	-	-	7	0.48	0.069	WHO (2011)
Ibuprofen	15687-27-1	-	-	150	3.50	0.023	Versteegh et al. (2007)
DTPA	67-43-6	100	100	350	13.4	0.038	Schriks et al. (2010)

Compound	CAS #	NOAEL (mg/kg day)	ADI/TDI (µg/kg day)	(Provisional) drinking water limit (µg/L)	C_{max} 2010-2014 (µg/L)	BQ	Reference[1]
EDTA	60-00-4	-	-	600	54.0	0.090	WHO (2011)
Iohexol	66108-95-0	125000	125000	375000	0.32	<0.001	Versteegh et al. (2007)
Iomeprol	78649-41-9	4000	1900	5700	0.52	<0.001	Schriks et al. (2010)
Iopamidol	60166-93-0	1380	138333	415000	0.24	<0.001	Versteegh et al. (2007);
Iopromide	73334-07-3	833	83333	250000	0.78	<0.001	Versteegh et al. (2007)
Acetone	67-64-1	900	900	2700	40	0.015	http://www.epa.gov/iris/toxreviews/0128tr.pdf
Amidotrizoic acid	117-96-4		83333	250000	0.48	<0.001	Versteegh et al. (2007)
AMPA (metabolite glyphosate)	1066-51-9	32	320	900	4.20	0.005	WHO (2011)
Glyphosate	1071-83-6	-	-	900	0.66	0.001	WHO (2011)
Desphenyl chloridazon (metabolite chloridazon)	6339-19-1	15	100	300	2.38	0.008	ADI from EFSA (2007), pGLV calculated
Chloridazon	1698-60-8	5.40	54	189	0.13	0.001	Schriks et al. (2010)
DMS (metabolite tolylfluanide)	3984-14-3	-	2000	6000	0.29	<0.001	http://www.ctb.agro.nl/ctb_files/13912_01.html
Nicosulfuron	111991-09-4	199	200	700	0.49	0.001	Schriks et al. (2010)
DEET	134-62-3	100	1800	6250	0.70	<0.001	Schriks et al. (2010)
Metoprolol	37350-58-6	-	-	50	0.29	0.006	Schriks et al. (2010)
Sotalol	3930-20-9	1.10	11	40	0.22	0.006	Houtman et al. 2014

[1] In case an official drinking water limit was available from WHO or RIVM, this one was chosen over the calculated pGLV *
[2] A pGLV could not be calculated, so the threshold of toxicological concern is used as pGLV instead

Appendix 5 — Data for scoring

Table 13. *Score of drinking water relevant compounds based on removal by water treatment, toxicity, odor/taste threshold and public perception (List 1, 2015). If a parent compound and metabolite are both listed, they are coupled together based on the highest score*

Compound	CAS	Polarity (log K_{ow})[2]		Volatility (vapor pressure, mm Hg)		BioWIN3		BQ		Odor / taste threshold (µg/L)		Harmful for public perception		Total
Guanylurea (metabolite metformin)	141-83-3	-1.22	3	8.68×10^{-4}	3	2.97	2	35.0	18	-	0	True	3	29
Metformin	657-24-9	-2.64	3	7.58×10^{-5}	3	2.91	2	0.011	6	-	0	True	3	17
ER-Calux (expressed in 17ß-estradiol equivalents)[1]	-	**4.01**	1	1.99×10^{-9}	3	2.45	2	1.37	18	-	0	True	3	27
DIPE (di-isopropyl Ether)	108-20-3	**1.52**	2	**1.49×10^{2}**	0	2.69	2	0.125	12	10	3		0	19
NTA	139-13-9	-3.81	3	7.16×10^{-9}	3	3.62	1	0.43	12	-	0		0	19
Fluoride	16984-48-8	[3]		[3]		[3]		1.14	18	-	0		0	>18
Benzo(a)pyrene	50-32-8	**6.13**	0	**5.49×10^{-9}**	3	1.84	3	0.126	12	-	0		0	18
DEHP	117-81-7	**7.60**	0	**1.42×10^{-7}**	3	3.21	2	0.76	12	-	0		0	17
Isoproturon	34123-59-6	**2.87**	2	**2.47×10^{-8}**	3	2.67	2	0.024	6	-	0	True	3	16
Paroxetine	61869-08-7	3.95	1	4.79×10^{-8}	3	1.89	3	0.051	6	-	0	True	3	16
Terbuthylazine	5915-41-3	**3.21**	1	**1.12×10^{-6}**	3	1.76	3	0.038	6	-	0	True	3	16
Ibuprofen	15687-27-1	**3.97**	1	**1.86×10^{-4}**	3	2.96	2	0.023	6	-	0	True	3	15
DTPA	67-43-6	-4.91	3	1.21×10^{-16}	3	3.39	1	0.038	6	-	0		0	13
EDTA	60-00-4	-3.86	3	3.77×10^{-12}	3	3.5	1	0.090	6	-	0		0	13
Iohexol	66108-95-0	**-3.05**	3	4.06×10^{-29}	3	2.05	3	<0.001	0	-	0	True	3	12

Compound	CAS	Polarity (log K_{ow})[2]		Volatility (vapor pressure, mm Hg)		BioWIN3		BQ		Odor / taste threshold (µg/L)		Harmful for public perception		Total
Iomeprol	78649-41-9	**-2.79**	3	3.04×10^{-29}	3	1.98	3	<0.001	0	-	0	True	3	12
Iopamidol	60166-93-0	**-2.42**	3	1.33×10^{-30}	3	1.98	3	<0.001	0	-	0	True	3	12
Iopromide	73334-07-3	**-2.05**	3	1.59×10^{-28}	3	1.78	3	<0.001	0	-	0	True	3	12
Acetone	67-64-1	**-0.24**	3	**2.32×10^{2}**	0	3.05	2	0.015	6	-	0		0	11
Amidotrizoic acid	117-96-4	1.37	2	3.57×10^{-15}	3	1.69	3	<0.001	0	-	0	True	3	11
AMPA (metabolite glyphosate)	1066-51-9	-2.47	3	5.76×10^{-5}	3	2.98	2	0.005	0	-	0	True	3	11
Glyphosate	1071-83-6	**-3.40**	3	**1.58×10^{-8}**	3	3.21	1	0.001	0	-	0	True	3	10
Desphenyl chloridazon (metabolite chloridazon)	6339-19-1	-1.59	3	4.23×10^{-6}	3	2.73	2	0.008	0	-	0	True	3	11
Chloridazon	1698-60-8	**1.14**	2	**4.50×10^{-7}**	3	2.58	2	0.001	0	-	0	True	3	10
DMS (metabolite tolylfluanide)	3984-14-3	-1.11	3	9.80×10^{-2}	3	2.92	2	0.001	0	-	0	True	3	11
Nicosulfuron	111991-09-4	**0.01**	2	1.98×10^{-12}	3	1.91	3	0.001	0	-	0	True	3	11
DEET	134-62-3	**2.18**	2	**2.00×10^{-3}**	3	2.65	2	<0.001	0	-	0	True	3	10
Metoprolol	37350-58-6	**1.88**	2	**2.88×10^{-7}**	3	2.65	2	0.006	0	-	0	True	3	10
Sotalol	3930-20-9	**0.24**	2	1.34×10^{-9}	3	2.78	2	0.006	0	-	0	True	3	10

[1] The physical parameters are given for the compound 17ß-estradiol

[2] Data depicted in bold is experimental data, the other data is estimated (Reference: EPI Suite 4.1)

[3] Inorganic compound; outside estimation domain of EPI Suite v4.1

Appendix 6 — Available analytical methods for list 2 candidates

Table 14. In this table the candidate compounds for list 2 (from Chapter 4.4, 4.5 and 4.6) are summarized and per compound is indicated if an analytical method is available. The laboratories where a method is performed are mentioned (ALZ = Aqualab Zuid, BDS = Biodetection Systems, HWL = Het Waterlaboratorium, KWR = KWR Watercycle Research Institute, SWDE = La société wallone des eaux, TZW = Technologie Zentrum Wasser). The source refers to the source in which the compound was found as a potential candidate (literature (LIT), screening (SCR) or incidental emissions (EMI)). In the column Monitoring 2016 is indicated with a "yes" is the compound will be monitored as part of the RIWA monitoring programme of 2016, SCR means that the compound will be followed via the screening methodologies. In the last column is indicated if it is recommended (REC) to develop a new analytical method.

#	Compound	CAS number	Application	Method?	ALZ	BDS	HWL	KWR	SWDE	TZW	Source	Monitoring 2016	Develop method?
	Industrial compounds			-									
1	Benzylalcohol	100-51-6	Solvent	yes						A	SCR	SCR	
2	4,4-Sulfonyldifenol	80-09-1	Industrial compound								SCR	SCR	REC
3	Aniline	62-53-3	Industrial compound	yes						A	SCR	SCR	
4	Capric acid	334-48-5	Industrial compound								SCR	SCR	REC
5	Melamine	108-78-1	Resin	yes				A		B	LIT		
6	Propiophenone	93-55-0	Industrial compound								SCR	SCR	REC
7	Pyrazool	288-13-1	Industrial compound	yes	v		v				EMI	Yes	
8	Tert-butyl alcohol (metabolite MTBE)	75-65-0	Fuel component	yes				B		C	LIT		
9	Urotropine	100-97-0	Industrial compound	yes				C		D	other	Yes	
10	Dichloroaniline	-	Industrial compound								LIT	SCR	REC
11	Dichlorobenzene	-	Industrial compound								LIT	SCR	REC
12	Tetra-acetyl-ethylene-diamine (TAED)	1054305-70-4	Industrial compound								SCR	SCR	REC
13	Tri-phenyl-phosphine oxide (TPPO)	791-28-6	Industrial compound								SCR	SCR	REC
14	Tetramethylbutanedinitrile	3333-52-6	Industrial compound								LIT	SCR	REC
15	Tributylamine	102-82-9	Industrial compound								LIT	SCR	REC
16	3'-(Trifluoromethyl)acetophenone	349-76-8	Industrial compound								LIT	SCR	REC
	Hormone disrupting compounds			-									
17	Anti-AR CALUX	-	Anti-androgenic activity	yes		v					LIT		
18	GR-CALUX	-	Glucocorticoid activity	yes		v					LIT		
19	Bisfenol A	80-05-7	Industrial compound	yes			v				other	Yes	

#	Compound	CAS number	Application	Method?	ALZ	BDS	HWL	KWR	SWDE	TZW	Source	Monitoring 2016	Develop method?
	Pesticides/biocides			-									
20	Dettol (chloroxylenol)	88-04-0	Biocide								SCR	SCR	REC
21	3,5,6-TCP (chlorpyrifos + triclopyr met)	6515-38-4	Herbicide	yes						E	LIT		
22	Metazachlor ethane sulfonic acid	171118-09-5	Herbicide	yes				D		F	LIT		
23	Metazachlor oxalic acid	1231244-60-2	Herbicide	yes				D		F	LIT		
24	Metolachlor ethane sulfonic acid	172960-62-2	Herbicide	yes				D		F	LIT		
25	Oxadiazon	19666-30-9	Herbicide	yes							SCR	SCR	
	Vetreinary pharmaceuticals												
26	Ivermectin	70288-86-7	Veterinary parasiticide								LIT		REC
	Pharmaceuticals			-									
27	3,4-Methylenedioxyethylamphetamine	82801-81-8	Amphetamine								SCR	SCR	REC
28	N-methyl-1-(1,3-benzodioxol-5-yl)-2-aminobutane	103818-46-8	Amphetamine								SCR	SCR	REC
29	4-FAA (metabolite metamizol)	1672-58-8	Analgesic	yes				D		G	LIT		
30	Cetobemidone	5965-49-1	Analgesic								SCR	SCR	REC
31	Codein	76-57-3	Analgesic								LIT		REC
32	Meperidine/pethidine	57-42-1	Analgesic								SCR	SCR	REC
33	Nortramadol (metabolite tramadol)	80456-81-1	Analgesic								LIT		REC
34	Propyphenazone	479-92-5	Analgesic	yes				E		G	LIT		
35	Tramadol	27203-92-5	Analgesic	yes				E	v	G	LIT/SCR	Yes	
36	4-AAA (metabolite metamizol)	83-15-8	Analgesic	yes				D		G	LIT		
37	Flecainide	54143-55-4	Antiarrhythmic agent								LIT/SCR	SCR	REC
38	Amoxicillin	26787-78-0	Antibiotic	yes				F		H	LIT	Yes	
39	Certomycin	56391-57-2	Antibiotic								SCR		REC
40	Ciclacilline	3485-14-1	Antibiotic								SCR		REC
41	Ciprofloxacin	85721-33-1	Antibiotic	yes						H	LIT	Yes	
42	Clarithromycin	81103-11-9	Antibiotic	yes					v	H	LIT	Yes	
43	Clindamycin	18323-44-9	Antibiotic	yes				E		H	LIT	Yes	
44	Erythromycin	114-07-8	Antibiotic	yes				E		H	LIT	Yes	
45	Roxithromycin	80214-83-1	Antibiotic	yes						H	LIT	Yes	

#	Compound	CAS number	Application	Method?	ALZ	BDS	HWL	KWR	SWDE	TZW	Source	Monitoring 2016	Develop method?
46	Gabapentin	60142-96-3	Anticonvulsant	yes				D		G	LIT/SCR	SCR	
47	Lamotrigine	84057-84-1	Anticonvulsant	yes				D		G	SCR	SCR	
48	Citalopram	59729-33-8	Antidepressant	yes					v	G	LIT		
49	Mirtazapine	61337-67-5	Antidepressant								LIT		REC
50	O-Desmethylvenlafaxine	93413-62-8	Antidepressant	yes				E		G	LIT		
51	Venlafaxine	93413-69-5	Antidepressant	yes				E	v	G	LIT/SCR	SCR	
52	Fluconazole	86386-73-4	Antifungal medication	yes						I	LIT		
53	Fexofenadine	83799-24-0	Antihistamine								LIT		REC
54	Eprosartan	133040-01-4	Antihypertensive								SCR	SCR	REC
55	Irbesartan	138402-11-6	Antihypertensive	yes					v	J	LIT		
56	Telmisartan	144701-48-4	Antihypertensive	yes						J	SCR	SCR	
57	Valsartan	137862-53-4	Antihypertensive	yes						J	SCR	SCR	
58	Etilefrine	709-55-7	Antihypotensive								SCR	SCR	REC
59	Oxilofrine	365-26-4	Antihypotensive								SCR	SCR	REC
60	Amisulpride	71675-85-9	Anti-psychotic	yes						J	SCR	SCR	
61	Sulpiride	15676-16-1	Anti-psychotic								SCR	SCR	REC
62	Celiprolol	57470-78-7	Beta-blocker								SCR	SCR	REC
63	Butetamate	14007-64-8	Bronchodilator								SCR	SCR	REC
64	Cyclopentamine	102-45-4	Decongestant								SCR	SCR	REC
65	Cimetidine	51481-61-9	H_2-receptor antagonist					G			LIT		REC
66	Thymopentin	177966-81-3	Immunostimulant								SCR	SCR	REC
67	3,4-Dimethoxyphenethylamine	120-20-7	Neurotransmitter								SCR	SCR	REC
	Unknown application												
68	1,2,3-Propanetriol, 1-nitrate	?	?								SCR	SCR	REC
69	1,2-Ethanediol, dinitrate	?	?								SCR	SCR	REC
70	3-Hexanone-2.5-dimethyl-4-nitro	?	?								SCR	SCR	REC
71	5-Methyl-1-hexeen	?	?								SCR	SCR	REC
72	Cyclotetradecane	?	?								SCR	SCR	REC