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Results of monitoring installations of the Pilot Mineral Concentrate in 2019-2020

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Referaat

In het kader van de pilot Mineralenconcentraten (MC) is een monitoringsprogramma uitgevoerd, gesubsidieerd door het Ministerie van Landbouw, Natuur en Voedselkwaliteit, om gegevens te verzamelen over de huidige kwaliteit van mineralenconcentraten in relatie tot de kwaliteitscriteria van de pilot MC en de uitvoering van het productieproces. Deze criteria zijn vastgesteld teneinde een hoge stikstofgebruiksefficiëntie van het concentraat te waarborgen zonder schadelijke gevolgen voor het milieu. De monitoring was gekoppeld aan informatie die werd gevraagd door het EU Joint Research Centre (JRC) ter ondersteuning van de ontwikkeling van kwaliteitscriteria voor stikstofhoudende meststoffen verkregen uit dierlijke mest, genaamd RENURE, in het kader van de Nitraat Richtlijn. De monitoring was bovendien gekoppeld aan relevante gegevens over de kwaliteit van het permeaat uit omgekeerde osmose (OO) voor onderbouwing van kwaliteitseisen voor het lozen op oppervlaktewater van effluënten uit mestverwerkingsinstallaties. Tot het monitoringsprogramma behoorden primaire en secundaire nutriënten, zware metalen, antibiotica, pathogene bacteriën en virussen. In dit rapport zijn de resultaten van de monitoring bijeengebracht.

Abstract

Within the framework of the Dutch pilot Mineral concentrate a monitoring program was conducted, subsidised by the Ministry of Agriculture, Nature and Food Quality, to generate data on the current quality of mineral concentrates, in relation to the criteria of the pilot Mineral concentrate (MC) of the Netherlands, and the performance of their production processes. These criteria are established to ensure high nitrogen use efficiency without an environmental impact of the concentrate. The monitoring was linked to information requested by the EU Joint Research Centre (JRC) to support the development of quality criteria for N fertilisers recovered from animal manure, referred to as RENURE, within the context of the Nitrates Directive. It was also linked to relevant data on the quality of the permeate from reverse osmosis to substantiate quality requirements for discharge on surface water of effluents from manure processing plants. The parameters of the monitoring included primary and secondary nutrients, heavy metals, antibiotics, pathogenic bacteria and viruses. This report presents the results of the monitoring.

Keywords: Reverse Osmosis, mineral concentrate, pig slurry, monitoring, minerals, nutrients, heavy metals, antibiotics, pathogens, viruses.

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Table of contents

Summary	5
Samenvatting	7
1 Introduction	9
2 Materials and methods	11
2.1 Production process	11
2.2 Pilot installations	13
2.3 Monitoring program	13
2.4 Laboratory analyses	15
2.4.1 Chemical parameters	15
2.4.2 Biological parameters	18
2.5 Data processing	19
3 Results	20
3.1 Composition of input	20
3.1.1 General parameters	20
3.1.2 Heavy metals	20
3.1.3 Antibiotics	20
3.1.4 Biological parameters	21
3.2 Composition of RO-concentrate	25
3.2.1 General parameters	25
3.2.2 Heavy metals	25
3.2.3 Antibiotics	25
3.2.4 Biological parameters	25
3.2.5 Average composition of RO-concentrate	31
3.2.6 Correlation between general parameters in RO-concentrate	32
3.3 Effect of installation and input on RO-concentrate	33
3.4 Test of RO-concentrate to quality criteria	36
3.4.1 Criteria pilot MC	36
3.4.2 Criteria RENURE	39
3.5 Composition of process flows	43
3.5.1 Input	43
3.5.2 Solid fraction	43
3.5.3 RO-input	43
3.5.4 RO-concentrate	43
3.5.5 RO-permeate	44
3.6 Comparison of type 1 and type 2 processes	48
4 Discussion and conclusions	50
References	55
Appendices	56

Summary

Within the framework of the Dutch pilot Mineral concentrate Wageningen Livestock Research conducted a monitoring program, subsidised by the Ministry of Agriculture, Nature and Food Quality, to generate data on the current quality of mineral concentrates, in relation to the criteria of the pilot Mineral concentrate (MC) of the Netherlands, and the performance of their production processes. These criteria are established to ensure high nitrogen use efficiency without an environmental impact of the concentrate. The monitoring was linked to information requested by the EU Joint Research Centre (JRC) to support the development of quality criteria for N fertilisers recovered from animal manure, referred to as RENURE, within the context of the Nitrates Directive. It was also linked to relevant data on the quality of the permeate from reverse osmosis to substantiate quality requirements for discharge on surface water of effluents from manure processing plants.

Complying with the quality criteria of the pilot MC and of RENURE can qualify the mineral concentrate as a high-end N fertiliser with chemical mineral nitrogen fertiliser characteristics that is safe to use, with minimal environmental impact and risk for human health. Results from former monitoring campaigns between 2009 and 2014 and a snapshot in 2018 showed that only part of the mineral concentrates met the criteria of the pilot MC.

The continued monitoring included 9 companies producing mineral concentrate (RO-concentrate) from pig slurry, 8 of them managing two production facilities each, a total of 17 installations. The general production process followed 3 main steps: (1) mechanical separation of raw slurry into a solid fraction and a liquid fraction, with the use of one or more coagulants, (2) removal of suspended solids and colloid particles from the liquid fraction by means of dissolved air flotation and filtration and (3) concentration of the polished liquid fraction by reverse osmosis producing a concentrate and leaving a permeate (water). At the 17 installations that were involved, in the period September 2019 - April 2020 three to six sampling and measuring rounds were carried out. The main process flows were sampled: raw slurry, solid fraction, RO-input (liquid fraction after polishing), RO-concentrate and RO-permeate. At 9 installations all process flows were sampled, at 8 installations RO-concentrate only. The parameters of the monitoring included primary and secondary nutrients, heavy metals, antibiotics, pathogenic bacteria and viruses. Bacteria and viruses were measured only twice.

The results from this monitoring campaign show that RO-concentrate can be characterized as a largely mineral fertiliser with N and K as the main nutrients. The greater part of Total-N is present as total ammoniacal nitrogen (TAN). Total organic carbon (TOC), S and Cl⁻ are the other components present in relatively high concentrations. The difference in concentrations of nutrients in the RO-input and the RO-concentrate is an increase of a factor 2.4 on average. RO-concentrates show detectable total concentrations of the metals Ba, Cr, Cu, Ni, Pb and Zn, with highest levels for Cu and Zn (>1 mg/kg) as can be expected through the use of Cu and Zn feed-additives. As to antibiotics, oxytetracycline, doxycycline and sulfadiazine are most frequently found, with concentrations varying from 1 to 100 µg/kg. These are the antibiotics most often found in raw pig slurry, which could be expected regarding the relatively high amounts used in pig husbandry. Bacteria and viruses were detected in nearly all RO-concentrates; concentrations varied log 2 – 4 (CFU/g) fold between installations. Concentrations of biological parameters in RO-concentrate are highly correlated to concentrations in RO-input (liquid fraction after polishing).

Significant differences in concentrations of general parameters between the RO-concentrates from different installations are observed, especially between installations (type 1) that use two or three coagulants and/or flocculants as aids for polishing, to improve separation efficiency of the raw slurry, and installations (type 2) that uses only one organic poly-electrolyte as a coagulant/flocculant. Compared to the RO-concentrates of type 2 the type 1 installations show higher concentrations of TS, N, TAN, S, Na and Cl while the concentrations of VS, Total-C, TOC, and P are lower. The average TAN/Total-N ratios of type 1 and type 2 installations are 0.90 and 0.78 resp. Concentrations of heavy metals, antibiotics and of *E.coli* and enterococci are significantly higher in the RO-concentrates of type 2 installations. From these findings it is concluded that type 1 installations effectively remove more

suspended solids from the liquid fraction which results in RO-concentrates that can meet proposed RENURE criterions.

A minority of the pilot installations produces on average an RO-concentrate that meets the pilot MC criterion $\text{TAN/Total-N} \geq 0.90$. The RO-concentrates of all type 2 installations do not meet this criterion. This observation means that the current RO-concentrate cannot be qualified as a mineral N-fertiliser with chemical mineral nitrogen fertiliser characteristics. Two installations of type 2 do not meet the criterions $\text{Total-N/P}_2\text{O}_5 \geq 15$ and $\text{EC} \geq 50$ mS/cm. All pilot installations meet all other RENURE quality criterions, which proves that the proposed RENURE criterions are less sharp and easier to meet than the pilot MC criterions.

Comparison of the composition of the RO-concentrates to the quality criterions of the pilot MC and of RENURE leads to the following results.

	Criterion	Percentage of measurements that meet criterion	Number of pilot installations that meet criterion		
			Type 1	Type 2	Total
Pilot MC	$\text{NH}_4\text{-N / Total-N}^1 \geq 0.90$	26	3	0	3
	$\text{Total-N / P}_2\text{O}_5^1 \geq 15$	86	12	3	15
	$\text{EC} \geq 50$ mS/cm	71	11	3	14
RENURE	$\text{NH}_4\text{-N / Total-N} \geq 0.90$	26	3	0	3
	$\text{TOC / Total-N} \leq 3$	100	12	5	17
	$\text{Cu} \leq 300$ mg/kg DM	100	12	5	17
	$\text{Zn} \leq 800$ mg/kg DM	100	12	5	17

¹ measured in wet product (w/w)

RO-permeate, the end product of the treatment process without fertiliser value, is water that is preferably discharged to surface water. For discharge to surface water high quality standards apply to prevent a burden on the aquatic environment and risk to human health. Based on the indications from previous studies and expert judgement, reverse osmosis is currently classified as best available technic (BAT) for producing dischargeable effluent. However, the results from this monitoring campaign show that the RO-permeate (after ion exchange) is not pure demineralised water as could be expected from RO filtration. The presence of several parameters in the permeate of a majority of the pilot installations could indicate suboptimal pre-treatment of the RO-input, deficient construction of the RO-unit and inadequate cleaning procedure of the RO-membranes.

This monitoring campaign has shown that most of the pilot installations are able to produce an RO-concentrate that meet the quality requirements but also that between installations there is a great variation in the composition of the RO-concentrate. This indicates that the way the installations are operated and the way they are maintained are success factors for optimal performance to achieve high quality end products. The production process needs to be further improved to meet the quality requirements of both concentrates and permeates. In this respect the following is recommended:

- improve pre-treatment technology and operation,
- aim for a constant and 'clean' polished RO-input, to minimise fouling and damage and to extend the durability of the RO-membranes,
- carefully choose the right type and dosage of flocculants and supporting additives, in tune with the raw slurry, to improve the first separation step; the use of PAM only is discouraged,
- optimize the cleaning procedure of the RO-unit, to improve the performance RO-membranes,
- use high quality RO-membranes that are designed for this input from animal manure.

Samenvatting

Wageningen Livestock Research heeft in het kader van de pilot Mineralenconcentraten (MC) een monitoringsprogramma uitgevoerd, gesubsidieerd door het Ministerie van Landbouw, Natuur en Voedselkwaliteit, om gegevens te verzamelen over de huidige kwaliteit van mineralenconcentraten in relatie tot de kwaliteitscriteria van de pilot MC en de uitvoering van het productieproces. Deze criteria zijn vastgesteld teneinde een hoge stikstofgebruiksefficiëntie van het concentraat te waarborgen zonder schadelijke gevolgen voor het milieu. De monitoring was gekoppeld aan informatie die werd gevraagd door het EU Joint Research Centre (JRC) ter ondersteuning van de ontwikkeling van kwaliteitscriteria voor stikstofhoudende meststoffen verkregen uit dierlijke mest, genaamd RENURE, in het kader van de Nitraat Richtlijn. De monitoring was bovendien gekoppeld aan relevante gegevens over de kwaliteit van het permeaat uit omgekeerde osmose (OO) voor onderbouwing van kwaliteitseisen voor het lozen op oppervlaktewater van effluënten uit mestverwerkingsinstallaties.

Indien aan de criteria van de pilot MC en RENURE wordt voldaan kan mineralenconcentraat worden gekwalificeerd als een hoogwaardige N meststof, met de eigenschappen van kunstmest, die veilig kan worden toegepast met minimale milieu-impact en humane gezondheidsrisico's. Resultaten van eerdere monitoringscampagnes, tussen 2009 en 2014, en een momentopname in 2018 laten zien dat slechts een deel van de mineralenconcentraten voldeden aan de criteria van de pilot MC.

De monitoring is in 2019 voortgezet met 9 producenten van mineralenconcentraten, waarvan 8 een samenwerking waren aangegaan met een andere producent, totaal 17 productie-installaties. Het algemene productieproces bestond uit 3 processtappen: (1) mechanische scheiding van ruwe drijfmest in een vaste en vloeibare fractie, met toevoeging van één of meer vlokmiddelen, (2) verwijdering van gesuspendeerde vaste stoffen uit de vloeibare fractie door middel van *dissolved air flotation* (DAF) en filtratie en (3) concentratie van de vloeibare fractie middels omgekeerde osmose met als eindproducten een concentraat en een permeaat (water). Bij de 17 installaties werden 3 tot 6 monsternamen- en meetrondes uitgevoerd, waarbij de belangrijkste processtromen werden bemonsterd: ruwe mest, vaste fractie, OO-invoer (dunne fractie na zuivering), OO-concentraat en OO-permeaat. Bij 9 installaties werden alle processtromen bemonsterd, bij 8 installaties alleen OO-concentraat. Tot het monitoringsprogramma behoorden primaire en secundaire nutriënten, zware metalen, antibiotica, pathogene bacteriën en virussen.

De resultaten van deze monitoringscampagne laten zien dat OO-concentraat beschouwd kan worden als een grotendeels minerale meststof met N en K als belangrijkste waardegevendende bestanddelen. Het grootste deel van de stikstof bestaat uit ammonium ($\text{NH}_4\text{-N}$). Verder zijn totaal organisch koolstof (TOC), S en Cl^- in hoge concentraties aanwezig. Het concentratieverschil tussen OO-invoer en OO-concentraat bedraagt gemiddeld ongeveer een factor 2.4. In OO-concentraat zijn meetbare concentraties aan Ba, Cr, Ni, Pb en Zn aangetroffen, met de hoogste concentraties voor Cu en Zn ($> 1\text{mg/kg}$), wat gezien het gebruik als additieven in varkensvoer mocht worden verwacht. Van de antibiotica zijn oxytetracycline, doxycycline en sulfadiazine het meest frequent aangetroffen, in concentraties variërend van 1 tot $100\ \mu\text{g/kg}$. Deze antibiotica worden eveneens het vaakst in ruwe varkensmest aangetroffen, wat verwacht kan worden gezien de relatief grote hoeveelheden die in de varkenshouderij worden gebruikt. Bacteriën en virussen werden in bijna alle OO-concentraten aangetoond; concentraties varieerden tussen installaties met een factor 10^2 en 10^4 (CFU/g). De concentraties van biologische parameters in OO-concentraat zijn sterk gecorreleerd aan de concentraties in de OO-invoer.

Er zijn significante verschillen gevonden in de concentraties van algemene parameters tussen de OO-concentraten van verschillende installaties, met name tussen installaties van type 1, die polyacrylamide en ijzer- en/of aluminiumsulfaat als vlokmiddelen gebruiken voor verhogen van de efficiëntie van de eerste scheidingsstap, en installaties van type 2, die uitsluitend polyacrylamide (PAM) als vlokmiddel gebruiken. Type 1 installaties tonen in vergelijking met type 2 installaties hogere concentraties aan droge stof, N-totaal, $\text{NH}_4\text{-N}$, S, Na en Cl^- en lagere concentratie aan organische stof, C-totaal, TOC en P. De gemiddelde verhouding $\text{NH}_4\text{-N/N-totaal}$ van de OO-concentraten van type 1 en

type 2 installaties was 0.90 resp. 0.78. De concentraties aan zware metalen, antibiotica en *E.coli* en enterokokken in de OO-concentraten van type 2 installaties zijn significant hoger dan van type 1 installaties. Uit deze bevindingen wordt geconcludeerd dat de eerste scheidingsstap van type 1 installaties efficiënter plaatsvindt dan van type 2 installaties. Bij type 1 wordt meer gesuspendeerd (organisch) materiaal uit de dunne fractie verwijderd wat resulteert in OO-concentraten die voldoen aan de voorgestelde RENURE kwaliteitscriteria.

Een minderheid van de pilot installaties produceren gemiddeld een OO-concentraat dat voldoet aan het pilot kwaliteitscriterium $\text{NH}_4\text{-N/N-totaal} \geq 0.90$. De OO-concentraten van geen van de type 2 installaties voldoen aan dit criterium. Dit betekent dat het huidige OO-concentraat gemiddeld niet kan worden gekwalificeerd als een minerale meststof met de eigenschappen van kunstmest. De concentraten van twee type 2 installaties voldoen niet aan de pilot criteria $\text{N-totaal/ P}_2\text{O}_5 \geq 15$ en $\text{EC} \geq 50 \text{ mS/cm}$. De concentraten van alle pilot installaties voldoen aan de andere RENURE criteria. Dit geeft aan dat de voorgestelde RENURE criteria minder strikt zijn de pilot MC criteria.

Vergelijking van de samenstelling van de OO-concentraten met de kwaliteitscriteria van de pilot MC en van RENURE levert het volgende beeld op:

	Criterium	Percentage van de metingen dat voldoet aan het criterium	Aantal installaties dat voldoet aan het criterium		
			Type 1	Type 2	Totaal
Pilot MC	$\text{NH}_4\text{-N / Total-N}^1 \geq 0.90$	26	3	0	3
	$\text{Totaal-N / P}_2\text{O}_5^1 \geq 15$	86	12	3	15
	$\text{EC} \geq 50 \text{ mS/cm}$	71	11	3	14
RENURE	$\text{NH}_4\text{-N / Totaal-N} \geq 0.90$	26	3	0	3
	$\text{TOC / Totaal-N} \leq 3$	100	12	5	17
	$\text{Cu} \leq 300 \text{ mg/kg DS}$	100	12	5	17
	$\text{Zn} \leq 800 \text{ mg/kg DS}$	100	12	5	17

¹ gemeten in nat product, op gewichtsbasis

OO-permeaat, het eindproduct van het verwerkingsproces zonder bemestende waarde, is water dat bij voorkeur geloosd wordt op oppervlaktewater. Voor lozing gelden hoge kwaliteitseisen om schade aan het watermilieu en risico's voor de humane gezondheid te voorkomen. Gebaseerd op indicaties uit eerdere studies en *expert judgement*, wordt omgekeerde osmose momenteel gekwalificeerd als best beschikbare techniek (BBT) voor het produceren van een losbaar effluent. Echter, de resultaten van deze monitoringscampagne laten zien dat het OO-permeaat (na ionenwisseling) niet het pure gedemineralseerde water is dat na omgekeerde osmose verwacht zou mogen worden. De aanwezigheid van verschillende parameters in het permeaat van een meerderheid van de pilot installaties zou kunnen wijzen op suboptimale voorbehandeling van de OO-invoer, gebrekkige constructie van de OO-unit en ontoereikende reiniging van de OO-membranen.

Deze monitoringscampagne heeft aangetoond dat de meeste pilot installaties in staat zijn een OO-concentraat te produceren dat voldoet aan de kwaliteitseisen maar ook dat er tussen de installaties aanzienlijke prestatieverschillen bestaan wat tot uiting komt in de grote variatie in de samenstelling van de concentraten. Dit geeft aan dat de manier waarop de installaties worden bediend en onderhouden succesfactoren zijn voor optimale prestaties en het voortbrengen van eindproducten van hoge kwaliteit.

Het productieproces dient verder geoptimaliseerd te worden om aan de kwaliteitseisen te voldoen voor zowel het concentraat als het permeaat. Daarom wordt het volgende geadviseerd:

- verbeter de voorbehandelingstechniek en -exploitatie,
- streef naar een 'schone' OO-invoer, voor minimaliseren van vervuiling en beschadiging en verhogen van duurzaamheid van OO-membranen,
- kies zorgvuldig het juiste type en de juiste dosering van vlokmiddelen en ondersteunende additieven, die afgestemd zijn op de te verwerken mest (het gebruik van uitsluitend PAM wordt afgeraden),
- verbeter de reinigingsprocedure van de OO-unit, voor optimaal presteren van de OO-membranen,
- gebruik hoge kwaliteit RO-membranen die ontwikkeld zijn voor dit type invoer.

1 Introduction

The Dutch Pilot Mineral Concentrate started in 2009 to gain experience with processing of animal slurry into products comparable to liquid mineral NK-fertilisers. From 2009 to 2014 at 10 pilot installations a monitoring program was conducted from which information was generated on the chemical composition and also the nitrogen use efficiency and nitrogen fertiliser replacement value of the mineral concentrate product. In 2015 quality criteria (see textbox) were established that would guarantee the fertiliser efficiency and nitrogen fertiliser replacement value and minimize environmental impact of mineral concentrate produced from animal slurry. Based on these criteria producers were selected for participation in the follow up of the pilot. Today 9 companies, both individual pig farms and agricultural contractors, with a total of 17 installations (production facilities) in operation are involved in the pilot.

In 2018 the mineral concentrates from all pilot installations were analysed on contents of primary and secondary plant nutrients. This snapshot showed that only half of the products complied with the quality criterion mineral-N to total-N ratio $\geq 90\%$. Besides that, a large range in nutrient content of the products between participating installations was observed. Two installations were able to upgrade the quality of mineral concentrate by adding a second RO-unit, which lifted up the concentrations of nutrients in the RO-concentrate by 30% on average. However, further quality improvement of mineral concentrates is necessary to meet the quality requirements of N-fertilising products derived from animal slurry (RENURE) which are being established by the EU Joint Research Centre within the context of the Nitrates Directive 91/676/EC (Huygens et al, 2020). If RENURE criteria are met, mineral concentrate does not have to comply with the thresholds on the use of animal slurry (i.e. 170 kg N/ha or 230/250 kg N/ha on grassland based on the derogation for the Netherlands). For free trade as a fertilising product in the European Union mineral concentrates have to comply with the new European fertilising product regulation EC/2019/1009 (FPR), with criterions for quality and environmental and human safety. The new criterions on use and quality of both Nitrates Directive 91/676/EC and FPR are designed to stimulate a circular economy in EU.

Within the framework of the continued pilot in 2019 and 2020 a monitoring program was conducted to generate data on the current quality of mineral concentrates, in relation to the criteria of the pilot mineral concentrates of the Netherlands, and the performance of the production processes. The monitoring was linked to information requested by JRC to support development of quality criteria for high-end fertilisers produced from animal manure.

Quality criterions¹ for mineral concentrates

Pilot MC criteria of the Netherlands	TAN ² : Total-N ≥ 0.90
	Total-N : P ₂ O ₅ ≥ 15
	EC ≥ 50 mS/cm
RENURE quality criteria as proposed by Huygens et al, 2020	TAN : Total-N ≥ 0.90 or TOC : Total-N ≤ 3
	Cu ≤ 300 mg/kg DM
	Zn ≤ 800 mg/kg DM
	(Next to these chemical criterions general measures to prevent emissions are proposed)

¹ Ratios are based on concentrations (i.e. mg/kg or g/kg).

² TAN: total ammoniacal nitrogen

The monitoring program aims at:

- Survey on the current quality of mineral concentrates with comparison to quality standards of the Fertiliser Act of the Netherlands and the new European fertilising products regulation.
- Collection of relevant data to support JRC within the context of adjusting the EU Nitrates Directive.
- Collection of relevant data to substantiate general quality requirements, including so called precautionary parameters, for discharge on surface water of effluents from manure processing plants.
- Evaluation and proposals for improvement of the production process of mineral concentrates.

Chapter 2 of this report describes the current pilot installations which differ in processes, feedstocks and operational parameters. This chapter also justifies the methods of sampling and analytical procedures of measuring physical-chemical and biological parameters of the monitoring program. In chapter 3 results are presented on the composition of the input, throughput and output flows of the pilot installations. The report concludes with chapter 4, with an evaluation of the data followed by conclusions and general recommendations to meet Dutch and new European standards for quality and use for fertilising products made from slurry.

2 Materials and methods

2.1 Production process

This paragraph is based on Hoeksma and Buissonjé (2015).

The general concept of the production process of a mineral concentrate from animal slurry as studied in this project is shown in Figure 2.1. Raw or digested animal slurry is mechanically separated into a solid fraction and a liquid fraction after adding a coagulant and/or a flocculant. Suspended solids and colloid particles are removed from the liquid fraction by air flotation. Solid/liquid separation and air flotation may be applied in reversed order. The liquid fraction from mechanical separation or air flotation is filtered and subsequently concentrated by reverse osmosis, leaving a mineral concentrate and an effluent (permeate).

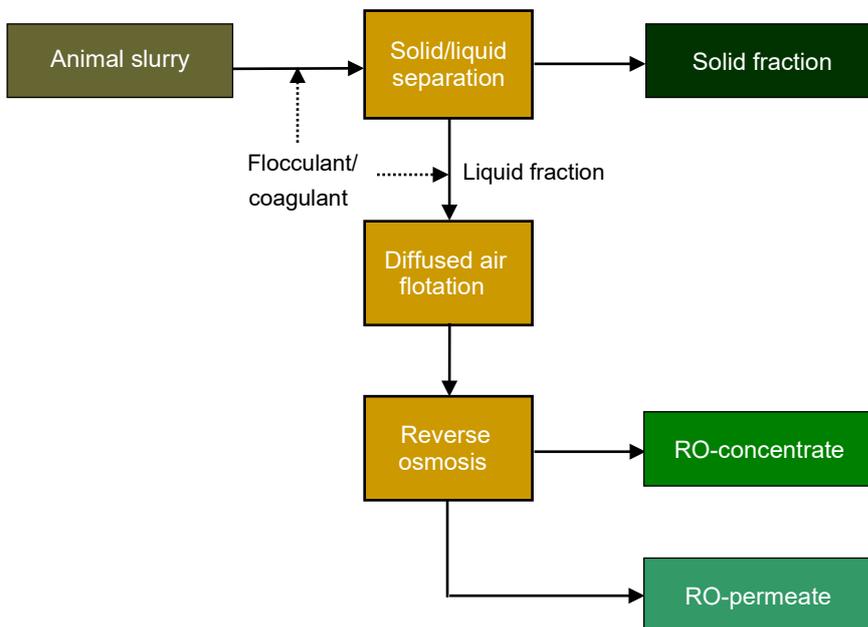


Figure 2.1 Production process of mineral concentrate using reverse osmosis

The principle of reversed osmosis is based on the ability of RO-membranes to let water pass and block salt ions. Water is pressed through the membrane. The pressure needed for the process to take place depends on the conductivity (EC value) of the feed. During the 2- or 3-step process, the conductivity of the concentrate increases by which increasing pressure is needed to get the desired volume of clean permeate. The maximal degree of concentration is limited by the osmotic pressure of the feed and the resulting driving force. The performance of RO-membranes drops if fouling of the membranes occurs. To avoid this, proper pre-treatment of the input is necessary. Flocculants and coagulants are added during pre-treatment to remove suspended solids more effectively. The filtration process is frequently interrupted for cleaning the membranes using acid and lye alternately.

Figure 2.2 shows characteristics of different types of pressure driven membrane technologies according to their pore size. RO-membranes, with a pore size of 0.1 – 1 nm, withhold all soluble salts and let only water pass, which leaves a clean permeate. All substances that are present in the input are held back and end up in the concentrate. However some ions (ammonia) and small particles, such as bacteria and viruses may pass the membrane if the membrane is damaged.

In the pilot installations spiral wound RO-membranes are applied. Figure 2.3 shows the structure of a spiral wound membrane element. RO-units consist of several modules (tubes) each holding 3 or 4 membrane elements. Figure 2.4 shows a picture of RO-unit of one of the pilot installations.

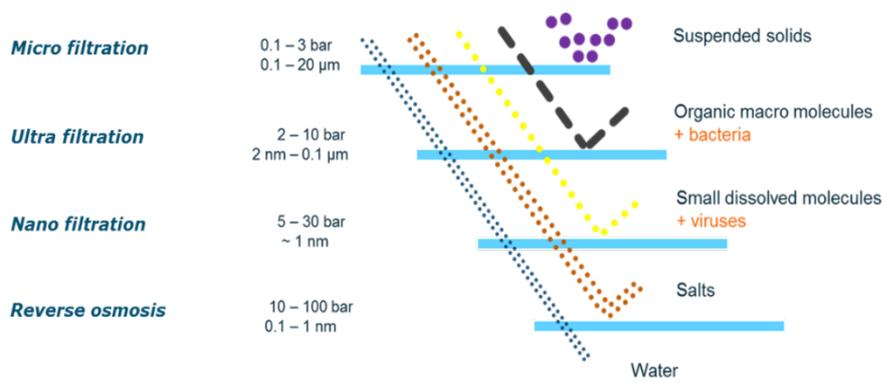


Figure 2.2 Characteristics of different types of membrane filtration (sources: www.watertool.be, Castro-Muñoz et al, 2018)

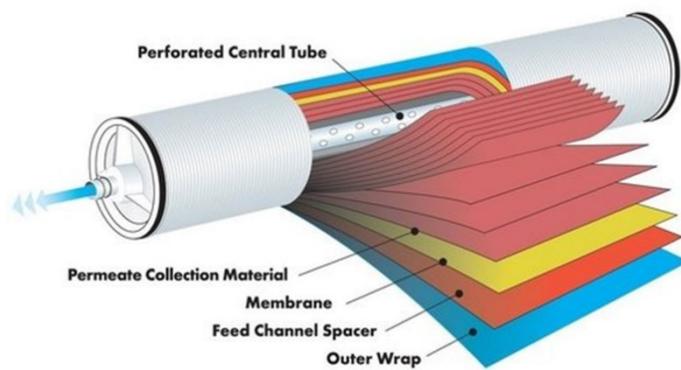


Figure 2.3 Structure of a spiral wound RO-membrane element (source: Schwinge et al, 2004)



Figure 2.4 RO-unit with 9 tubes holding 4 membrane elements each (Photo: WLR).

2.2 Pilot installations

A total of 9 producers of mineral concentrate, operating 17 pilot installations (coded A – Q), participated in the pilot during the monitoring campaign in 2019-2020 covered by this report. The main characteristics of the pilot installations are given in Table 1. Owners of installations K – Q entered into a formal co-operation with the owners of 7 installations that were participating in the pilot from the start, as indicated in Table 1.

All installations processed pig slurry mainly¹. Through several process steps the slurry was processed into a mineral concentrate and water (permeate), applying reversed osmosis as the final technique and in most cases followed by ion exchange on permeate. Pre-treatment of the slurry, to remove solid material and suspended particles from the slurry before feeding the RO-unit, was done by mechanical separation, dissolved air flotation and paper filtration. Additives such as cationic polyacrylamide (PAM) and iron(III)sulphate were commonly used to support the separation process. Installations that use both are indicated as type 1 installations, and installations that use PAM only are indicated as type 2 installations. In some installations sulphuric acid was added to control the pH of the RO-input.

2.3 Monitoring program

The monitoring was carried out in the period September 2019 - April 2020, with approximately two months between samplings leading to three to six sampling rounds per participating installation. The parameters of the monitoring are given in Table 2.

The parameters were divided up in groups, each with a specific purpose:

- General parameters were measured to get a detailed picture of the quality of the produced mineral concentrates as a fertilising product and to mirror the values of the key parameters to the quality criteria of the pilot.
- Heavy metals were measured to generate data that connect to the criteria as are being developed within the context of the new European fertilising products regulation and within the context of the Nitrates Directive (RENURE) next to general parameters on quality.
- Veterinary drugs/antibiotics and pathogenic microorganisms (as precautionary parameters) were measured to procure a basis for designation of a best available technique for effluents from slurry processing plants. These measurements were done to support development of legislation supported by the Dutch Waterboards on discharge to surface water of effluents from slurry processing plants.

From 9 installations (coded A – I) the following process flows were sampled:

- raw animal slurry (input)
- solid fraction
- input of reverse osmosis (RO-input)
- concentrate from reverse osmosis (RO-concentrate)
- permeate from reverse osmosis (RO-permeate)

From 8 installations (coded J – Q) the RO-concentrate was sampled only.

Liquid process flows were sampled using taps in the piping of the installations. A 10 l bucket was filled from which a sample of 1 l was taken. The RO-permeate was sampled after ion exchange, if present. The solid fraction was sampled directly after solid/liquid separation by collecting 1 l material from the transport conveyer belt.

¹ In some cases part of the feed stock dairy cattle slurry but this was limited to < 5% of the total input.

Table 1 Characteristics of pilot installations monitored in 2019 - 2020.

Code	Co-operation		Capacity (ton/a)	Input	Techniques sequentially applied	Additives used
A	A + K	Type 2	230,000	Pig slurry	Air flotation – Belt press - Paper + Candle filtration - Reverse osmosis – Ion exchange	PAM
B	B + P	Type 1	25,000	Sow slurry	Belt press – Sedimentation - Paper filtration – Reverse osmosis	PAM + iron(III)sulphate*
C	C + O	Type 2	16,000	Sow slurry	Air flotation - Screw press - Vibrating screen – Reverse osmosis	PAM
D		Type 1	44,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
E	E + L	Type 1	15,000	Sow slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis	PAM + iron(III)sulphate
F	F + M	Type 2	25,000	Pig slurry	Air flotation – Belt press - Candle filtration – Reverse osmosis	PAM
G		Type 1	85,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
H	H + N	Type 1	80,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
I	I + J	Type 1	25,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
J		Type 1	100,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
K		Type 2	80,000	Pig slurry	Air flotation – Belt press - Paper filtration – Reverse osmosis – Ion exchange	PAM
L		Type 1	100,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
M		Type 2	25,000	Pig slurry	Air flotation – Belt press - Paper filtration – Reverse osmosis – Ion exchange	PAM
N		Type 1	200,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
O		Type 1	40,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate
P		Type 1	35,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis	PAM + iron(III)sulphate (or Al ₃ (SO ₄) ₂)
Q**		Type 1	80,000	Pig slurry	Belt press - Air flotation – Paper filtration – Reverse osmosis – Ion exchange	PAM + iron(III)sulphate

* iron(III)sulphate is solved in sulphuric acid, delivered in a 42% solution

** Co-operation with an installation that was not in operation during the monitoring

Table 2 Parameters of the monitoring program of the pilot installations in 2019 - 2020.

Group	Measured parameters
General quality parameters*	Total solids (TS), Volatile solids (VS) Total organic Carbon (TOC) Total Nitrogen (Total-N) Total Ammonia Nitrogen (TAN) Nitrate (NO ₃ -N) Nitrite (NO ₂ -N) Phosphorus (P) Potassium (K) Ca Mg Na S Cl ⁻ pH Electrical conductivity (EC)
Heavy metals*	Ba, Cd, Cr, Cu, Hg, Ni, Pb, Zn, As
Antibiotics**	Ciprofloxacin Doxycycline Enrofloxacin Florfenicol Flumequine Lincomycin Oxytetracycline Sulfadiazine Sulfadimidine Tilmicosine Trimethoprim
Bacteria**	Escherichia coli Intestinal enterococci Spores of sulphite reducing Clostridia
Viruses***	Somatic coliphages (bacteriophages) Hepatitis E virus

* Measured 3 to 6 times at all installations

** Measured twice at all installations

*** Measured twice at two installations (A and H)

2.4 Laboratory analyses

2.4.1 Chemical parameters

The analytical methods that were used for measuring chemical parameters are given in Table 3.

Table 3 Analytical methods used for chemical parameters.

Parameter	Analytical method
TS	Raw sample 16 h dried at 105 °C (NEN-ISO-11465: 2005)
VS	Dried sample 3 h loss of ignition at 550 °C (NEN-EN-15935:2012)
C	Dried sample measured with LECO TruSpec CHN
Total-N, P	H ₂ SO ₄ -Se digestion; measured with segmented flow analyser at 660 nm (N) and 880 nm (P)
Ca, K, Mg, Na, S	Microwave digestion; measured with ICP-OES
TAN, NO ₂ -N, NO ₃ -N	1 M KCl extraction: 3 g raw material with 30 ml 1 M KCl shaken for 1 hour, filtered 0.45 µm and 10x diluted with water; measured with spectrophotometer
Cl, P-PO ₄ , TOC	Water extraction: 3 g raw material with 30 ml demi water shaken for 1 hour, centrifugated at 3000 rpm, filtered 0.45 µm and 10x diluted with water; measured with spectrophotometer
EC	Fluids directly in sample; solid fraction after extraction 5 g with 25 ml demi water and shaken for 1 hour
pH	Fluids directly in sample; solid fractions after extraction 5 g with 25 ml demi water and shaken for 1 hour

Laboratory: Wageningen Soil Chemistry Laboratory

Heavy metals

Samples were pre-treated using acid digestion in a microwave oven (MARS-6, CEM Corporation, Matthews, NC, USA). For the microwave digestion 10 mL of concentrated nitric acid (67-70% superpure nitric acid, Carlo Erba, Val-de-Reuil, France) was added to 0.5 -2.5 g of sample in Teflon digestion vessels. The amount of sample depended on the estimated moisture content of the samples, with 2.5 g for wet samples (raw slurry, RO-input, RO-concentrate and RO-permeate) and 0.5 g for dry samples (solid fraction). The samples were digested in the microwave oven at a temperature of 210°C. The digests were quantitatively transferred to 50 ml polypropylene (PP) centrifuge tubes (Greiner Bio-One, Frickenhausen, Germany) and diluted with de-ionized water to a final volume of 50 ml. The digests were diluted 5-fold and 200-fold prior to analysis with a final acid concentration of 2.8% v/v. Rhodium and thallium were added to the diluted sample digests as well as to the calibration standards as internal standards at a final concentration of 10 µg/L. Gold chloride was added to both the sample digests and the calibration standards for mobilization of mercury at a final concentration of 1 mg gold/L. Multi-element external calibration standards were prepared in 2.8% nitric acid with concentrations of 10-200 µg/L for copper and zinc, 1-20 µg/L for vanadium, chromium, nickel, arsenic, barium and uranium, 0.25-5 µg/L for cadmium and lead; and 0.1-2 µg/L for mercury.

Concentrations of the elements were determined against the external calibration curves using ICP-MS (NexION 300D or NexION 350D, Perkin Elmer, Waltham, MA, USA) equipped with an autosampler. The ICP-MS was equipped with a PFA-ST nebulizer, a quartz cyclonic spray chamber and a quartz torch with a sapphire injector. Cadmium, mercury and lead were measured without a collision or reaction gas (standard-mode), whereas all other elements were analysed in collision mode (KED) with helium as collision gas at a flow rate of 3.9 mL/min and using an RPq of 0.25. Thallium (m/z 205) was used as the internal standard for mercury and lead, whereas rhodium (m/z 103) was used as internal standard for all other elements. The isotopes used for quantification were ⁵¹V (vanadium) ⁵²Cr (chromium), ⁶⁰Ni (nickel), ⁶⁵Cu (copper), ⁶⁶Zn (zinc), ⁷⁵As (arsenic), ¹¹⁴Cd (cadmium), ¹³⁸Ba (barium), ²⁰¹Hg (mercury) and ²³⁸U (uranium). Lead was quantified using the sum of the three isotopes ²⁰⁶Pb, ²⁰⁷Pb and ²⁰⁸Pb.

Certified reference materials BCR 482 lichen and SETOC 733 sediment were used for quality control. Because no certified reference values are available for the elements vanadium, barium and uranium, the recovery of these elements was determined by spiking BCR 482 lichen at a level of 20 mg/kg.

Laboratory: Wageningen Food Safety Research

Antibiotics

The confirmation of the identity of antibiotics was carried out in accordance to the Commission Decision EC 2002/657/EC.

All compounds were analyzed simultaneously. The target limits in slurry for sulfadimidine, sulfadiazine and trimethoprim were 5 µg/kg and for the other compounds were set at 20 µg/kg.

Raw samples and pretreated test samples were stored at -18 °C.

The samples were extracted with Mcllvain buffer and acetonitrile after which the proteins were precipitated with lead acetate. After centrifuging and pouring out the extract, the lead was removed with EDTA. The extracts were transferred through a Reversed Phase SPE column and the components were eluted with a methanol/water mixture. After evaporation, dissolving and if necessary centrifugation, the extracts were measured using LC-MS/MS.

LC-MS system:

-Waters Acquity LC

-Analytical column: Kinetex 1.7 µ C18 100A, 100*2.1 mm

-Mass spectrometer: AB Sciex QTrap6500 provided with a ESI-interface.

All reagents were of pre-analysis quality or a higher quality. Water cleaned over a MilliQ® installation with a minimum resistance of 18.2 MΩ.cm⁻¹ was used.

The following checks were performed for each analytical series (10 – 20 samples):

- A series of a blank sample (preferably matrix corresponding to the samples) with various additions, based on which identification and quantification of the samples take place (MMS);
- All samples were treated in duplicate. The second sample contained an addition at target level, or when sensitivity was in question two times target level.

Defrosted samples were homogenized by stirring with a disposable wooden spatula. The test unit was 2.00 ± 0.05 grams.

All samples were analyzed in duplicate. One of the duplicates was spiked at target level or twice that level. Two grams of sample was weighed into a 50 ml tube. Internal standard solution was added to all samples and the standard solution to the duplicate samples and mixed. The samples rested for 20 minutes. Mcllvain-buffer was added to all tubes. After vigorously shaking acetonitrile containing TFA was added. All samples were placed in a Head over Head mixing device for 15 minutes. Lead acetate solution was added to all tubes, shaken vigorously and then centrifuged at high level. The supernatant was poured into a glass tube.

The acetonitrile was removed by nitrogen-current at 40° C. EDTA-solution was added and the complete sample was transferred on a conditioned SPE-column. The column was washed and eluted. The samples were evaporated by using nitrogen-flow at 40° C. The residue was dissolved in water/methanol mixture. The samples were filtered when necessary and placed in vials.

Before the start of the analyses by LC-MS/MS the sensitivity and retention time of components was checked using a solvent standard and the lower MMS by means of signal-to-noise.

When high concentrations were expected extra solvent measurements were added.

Data processing:

Four calculation sheets were used for data processing (one for each group of antibiotics). The limit of detection (LOD) was determined individually for each sample based on the response of the internal standard in combination with the signal-to-noise of the standard line (MMS). The concentration found in a sample was compared with this LOD and reported as positive (with a concentration) or negative (<LOD).

Laboratory: Wageningen Food Safety Research

2.4.1 Biological parameters

Sample transport

All samples were directly transported to a cold storage (4°C) and analysed within 24 hours after sampling (maximally within 29 hours after sampling).

Pre-treatment of "liquid" fractions for bacterial and viral determinations

For the determination of *E. coli*, enterococci and spores of sulphite reducing Clostridia in "liquid" fractions (= animal slurry, liquid fraction/RO-input, RO-concentrate), the following pre-treatment was used: 50 grams of the liquid fractions were diluted 1:1 in peptone saline (PS) and homogenized using a pulsifier (Microgen Bioproducts Ltd). Decimal dilutions of the homogenized suspensions were prepared in PS (10⁻¹ to 10⁻⁴).

Pre-treatment of solid fraction for bacterial and viral determinations

For the determination of *E. coli*, enterococci and spores of sulphite reducing Clostridia in the solid fraction, the following pre-treatment was used: 15 grams of the dry fractions were diluted 1:1 in PS. The 1:1 dilution was diluted 1:5 in PS and homogenized using a pulsifier (Microgen Bioproducts Ltd). Decimal dilutions of the homogenized suspensions were prepared in PS (10⁻² to 10⁻⁴).

Pre-treatment of RO-permeate for bacterial determinations

500 ml of the RO-permeate was filtered through a 0.45 µm membrane filter twice. The filters were then placed on solid culture media.

Pretreatment of RO-permeate for viral determination

Approximately 20 l of RO-permeate were collected and concentrated by a conventional filter adsorption-elution method. Magnesium chloride (4.17 mol/L, Sartorius) was added to the water sample to a final concentration of 0.05 M. The pH was reduced to 3.8 by adding 1M HCl. The viruses and bacteriophages were eluted after filtration through a 1.2 µm negatively charged cartridge filter (4 inch cellulose acetate, Milligard cartridge filter, Millipore) with approximately 600 ml 3% beef extract solution (pH > 9.0) (Gibco). The eluate was neutralized with 55 ml acetic acid buffer (pH 5.0) and further concentrated by ultrafiltration using a cellulose acetate filter ((Cellulose triacetate, MWCO 10 kD, ø100 mm Sartorius) under high pressure (3 bars). The ultrafilter was rinsed with 3% beef extract solution (pH > 0.9) to a volume ranging from 10 to 30 grams. The remaining concentrate was analysed for numbers of somatic coliphages and hepatitis E viruses.

E. coli

E. coli was enumerated according to ISO 9308-1 (as regards membrane filtration of permeate) and ISO 16649-2 (as to the media and incubation conditions). For slurry fractions, 100 µl of the 1:1 to 10⁻⁴ dilutions (liquid fractions) or of the 10⁻¹ to 10⁻⁴ dilutions (dry fractions) were enumerated by spread plating on 'Tryptone bile X-glucuronide agar (TBX) (0.06111 and 0.01111 grams of slurry in total, respectively). The filters with filtered RO-permeate (2x500ml) were also placed on TBX agar. All cultures were incubated at 37 °C for 4-5 hours, followed by 44-22 hours at 18 °C. The green-blue colonies were counted.

Intestinal enterococci

Intestinal enterococci were enumerated according to ISO 7899-2. Membrane filtration was performed for permeate samples; all other fractions were enumerated by spread plating on the same media. For slurry fractions, 100 µl of the 1:1 to 10⁻³ dilutions (liquid fractions) or of the 10⁻¹ to 10⁻³ dilutions (dry fractions) were enumerated by spread plating on 'Slanetz and Bartley agar' (S&B) (a total of 0.0611 and 0.0111 grams of slurry, respectively). The filters with filtered RO-permeate (2x100ml) were also placed on S&B agar. All cultures were incubated at 37 °C for 4-5 hours, followed by 40-44 hours at 44 °C according to the media specifications (Oxoid). After incubation, maroon colonies were counted. For confirmation, 10 colonies per sample were sub-cultured onto Bile Esculin agar (BEA) followed by incubation at 44 °C for 18-24 hours. Counts on S&B were corrected by multiplying the fraction of colonies confirmed as intestinal enterococci (black colonies with black halo) with the number of colonies counted on S&B.

Spores of sulphite reducing Clostridia (SSRC)

The determination of spores of sulphite reducing Clostridia was based on ISO 6461-2. The membrane filtration method described in this standard was performed only for the RO-permeates whereas the cultivation media were used for all fractions. For the determinations in other fractions, 100 µl of the 1:1 to 10⁻⁴ dilutions (liquid fractions) or of the 10⁻¹ to 10⁻⁴ dilutions (solid fractions) were spread plated on 'Iron Sulphite agar' (ISA) (in total 0.06111 and 0.01111 grams of slurry respectively). After filtrating 2x500 ml of RO-permeate, filters were also placed on ISA agar. All cultures were incubated under anaerobic conditions at 37 °C for 18-24 hours and 40-48 hours, and the black colonies were counted.

Somatic coliphages

The number of somatic coliphages were determined according to ISO 10705-2. 10 gram of the slurry fractions were diluted 1:10 in PS and homogenized. A dilution series of the homogenized suspensions was made in PS (10⁻² to 10⁻⁵). For the enumeration of somatic coliphages, a mixture of 1 ml of the nalidixic acid resistant mutant Escherichia coli CN (WG5), 1 ml of the sample or the ten-fold dilutions of the sample and 2.5 ml of molten culture medium (Scholtens' agar, Oxoid) poured into a petri dish over a solid culture medium (Scholtens' agar, Oxoid). All plates were incubated at 37 °C for 16-20 hours, followed by plaque counting.

Hepatitis E virus

A 10% suspension was made from the samples by adding 900µl Hanks' Balanced Salt Solution (HBSS, calcium, magnesium (Gibco, ThermoFischer scientific) with 0.05 mg/ml gentamycin to 100 mg of the slurry fractions and vortexing for 30 seconds. After centrifugation at 3000xg for 15 minutes, the supernatant was used for analysis.

For the RNA isolation from the slurry suspensions or the concentrate, a Fast Spin column kit with silica membrane (QIAmp Viral RNA Mini Kit, Qiagen) was used. Presence of HEV was tested in undiluted and 10x diluted RNA extracts by real-time RT-PCR. Primers and probes as described by Bouwknegt et al. (2009) were used. The TaqMan Fast Virus 1-step kit (Invitrogen) was used for RT-PCR.

Laboratory: National Institute for Public Health and Environment

2.5 Data processing

The average value and standard deviation of the parameters measured in the process flows overall and of individual pilot installations were determined.

Regression and variance analysis were executed with Genstat 19nd edition or R (for biological parameters only). Statistical comparisons of chemical parameters were performed with ANCOVA, of biological parameters with ANOVA followed by a Tukey post-hoc test. Significance of differences was tested with p<0.05. The impact of the composition of the input on the composition of RO-concentrate was tested with ANCOVA. Correlations between parameters in the RO-concentrate were conducted using R package 'ggcorrplot'.

The reported real average concentrations of heavy metals and antibiotics in process flows of individual installations were calculated as sum of measured values divided by the number of measurements; reported values < detection limit represent the average detection limit.

3 Results

The presentation of results in this chapter is primarily focused on the qualitative characteristics of RO-concentrate and the impact of the input on the composition of RO-concentrate. The results will be discussed considering the quality criteria set for the pilot MC and the proposed criteria for RENURE. The relationships between RO-concentrates and the other process flows will be discussed.

The average values of the concentrations, in wet product and in dry matter, and standard deviations of the measured chemical parameters in the process flows of the individual installations are given in Appendices 1-4.

3.1 Composition of input

3.1.1 General parameters

Table 4 gives the average values of the general parameters in the input (raw slurry) of pilot installations A-I. The table shows significant differences between installations, especially between sow slurry (B, C and E) and slurry from fattening pigs. Sow slurry shows lower concentrations of almost all general parameters. Noteworthy is the detection of NO₂-N+NO₃-N in the input of installations C to H as in stored slurry an anoxic environment may be assumed. Stirring and pumping the slurry into a buffer tank may have caused the supply of some oxygen. The input pig slurry of the pilot installations originates from a great number (> 200) of pig farms. Based on the average concentrations of the general parameters the input may be considered as representative for the pig slurry produced at Dutch pig farms today (Commissie Bemesting Grasland en Voedergewassen, 2020).

3.1.2 Heavy metals

Table 5 gives the average values of heavy metals in the input of pilot installations A-I. There are no significant differences in heavy metal concentrations between the inputs of the different installations. C and E show lower concentrations than the other installations. Relatively high concentrations of Zn, Cu and Ba were found in all slurry samples. Ni and Pb were found in all input samples. Concentrations of the other heavy metals were lower than 1 mg/kg or below detection limit.

On average concentrations of Cd, Cr, Cu, Ni and Zn were comparable to the concentrations in the input pig slurry of 9 pilot installations as reported by Hoeksma et al. (2011) from the monitoring in 2009-2010. The other metals from Table 5 were not included in that monitoring program.

3.1.3 Antibiotics

Table 6 gives the average values of antibiotics in the input of pilot installations A-I. Oxytetracycline and doxycycline were found in the input of all installations in relatively high concentrations. Sulfadiazine was found in 6 out of 9 inputs. Overall, there is no significant difference between the input of the different pilot installations regarding antibiotics.

Considering the amounts of antibiotics that are used in pig husbandry, the part of the administered amounts that is excreted with urine and feces and the persistence of the substances during slurry storage, the detected antibiotics in the input of the pilot installations could be expected. Lahr et al. (2014) found in stored pig slurry of 4 Dutch pig farms the following concentrations: oxytetracycline 95-883 µg/kg, doxycycline 2454-4787 µg/kg, sulfadiazine <1-225 µg/kg and flumequine 40 µg/kg (one measurement).

3.1.4 Biological parameters

Table 7 shows the average values of the count of measured microorganisms in the input of pilot installations A-I. *E. coli*, spores of sulphite reducing Clostridia and intestinal enterococci were found in the input of all pilot installations, as expected. On average, concentrations of *E. coli* and enterococci were around 10^4 CFU/g and slightly higher for SSRC. The concentrations of these bacteria varied between and partly also within the installations, with *E. coli* showing the greatest differences between installations (up to 100 fold). Differences between installations were however not statistically significant, based on two measurements per installation. Hoeksma et al. (2015) found comparable average concentrations of *E.coli* and intestinal enterococci in the input of 6 pilot installations. More recently, these concentration levels of *E.coli* and enterococci spp. in (Belgian) pig slurry were confirmed by Rasschaert et al. (2020).

Table 4 Average concentrations of **general parameters** in the input of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is significant difference in concentration between installations for the designated parameter ($p < 0.05$). The number between brackets represents the number of samples.

Parameter	Unit	A		B		C		D		E		F		G		H		I	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
TS	g/kg	86 ^c	(5)	30 ^a	(3)	36 ^{ab}	(3)	58 ^{abc}	(3)	37 ^a	(4)	85 ^c	(3)	76 ^c	(5)	68 ^{bc}	(6)	55 ^{abc}	(3)
VS	g/kg DM	706 ^c	(5)	634 ^c	(3)	470 ^a	(3)	664 ^c	(3)	554 ^{ab}	(4)	703 ^c	(3)	712 ^c	(5)	695 ^c	(6)	676 ^c	(3)
C-total	g/kg DM	364 ^c	(5)	341 ^{bc}	(3)	252 ^a	(3)	334 ^{bc}	(3)	309 ^b	(3)	352 ^{bc}	(3)	367 ^c	(5)	359 ^c	(6)	344 ^{bc}	(3)
TOC	mg/kg	3498 ^c	(5)	1296 ^b	(3)	1812 ^{ab}	(3)	2049 ^{ab}	(3)	941 ^a	(4)	3986 ^c	(3)	2648 ^{bc}	(5)	2437 ^{ab}	(6)	2631 ^{bc}	(3)
Total-N	mg/kg	6460 ^d	(5)	2829 ^a	(3)	3831 ^{ab}	(3)	4958 ^{bcd}	(3)	2799 ^a	(4)	5800 ^{bcd}	(3)	5510 ^{cd}	(5)	5033 ^{abc}	(6)	5191 ^{bcd}	(3)
TAN	mg/kg	3715 ^e	(5)	1652 ^{ab}	(3)	2383 ^{bc}	(3)	2790 ^{cd}	(3)	1506 ^a	(4)	3307 ^{de}	(3)	3050 ^{cd}	(5)	2822 ^{cd}	(6)	3198 ^{de}	(3)
P	mg/kg	1727 ^c	(5)	742 ^a	(3)	1080 ^{abc}	(3)	1314 ^{abc}	(3)	972 ^{ab}	(4)	1539 ^{abc}	(3)	1678 ^{bc}	(5)	1432 ^{abc}	(6)	1269 ^{abc}	(3)
P-PO ₄	mg/kg	241 ^b	(5)	201 ^{ab}	(3)	158 ^{ab}	(3)	150 ^{ab}	(3)	129 ^a	(4)	248 ^b	(3)	232 ^b	(5)	234 ^{ab}	(6)	203 ^{ab}	(3)
K	mg/kg	4680 ^d	(5)	1633 ^a	(3)	3067 ^{abc}	(3)	4033 ^{cd}	(3)	2350 ^{ab}	(4)	4367 ^{cd}	(3)	3900 ^{cd}	(5)	3920 ^{bc}	(6)	3633 ^{bcd}	(3)
EC	mS/cm	29 ^c	(5)	17 ^a	(3)	24 ^b	(3)	25 ^{bc}	(3)	16 ^a	(4)	26 ^{bc}	(3)	25 ^b	(5)	24 ^b	(6)	27 ^{bc}	(3)
pH		7.8 ^{bc}	(5)	7.5 ^a	(3)	8.2 ^d	(3)	7.8 ^{bc}	(3)	7.7 ^{ab}	(4)	7.9 ^c	(3)	7.7 ^{bc}	(5)	7.7 ^b	(6)	7.8 ^{bc}	(3)
NO ₂ -N + NO ₃ -N	mg/kg	0 ^a	(5)	0 ^a	(3)	0.37 ^a	(3)	0.36 ^a	(3)	0.55 ^a	(4)	0.42 ^a	(3)	0.24 ^a	(5)	0.43 ^a	(6)	0 ^a	(3)
Ca	mg/kg	2060 ^a	(5)	1067 ^a	(3)	1377 ^a	(3)	1427 ^a	(3)	2408 ^a	(4)	2240 ^a	(3)	2202 ^a	(5)	2020 ^a	(6)	1643 ^a	(3)
Mg	mg/kg	1286 ^c	(5)	507 ^a	(3)	707 ^{ab}	(3)	837 ^{abc}	(3)	548 ^a	(4)	1280 ^{bc}	(3)	1148 ^{bc}	(5)	1050 ^{abc}	(6)	877 ^{abc}	(3)
Na	mg/kg	1148 ^b	(5)	668 ^a	(3)	975 ^b	(3)	995 ^b	(3)	572 ^a	(4)	1156 ^b	(3)	1036 ^b	(5)	1096 ^b	(6)	1107 ^b	(3)
S	mg/kg	972 ^d	(5)	253 ^a	(3)	413 ^{ab}	(3)	1433 ^e	(3)	398 ^{ab}	(4)	807 ^{cd}	(3)	1040 ^d	(5)	764 ^{bc}	(6)	663 ^{bc}	(3)
Cl-	mg/kg	2022 ^d	(5)	1256 ^{bc}	(3)	1147 ^{ab}	(3)	1591 ^{bcd}	(3)	603 ^a	(4)	2172 ^d	(3)	1698 ^{bcd}	(5)	1645 ^{cd}	(6)	1861 ^{cd}	(3)

Table 5 Average concentrations of **heavy metals** (mg/kg) in the input of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is significant difference in concentration between installations for the designated parameter ($p < 0.05$). For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of samples.

Parameter	A		B		C		D		E		F		G		H		I	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
As	<0.11	(0/5)	<0.11	(0/3)	0.04	(1/3)	<0.11	(0/2)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/6)	<0.11	(0/2)
Ba	4.0 ^c	(5/5)	1.4 ^a	(3/3)	4.2 ^c	(3/3)	2.9 ^{bc}	(2/2)	1.4 ^a	(3/3)	3.0 ^{bc}	(3/3)	2.8 ^{bc}	(3/3)	3.3 ^c	(6/6)	2.2 ^{ab}	(2/2)
Cd	0.04 ^a	(5/5)	<0.03	(0/3)	0.03 ^a	(3/3)	0.04 ^a	(1/2)	<0.02	(0/3)	0.04 ^a	(3/3)	0.04 ^a	(3/3)	0.04 ^a	(6/6)	0.02 ^a	(2/2)
Cr	0.77 ^{bc}	(5/5)	0.41 ^{ab}	(3/3)	0.60 ^{abc}	(3/3)	0.46 ^{abc}	(2/2)	0.20 ^a	(1/3)	0.90 ^c	(3/3)	0.72 ^{bc}	(3/3)	0.80 ^c	(6/6)	0.50 ^{abc}	(2/2)
Cu	23 ^{abc}	(5/5)	10 ^{ab}	(3/3)	31 ^{bc}	(3/3)	24 ^{abc}	(2/2)	5.3 ^a	(3/3)	38 ^c	(3/3)	18 ^{abc}	(3/3)	19 ^{abc}	(6/6)	25 ^{abc}	(2/2)
Hg	<0.01	(0/5)	<0.01	(0/3)	<0.01	(0/3)	<0.01	(0/2)	<0.01	(0/3)	<0.01	(0/3)	<0.01	(0/3)	<0.01	(0/6)	<0.01	(0/2)
Ni	0.82 ^{bc}	(5/5)	0.32 ^a	(3/3)	0.90 ^{bc}	(3/3)	0.72 ^{bc}	(2/2)	0.29 ^a	(3/3)	1.0 ^c	(3/3)	0.76 ^{bc}	(3/3)	0.68 ^b	(6/6)	0.56 ^{ab}	(2/2)
Pb	0.22 ^{ab}	(5/5)	0.06 ^a	(3/3)	0.10 ^{ab}	(3/3)	0.10 ^{ab}	(2/2)	0.04 ^a	(3/3)	0.70 ^b	(3/3)	0.08 ^{ab}	(3/3)	0.09 ^a	(6/6)	0.08 ^{ab}	(2/2)
Zn	81 ^c	(5/5)	34 ^{ab}	(3/3)	92 ^c	(3/3)	75 ^{bc}	(2/2)	25 ^a	(3/3)	76 ^{bc}	(3/3)	63 ^{abc}	(3/3)	66 ^{bc}	(6/6)	62 ^{abc}	(2/2)

Table 6 Average concentrations of **antibiotics** ($\mu\text{g}/\text{kg}$) in the input of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is significant difference in concentration between installations for the designated parameter ($p < 0.05$). Values above detection limit are presented in bold. For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of values > detection limit and the total number of measurements.

Parameter	A		B		C		D		E		F		G		H		I	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
Oxytetracycline	34 ^a	(3/3)	248 ^a	(2/2)	70 ^a	(2/2)	58 ^a	(1/2)	3.5 ^a	(1/2)	47 ^a	(1/2)	62 ^a	(2/2)	245 ^a	(5/5)	40 ^a	(2/2)
Doxycycline	272 ^a	(3/3)	351 ^a	(2/2)	670 ^a	(2/2)	89 ^a	(1/2)	12 ^a	(2/2)	71 ^a	(1/2)	467 ^a	(2/2)	366 ^a	(5/5)	292 ^a	(2/2)
Sulfadiazine	14 ^a	(2/3)	<1.0	(0/2)	3.5 ^a	(2/2)	1.0 ^a	(0/2)	2.5 ^a	(1/2)	15 ^a	(0/2)	1.0 ^a	(2/2)	3.8 ^a	(2/5)	1.0 ^a	(2/2)
Sulfadimidine	<1.0	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/5)	<1.0	(0/2)
Trimethoprim	<1.7	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<1.2	(0/5)	<1.5	(0/2)
Ciprofloxacin	<5.7	(0/3)	<3.0	(0/2)	<3.0	(0/2)	<5.5	(0/2)	<1.5	(0/2)	<4.0	(0/2)	<1.5	(0/2)	<5.2	(0/5)	<2.0	(0/2)
Enrofloxacin	<4.7	(0/3)	<1.0	(0/2)	<2.0	(0/2)	<1.5	(0/2)	1.0 ^b	(1/2)	<3.5	(0/2)	<3.5	(0/2)	<6.8	(0/5)	<3.0	(0/2)
Flumequine	0.67 ^a	(1/3)	<1.0	(0/2)	0.50 ^a	(1/2)	<1.0	(0/2)	<1.0	(0/2)	0.50 ^a	(1/2)	<1.5	(0/2)	0.20 ^a	(1/5)	<1.5	(0/2)
Lincomycine	<1.7	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<1.4	(0/5)	<1.5	(0/2)
Tilmicosine	2.0 ^a	(1/3)	5.0 ^a	(2/2)	<10	(0/2)	<5.5	(0/2)	<5.0	(0/2)	<6.0	(0/2)	<7.5	(0/2)	<1.3	(0/5)	2.5 ^a	(1/2)
Florfenicol	<2.3	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<3.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.8	(0/5)	<1.0	(0/2)

Table 7 Average concentrations of **bacteria and viruses** in the input of pilot installations A – R, based on the geometric mean. Values are shown in two units: the decadic logarithm of the concentration, and the concentration. As example: +4 $\log_{10}(\text{CFU/g})$ equals 10^4 CFU/g, and 0 $\log_{10}(\text{CFU/g})$ equals 10^0 or 1 CFU/g. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter, calculated for the geometric mean ($p < 0.05$). For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of samples. NA = not available (not measured)

Parameter	Unit	A		B		C		D		E		F		G		H		I	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
<i>E. coli</i>	$\log_{10}(\text{CFU/g})$	2.2 ^a	(1/2)	4.5 ^a	(2/2)	2.6 ^a	(2/2)	3.2 ^a	(2/2)	3.9 ^a	(2/2)	4.3 ^a	(2/2)	4.4 ^a	(2/2)	4.7 ^a	(2/2)	3.7 ^a	(2/2)
Enterococci	$\log_{10}(\text{CFU/g})$	3.6 ^a	(2/2)	3.1 ^a	(2/2)	3.5 ^a	(2/2)	1.6 ^a	(1/2)	<1.2	(0/2)	4.1 ^a	(2/2)	3.2 ^a	(2/2)	3.0 ^a	(2/2)	3.2 ^a	(2/2)
Clostridia	$\log_{10}(\text{CFU/g})$	3.8 ^{bc}	(2/2)	3.5 ^{ab}	(1/2)	4.6 ^b	(2/2)	1.7 ^a	(1/2)	3.0 ^{ab}	(2/2)	3.6 ^{ab}	(2/2)	3.1 ^{ab}	(2/2)	2.4 ^{ac}	(2/2)	3.1 ^{ab}	(2/2)
Bacteriophages	$\log_{10}(\text{pfu/g})$	4.9 ^a	(2/2)	NA		NA		NA		NA		NA		NA		5.0 ^a	(2/2)	NA	
Hepatitis E virus	positive		(2/2)	NA		NA		NA		NA		NA		NA		positive	(2/2)	NA	

3.2 Composition of RO-concentrate

3.2.1 General parameters

Table 8a and 8b gives the average values of the general parameters in the RO-concentrate of all individual pilot installations. The RO-concentrates of installations B, C, and E are characterized by lower concentrations of NH₄-N and K compared with the other installations. These installations process sow slurries. The RO-concentrates of installations A, C, F, K and M show high concentration of P-total compared with the rest of the installations. These installation do not use iron salts while others do. Differences in the composition of RO-concentrates between process type 1 and process type 2 are further elaborated in section 3.6.

3.2.2 Heavy metals

Table 9 gives the average values of the measured heavy metals in the RO-concentrate of all pilot installations. The following heavy metals were found in concentrations above detection limit in a various number of RO-concentrates: Ba (5), Cr (7), Cu (5), Ni (all 17), and Zn (7). Other metals were not detected. The concentrations of most of the detected metals were <1 mg/kg. In the concentrates of A, C, F, L and K the concentrations of Cu and/or Zn were >1 mg/kg. The monitoring in 2009-2010 showed comparable results (Hoeksma et al., 2011).

3.2.3 Antibiotics

Table 10 gives the average values of the measured antibiotics in the RO-concentrate of pilot installations A-I. Oxytetracycline, doxycycline and sulfadiazine were most often detected. Highest concentrations were measured for doxycycline, as were in the input. Flumequine, lincomycine and tilimicosine were occasionally found. In general, the concentrations of antibiotics in the RO-concentrate were substantially lower (factor 7-100) than in the input.

3.2.4 Biological parameters

Table 11 shows the average values of the measured microorganisms in the RO-concentrate of pilot installations A-I. In general, the measured microorganisms were detected in nearly all RO-concentrates, with the exception of intestinal enterococci for which concentrations were below the limit of detection in installation E. Concentrations varied between installations; for *E. coli* and enterococci, up to > 300 fold differing concentrations could be found between installations. In addition, smaller variation was occasionally observed within the installations. As expected, concentrations in RO-concentrate were highly correlated to concentrations in RO-input (Figure 3.1). Comparable concentrations of *E. coli* and instestinal enterococci were observed in the RO-concentrates of 6 pilot installations by Hoeksma et al. (2015), which means that in the last 5 years there was no change in concentration of pathogens in the RO-concentrates of these installations.

Table 8a Average concentrations of **general parameters** in the RO-concentrates of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

Parameter	Unit	A		B		C		D		E		F		G		H		I	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
TS	g/kg	29 ^{bc}	(5)	22 ^{ab}	(3)	22 ^{ab}	(3)	32 ^{bcd}	(3)	17 ^a	(4)	35 ^{bcde}	(3)	44 ^{ef}	(5)	54 ^f	(6)	47 ^{ef}	(3)
VS	g/kg DM	422 ^f	(5)	301 ^{cde}	(3)	357 ^{ef}	(3)	243 ^{bcd}	(3)	131 ^a	(4)	373 ^{ef}	(3)	354 ^{ef}	(5)	354 ^{ef}	(6)	291 ^{cde}	(3)
C-total	g/kg DM	255 ^g	(5)	132 ^d	(3)	214 ^f	(3)	56 ^a	(3)	80 ^{ab}	(3)	221 ^f	(3)	77 ^a	(5)	62 ^a	(6)	66 ^a	(3)
TOC	mg/kg	5038 ^e	(5)	3258 ^{bcd}	(3)	2856 ^{bcd}	(3)	1819 ^{ab}	(3)	1014 ^a	(4)	5481 ^e	(3)	3469 ^{cd}	(5)	4038 ^{de}	(6)	3367 ^{bcd}	(3)
Total-N	mg/kg	7145 ^{ef}	(5)	5472 ^{bcd}	(3)	4492 ^{ab}	(3)	5276 ^{bc}	(3)	3078 ^a	(4)	8328 ^f	(3)	6778 ^{def}	(5)	7989 ^f	(6)	6428 ^{cde}	(3)
TAN	mg/kg	5599 ^{cde}	(5)	4536 ^{bc}	(3)	3448 ^{ab}	(3)	4574 ^{bc}	(3)	2659 ^a	(4)	6485 ^{efg}	(3)	6075 ^{defg}	(5)	6944 ^{fg}	(6)	5898 ^{cdef}	(3)
P	mg/kg	169 ^d	(5)	39 ^{ab}	(3)	159 ^d	(3)	22 ^a	(3)	32 ^{ab}	(4)	244 ^e	(3)	27 ^a	(5)	25 ^a	(6)	27 ^{ab}	(3)
P-PO ₄	mg/kg	110 ^d	(5)	8.7 ^{ab}	(3)	110 ^d	(3)	1.2 ^a	(3)	20 ^{ab}	(4)	174 ^e	(3)	5.6 ^a	(5)	6.8 ^a	(6)	13 ^{ab}	(3)
K	mg/kg	6500 ^{bcd}	(5)	4333 ^a	(3)	4767 ^{ab}	(3)	6300 ^{abcd}	(3)	4325 ^a	(4)	8500 ^{defghi}	(3)	7580 ^{defg}	(5)	9417 ^{fhi}	(6)	7400 ^{cdef}	(3)
EC	mS/cm	50 ^{cd}	(5)	51 ^{cde}	(3)	38 ^{ab}	(3)	52 ^{cde}	(3)	33 ^a	(4)	60 ^{cdefg}	(3)	62 ^{efgh}	(5)	72 ^h	(6)	62 ^{cefgh}	(3)
pH		8.4 ^e	(5)	7.8 ^{ab}	(3)	8.2 ^{de}	(3)	7.8 ^{abc}	(3)	7.8 ^{abc}	(4)	8.1 ^{cd}	(3)	7.7 ^a	(5)	7.7 ^a	(6)	7.7 ^a	(3)
NO ₂ -N + NO ₃ -N	mg/kg	0.20 ^a	(5)	3.3 ^a	(3)	0.37 ^a	(3)	0 ^a	(3)	0.81 ^a	(4)	0.37 ^a	(3)	0.19 ^a	(5)	2.8 ^a	(6)	0.70 ^a	(3)
Ca	mg/kg	128 ^{ab}	(5)	213 ^{abcd}	(3)	150 ^{abcd}	(3)	170 ^{abcd}	(3)	165 ^{abcd}	(4)	140 ^{abc}	(3)	314 ^{def}	(5)	430 ^e	(6)	303 ^{cde}	(3)
Mg	mg/kg	30 ^a	(5)	337 ^{cdef}	(3)	27 ^a	(3)	393 ^{efg}	(3)	168 ^{abc}	(4)	20 ^a	(3)	432 ^{efg}	(5)	557 ^g	(6)	270 ^{bcde}	(3)
Na	mg/kg	1690 ^{abc}	(5)	1860 ^{abcd}	(3)	1568 ^a	(3)	1692 ^{ab}	(3)	1381 ^a	(4)	2069 ^{abcdef}	(3)	2071 ^{abcdefg}	(5)	2681 ^{efgh}	(6)	4087 ⁱ	(3)
S	mg/kg	482 ^a	(5)	107 ^a	(3)	267 ^a	(3)	5180 ^{def}	(3)	1818 ^{ab}	(4)	1027 ^{ab}	(3)	6596 ^{ef}	(5)	8428 ^g	(6)	7387 ^{fg}	(3)
Cl-	mg/kg	3097 ^{cd}	(5)	6943 ⁱ	(3)	2020 ^{ab}	(3)	2796 ^{bc}	(3)	1799 ^a	(4)	3854 ^{defgh}	(3)	3555 ^{cdefg}	(5)	4328 ^{fh}	(6)	3696 ^{cdefgh}	(3)

Table 8b Average concentrations of **general parameters** in the RO-concentrates of pilot installations J –Q. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

Parameter	Unit	J		K		L		M		N		O		P		Q	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
TS	g/kg	31	bcd (3)	28	abc (4)	39	cde (4)	27	ab (4)	42	de (4)	24	ab (3)	33	bcd (4)	42	de (4)
VS	g/kg DM	217	bc (3)	366	ef (4)	190	ab (4)	309	de (4)	259	bcd (4)	242	bcd (3)	217	bc (4)	324	de (4)
C-total	g/kg DM	120	cd (3)	227	f (4)	78	ab (4)	181	e (4)	101	bc (4)	138	d (3)	56	a (4)	79	ab (4)
TOC	mg/kg	2950	bcd (3)	4309	de (4)	2922	bcd (4)	2871	bcd (4)	4073	de (4)	2123	abc (3)	2025	abc (4)	4260	de (4)
Total-N	mg/kg	5997	bcd (3)	6214	cde (4)	6382	cde (4)	5932	bcd (4)	8177	f (4)	5837	bcd (3)	5544	bcd (4)	7202	ef (4)
TAN	mg/kg	5330	cde (3)	4891	c (4)	5758	cdef (4)	4888	c (4)	7272	g (4)	5004	cd (3)	4848	c (4)	6603	efg (4)
P	mg/kg	40	ab (3)	184	d (4)	34	ab (4)	105	c (4)	56	b (4)	59	b (3)	24	a (4)	17	a (4)
P-PO ₄	mg/kg	9.3	ab (3)	118	d (4)	14	ab (4)	55	c (4)	14	ab (4)	28	b (3)	9.4	ab (4)	1.7	a (4)
K	mg/kg	8000	defghi (3)	6575	bcd (4)	9700	i (4)	7300	cde (4)	9000	efghi (4)	5467	abc (3)	7675	defgh (4)	6850	cd (4)
EC	mS/cm	56	cdef (3)	49	bcd (4)	64	fgh (4)	52	cde (4)	69	gh (4)	49	bc (3)	55	cdef (4)	64	fgh (4)
pH		7.8	abc (3)	8.1	bcd (4)	7.9	abcd (4)	7.9	abc (4)	7.9	abcd (4)	8.0	abcd (3)	8.0	bcd (4)	7.8	ab (4)
NO ₂ -N + NO ₃ -N	mg/kg	0.35	a (3)	1.9	a (4)	1.1	a (4)	0.80	a (4)	1.1	a (4)	0	a (3)	58	b (4)	0.53	a (4)
Ca	mg/kg	70	a (3)	193	abcd (4)	173	abcd (4)	185	abcd (4)	248	bcd (4)	220	abcd (3)	118	ab (4)	255	bcd (4)
Mg	mg/kg	90	ab (3)	25	a (4)	303	cde (4)	55	a (4)	335	def (4)	193	abcd (3)	325	cdef (4)	463	fg (4)
Na	mg/kg	2045	abcdef (3)	1718	abc (4)	2917	h (4)	1764	abcd (4)	2459	bdefgh (4)	1893	abcde (3)	2057	abcdefg (4)	2816	fh (4)
S	mg/kg	2847	bc (3)	390	a (4)	5098	de (4)	323	a (4)	5233	def (4)	1627	ab (3)	4603	cd (4)	6095	def (4)
Cl-	mg/kg	3380	cde (3)	3161	cd (4)	3588	cdefgh (4)	3436	cde (4)	4143	efgh (4)	3289	cde (3)	3351	cde (4)	3547	cdef (4)

Table 9 Average concentrations of **heavy metals** (mg/kg) in the RO-concentrates of pilot installations A –Q. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of measurements.

Parameter	A		B		C		D		E		F		G		H		I	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
As	<0.11	(0/5)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/2)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/6)	<0.11	(0/2)
Ba	0.2 ^a	(4/5)	<0.16	(0/3)	0.36 ^b	(3/3)	<0.16	(0/2)	<0.16	(0/3)	0.24 ^{ab}	(3/3)	<0.16	(0/3)	<0.16	(0/6)	<0.16	(0/2)
Cd	<0.03	(0/5)	<0.03	(0/3)	<0.03	(0/3)	<0.03	(0/2)	<0.03	(0/3)	<0.03	(0/3)	<0.027	(0/3)	<0.03	(0/6)	<0.03	(0/2)
Cr	0.1 ^{ab}	(5/5)	0.03 ^a	(1/3)	0.15 ^{bc}	(3/3)	<0.07	(0/2)	<0.07	(0/3)	0.1 ^{ab}	(3/3)	<0.07	(0/3)	0.02 ^a	(1/6)	<0.07	(0/2)
Cu	0.71	(5/5)	<0.57	(0/3)	2.4 ^b	(3/3)	0.75 ^a	(1/2)	<0.57	(0/3)	0.97 ^a	(3/3)	<0.57	(0/3)	<0.57	(0/6)	<0.57	(0/2)
Hg	<0.01	(0/5)	<0.01	(0/3)	<0.01	(0/3)	<0.01	(0/2)	<0.01	(0/3)	<0.01	(0/3)	<0.012	(0/3)	<0.01	(0/6)	<0.01	(0/2)
Ni	0.48 ^{efgh}	(5/5)	0.25 ^{abc}	(3/3)	0.62 ^h	(3/3)	0.38 ^{bcde}	(2/2)	0.14 ^a	(3/3)	0.58 ^{fgh}	(3/3)	0.47 ^{efgh}	(3/3)	0.45 ^{fgh}	(5/6)	0.25 ^{ab}	(1/2)
Pb	<0.01	(0/5)	<0.01	(0/3)	<0.01	(0/3)	0.01 ^a	(1/2)	<0.01	(0/3)	0.01 ^a	(1/3)	<0.013	(0/3)	<0.01	(0/6)	<0.01	(0/2)
Zn	1.8 ^a	(5/5)	<1.1	(0/3)	6.6 ^b	(3/3)	<1.1	(0/2)	<1.1	(0/3)	2.1 ^a	(3/3)	0.57 ^a	(1/3)	<1.1	(0/6)	<1.1	(0/2)

Parameter	J		K		L		M		N		O		P		Q	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
As	<0.11	(0/3)	<0.11	(0/2)	<0.11	(0/3)	<0.11	(0/4)	<0.11	(0/3)	<0.11	(0/2)	<0.11	(0/1)	<0.11	(0/2)
Ba	<0.16	(0/3)	0.23 ^b	(2/2)	<0.16	(0/3)	0.09 ^a	(2/4)	<0.16	(0/3)	<0.16	(0/2)	<0.16	(0/1)	<0.16	(0/2)
Cd	<0.03	(0/3)	<0.03	(0/2)	<0.027	(0/3)	<0.03	(0/4)	<0.027	(0/3)	<0.03	(0/2)	<0.03	(0/1)	<0.03	(0/2)
Cr	0.22 ^b	(2/3)	<0.07	(0/2)	<0.074	(0/3)	<0.07	(0/4)	<0.074	(0/3)	<0.07	(0/2)	0.1 ^a	(1/1)	<0.07	(0/2)
Cu	<0.57	(0/3)	<0.57	(0/2)	0.57	(1/3)	<0.57	(0/4)	<0.57	(0/3)	<0.57	(0/2)	<0.57	(0/1)	<0.57	(0/2)
Hg	<0.01	(0/3)	<0.01	(0/2)	<0.012	(0/3)	<0.01	(0/4)	<0.012	(0/3)	<0.01	(0/2)	<0.01	(0/1)	<0.01	(0/2)
Ni	0.6 ^{fh}	(3/3)	0.43 ^{bdef}	(2/2)	0.49 ^{efgh}	(3/3)	0.44 ^{efg}	(4/4)	0.54 ^{fgh}	(3/3)	0.25 ^{abcd}	(2/2)	0.58 ^{efgh}	(1/1)	0.48 ^{efgh}	(2/2)
Pb	<0.01	(0/3)	<0.01	(0/2)	0.02 ^b	(1/3)	<0.01	(0/4)	<0.013	(0/3)	<0.01	(0/2)	<0.01	(0/1)	<0.01	(0/2)
Zn	0.37 ^a	(1/3)	1.2 ^a	(2/2)	<1.1	(0/3)	0.33 ^a	(1/4)	<1.1	(0/3)	<1.1	(0/2)	<1.1	(0/1)	<1.1	(0/2)

Table 10 Average concentrations of **antibiotics** ($\mu\text{g}/\text{kg}$) in the RO-concentrates of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). Values above detection limit are presented in bold. For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of measurements.

Parameter	A		B		C		D		E		F		G		H		I	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
Oxytetracycline	42	(5/5)	32	(1/2)	<15	(0/2)	<2.0	(0/2)	<5.5	(0/2)	45	(2/2)	<10	(0/2)	1.5	(1/6)	<10	(0/2)
Doxycycline	87 ^c	(5/5)	37 ^b	(2/2)	94 ^c	(2/2)	<3.0 ^a	(0/2)	<3.0 ^a	(0/2)	96 ^c	(2/2)	6.0 ^a	(1/2)	3.2 ^a	(2/6)	5.5 ^a	(1/2)
Sulfadiazine	9.6 ^c	(4/5)	<1.0 ^a	(0/2)	6.5 ^{bc}	(2/2)	2.5 ^{ab}	(2/2)	4.5 ^{bc}	(1/2)	4.5 ^{bc}	(2/2)	<5.5 ^a	(0/2)	6.5 ^{bc}	(5/6)	2.5 ^{ab}	(1/2)
Sulfadimidine	<1.0	(0/5)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.2	(0/6)	<1.0	(0/2)
Trimethoprim	<1.4	(0/5)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<2.0	(0/2)	<1.5	(0/2)	<2	(0/6)	<2.0	(0/2)
Ciprofloxacin	<4.4	(0/5)	<1.5	(0/2)	<3.0	(0/2)	<3.0	(0/2)	<1.0	(0/2)	<7.5	(0/2)	<1.5	(0/2)	<5.2	(0/6)	<5.5	(0/2)
Enrofloxacin	<5.0	(0/5)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<6.0	(0/2)	<3.5	(0/2)	<6.5	(0/6)	<6	(0/2)
Flumequine	0.40 ^a	(1/5)	<1.0 ^a	(0/2)	<1.0 ^a	(0/2)	<1.0 ^a	(0/2)	<1.5 ^a	(0/2)	0.50 ^a	(1/2)	<1.5 ^a	(0/2)	<2.3 ^a	(0/6)	<1.5 ^a	(0/2)
Lincomycine	0.40 ^a	(1/5)	<1.0 ^a	(0/2)	<1.0 ^a	(0/2)	<1.0 ^a	(0/2)	<1.0 ^a	(0/2)	<1.0 ^a	(0/2)	<1.5 ^a	(0/2)	<2.2 ^a	(0/6)	<1.5 ^a	(0/2)
Tilmicosine	0.40 ^{ab}	(1/5)	<10 ^a	(0/2)	<10 ^a	(0/2)	<5.5 ^a	(0/2)	<7.5 ^a	(0/2)	<6.0 ^a	(0/2)	<7.5 ^a	(0/2)	<3.5 ^a	(0/6)	2.5 ^b	(1/2)
Florfenicol	<1.8	(0/5)	<1.0	(0/2)	<1.0	(0/2)	<3.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<2.7	(0/6)	<1.0	(0/2)

Pilot installations J – Q were not included in the monitoring on antibiotics.

Table 11 Average concentrations of **bacteria and viruses** in the RO-concentrates of pilot installations A – I, based on the geometric mean. Values are shown in two units: the decadic logarithm of the concentration, and the concentration. As example: $+4 \log_{10}(\text{CFU/g})$ equals 10^4 CFU/g , and $0 \log_{10}(\text{CFU/g})$ equals 10^0 or 1 CFU/g. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter, calculated for the geometric mean ($p < 0.05$). For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of measurements. NA: not analyzed

Parameter	Unit	A		B		C		D		E		F		G		H		I	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
<i>E. coli</i>	$\log_{10}(\text{CFU/g})$	2.2 ^a	(1/2)	4.5 ^a	(2/2)	2.6 ^a	(2/2)	3.2 ^a	(2/2)	3.9 ^a	(2/2)	4.3 ^a	(2/2)	4.4 ^a	(2/2)	4.7 ^a	(2/2)	3.7 ^a	(2/2)
Enterococci	$\log_{10}(\text{CFU/g})$	3.6 ^a	(2/2)	3.1 ^a	(2/2)	3.5 ^a	(2/2)	1.6 ^a	(1/2)	<1.2 ^a	(0/2)	4.1 ^a	(2/2)	3.2 ^a	(2/2)	3.0 ^a	(2/2)	3.2 ^a	(2/2)
Clostridia	$\log_{10}(\text{CFU/g})$	3.8 ^{bc}	(2/2)	3.5 ^{ab}	(1/2)	4.6 ^b	(2/2)	1.7 ^a	(1/2)	3.0 ^{ab}	(2/2)	3.6 ^{ab}	(2/2)	3.1 ^{ab}	(2/2)	2.4 ^{ac}	(2/2)	3.1 ^{ab}	(2/2)
Bacteriophages	$\log_{10}(\text{pfu/g})$	4.9 ^a	(2/2)	NA		NA		NA		NA		NA		NA		5.0 ^a	(2/2)	NA	
Hepatitis E virus	positive		(2/2)	NA		NA		NA		NA		NA		NA		positive	(2/2)	NA	
<i>E. coli</i>	CFU/g	162		31289		395		1643		7211		18396		26552		51614		4970	
Enterococci	CFU/g	4416		1301		2898		39		17		12296		1697		978		1510	
Clostridia	CFU/g	6261		3407		35496		53		1088		4047		1212		269		1202	
Bacteriophages	pfu/g	86487		NA		NA		NA		NA		NA		NA		93220		NA	

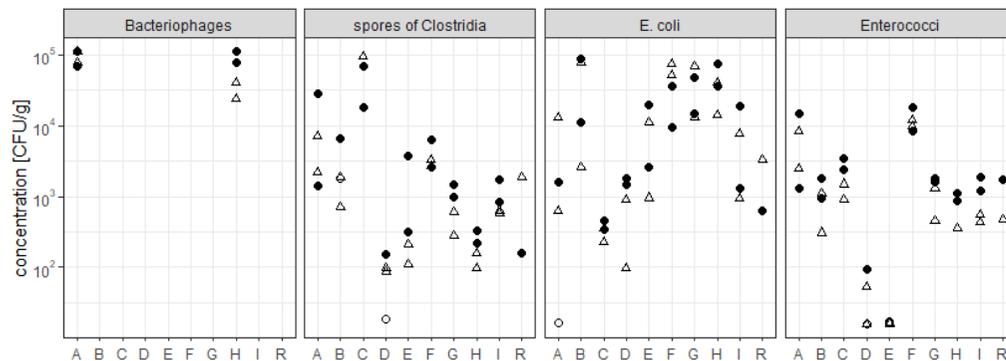


Figure 3.1 Bacteria and viruses in RO-inputs and RO-concentrates from 10 pilot installations. Solid circles indicate concentrations in RO-concentrates, open triangles indicate concentrations in RO-input

3.2.5 Average composition of RO-concentrate

Table 12 shows median and average values and standard deviations of chemical parameters of RO-concentrate as collected from 17 pilot installations. Regarding the general parameters the RO-concentrate can be characterised as a largely mineral fertiliser with N and K as the main nutrients. The greater part (86%) of Total-N is present as TAN. The average concentrations of N and K are similar to the average concentrations found in the monitoring of pilot plants in 2012-2014 (Hoeksma and de Buissonjé, 2015). TS and EC values are also at the same level. The main difference between the RO-concentrates then and now is that the RO-concentrate as presented in Table 12 shows a significantly higher concentration of volatile solids and a lower concentration of TAN than reported in 2015. This indicates that pre-treatment of the RO-input during this measuring campaign was less effective than it was in 2012-2014, possibly due to decreased use of PAM and/or Fe-salts to cut operational costs. Table 12 shows detectable concentrations of Ba, Cr, Cu, Ni, Pb and Zn in the RO-concentrate, with highest levels for Cu and Zn. Ni was most often found. Concentrations in RO-concentrate of Cr, Cu, Ni and Zn that were reported by Hoeksma et al. (2011) show no significant (not tested) differences with the results shown in this report.

From the measured antibiotics in the RO-concentrate oxytetracycline, doxycycline and sulfadiazine are most frequently found, with the highest concentration for doxycycline. Antibiotics were not included in former monitoring programs of the pilot installations.

Table 12 Median, mean and standard deviation (SD) and counts (n) of measured chemical parameters in RO-concentrate from 17 pilot installations. Numbers between () represent counts above detection limit and total counts.

Parameters	Unit	Median	Mean	SD	n
General parameters					
TS	g/kg	31	35	13	65
VS	g/kg DM	308	296	91	65
C-total	g/kg DM	101	123	69	65
TOC	mg/kg	3340	3357	1463	65
Total-N	mg/kg	6087	6356	1607	65
TAN	mg/kg	5360	5439	1404	65
P-total	mg/kg	42	72	69	65
P-PO ₄	mg/kg	13	40	52	65
K	mg/kg	7300	7183	2010	65
EC	mS/cm	55	56	13	65
pH		7.9	7.9	0.27	65
NO ₂ -N + NO ₃ -N	mg/kg	0	4.4	2.8	65
Ca	mg/kg	180	214	130	65
Mg	mg/kg	260	251	204	65
Na	mg/kg	1948	2169	765	65
S	mg/kg	3050	3623	3129	65
Cl-	mg/kg	3404	3527	1120	65
Heavy metals					
As	mg/kg DM	<3.6	<3.5		(0/50)
Ba	mg/kg DM	8.9	9.6	4.7	(14/50)
Cd	mg/kg DM	<0.89	<0.85		(0/50)
Cr	mg/kg DM	3.3	4.5	3	(16/50)
Cu	mg/kg DM	38	45	30	(13/50)
Hg	mg/kg DM	<0.40	<0.38		(0/50)
Ni	mg/kg DM	13	14	5.5	(48/50)
Pb	mg/kg DM	0.05	0.02	0.24	(3/50)
Zn	mg/kg DM	51	93	75	(16/50)
Antibiotics					
Oxytetracycline	µg/kg	32	14	21	(13/29)
Doxycycline	µg/kg	29	35	50	(19/29)
Sulfadiazine	µg/kg	6.0	9.0	12	(21/29)
Sulfadimidine	µg/kg	<1.0	<1.1		(0/29)
Trimethoprim	µg/kg	<1.0	<1.5		(0/29)
Ciprofloxacin	µg/kg	<2.0	<3.8		(0/29)
Enrofloxacin	µg/kg	<3.0	<4.7		(0/29)
Flumequine	µg/kg	1.0	0.07	0.42	(3/29)
Lincomycine	µg/kg	2.0	0.07	0.38	(2/29)
Tilmicosine	µg/kg	4.5	0.33	1.0	(3/29)
Florfenicol	µg/kg	<1.0	<1.6		(0/29)

3.2.6 Correlation between general parameters in RO-concentrate

The correlation between the general parameters in the RO-Concentrate was calculated with R package 'ggcorrplot'. The correlation matrix (Figure 3.2) shows the correlation coefficients between individual parameters. The degree of correlation is expressed in different colours, representing strong correlation (coefficient 0.6-1), moderate correlation (coefficient 0.3-0.6) and low correlation (coefficient 0.0-0.3). The correlation analysis shows that EC, dry matter and N-NH₄ have strong positive correlation with the largest number of parameters in the RO-concentrate. They show strong mutual relationship and strong relationship with total-N, K, Na and Cl⁻. EC and N-NH₄ would be strong indicators for the identity of the RO-concentrate.

Expected correlations between P-total and Dry matter or Org. matter or TOC and between TOC and Org. matter are not observed, which can possibly be explained by a variation in the dosage of flocculants.

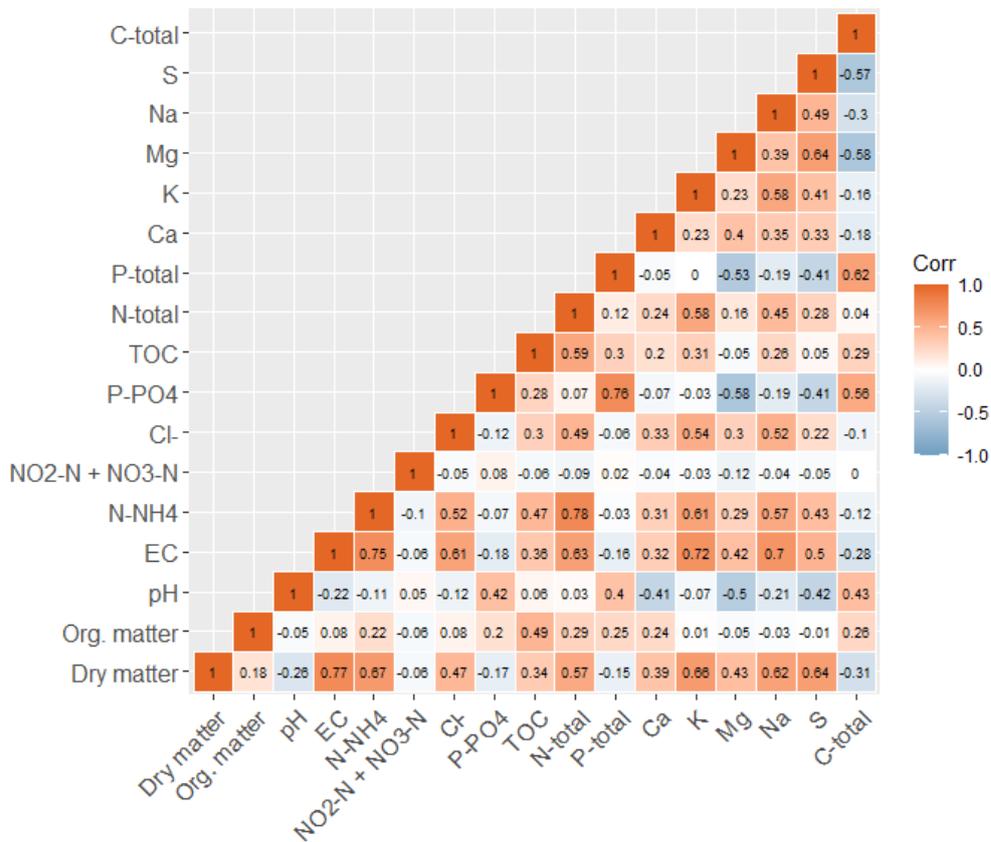


Figure 3.2 Correlation between general parameters in RO-Concentrate of 17 pilot installations. The numbers indicate the Kendall's Tau correlation coefficients (theoretical range, -1 to +1). Degrees of correlation: strong (correlation coefficient 0.6-1), moderate (correlation coefficient 0.3-0.6) and low (correlation coefficient 0.0-0.3)

3.3 Effect of installation and input on RO-concentrate

The effect of the installation and input on the composition of RO-concentrate was tested for all parameters. The installation and concentration in the input were considered as a factor and co-variable, respectively. Table 13 shows the p-values of the ANCOVA tests for the different parameters. For most of the parameters the effect of installation on RO-concentrate was significant at 0.01 and 0.05. The impact of the input on RO-concentrate was significant only for the general parameters P-total, pH, NO₂-N+NO₃-N, heavy metals Cu and Zn and antibiotic flumequine. These results illustrate the higher impact of the installation compared to the input on RO-concentrate.

Table 13 The effect of installation and input on the concentration of RO-concentrate. Installation and input were considered as a factor and co-variable, respectively. *p*-values show significance levels.

		Installation	Input
	unit	P-value	P-value
General parameters			
TS	g/kg	0.01	ns
VS	g/kg DM	0.01	ns
C-total	g/kg DM	0.01	ns
TOC	mg/kg	0.05	ns
Total-N	mg/kg	0.01	ns
TAN	mg/kg	0.01	ns
P-total	mg/kg	0.01	0.05
P-PO4	mg/kg	0.01	ns
K	mg/kg	0.01	ns
EC	mS/cm	0.01	ns
pH		0.01	0.05
NO ₂ -N + NO ₃ -N	mg/kg	ns	0.05
Ca	mg/kg	0.05	ns
Mg	mg/kg	0.01	ns
Na	mg/kg	0.01	ns
S	mg/kg	0.01	ns
Cl-	mg/kg	0.01	ns
Heavy metals			
As	mg/kg	NA	NA
Ba	mg/kg	0.01	ns
Cd	mg/kg	NA	NA
Cr	mg/kg	0.01	ns
Cu	mg/kg	ns	0.01
Hg	mg/kg	NA	NA
Ni	mg/kg	ns	ns
Pb	mg/kg	ns	ns
Zn	mg/kg	0.05	0.01
Antibiotics			
Oxytetracycline	µg/kg	0.01	ns
Doxycycline	µg/kg	0.05	ns
Sulfadiazine	µg/kg	ns	ns
Sulfadimidine	µg/kg	NA	NA
Trimethoprim	µg/kg	NA	NA
Ciprofloxacin	µg/kg	NA	NA
Enrofloxacin	µg/kg	NA	NA
Flumequine	µg/kg	ns	0.01
Lincomycine	µg/kg	NA	NA
Tilmicosine	µg/kg	ns	ns
Florfenicol	µg/kg	NA	NA

ns = not significant

NA = not available (not measured)

To study the relationship between the measured parameters in the input and RO-concentrate, regression analysis was applied for the parameters which were significant in the ANCOVA test. Figure 3.3 shows the relationship between the concentrations in the input (raw slurry) and in RO-concentrate of total-P, pH, NO₂-N+NO₃-N, Cu, Zn and Flumequine. Based on the obtained ANCOVA test results, RO-concentrate can be explained from the input concentrations of total-P, pH, N₂O-N+NO₃-N, Cu, Zn and flumequine, by 6%, 24%, 11%, 37%, 37%, and 69%, respectively. Please note, the test results of flumequine are based on only 4 measured values.

It can be concluded that, on average, the input of the pilot installations has little impact on the composition the RO-concentrate.

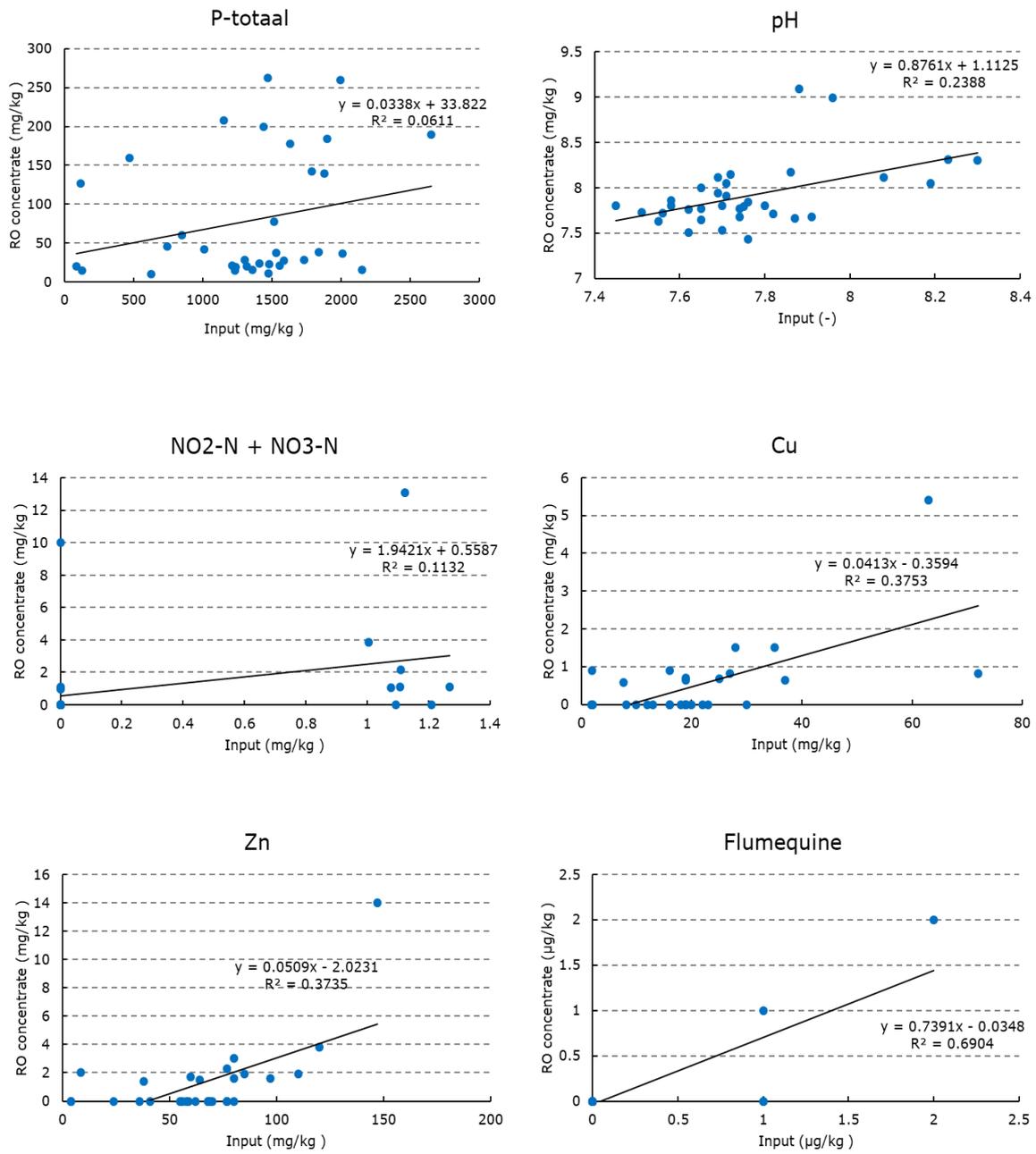


Figure 3.3 The relationship between concentration of input and RO-concentrate for total-P, pH, NO₂-N+NO₃-N, Cu, Zn and flumequine.

3.4 Test of RO-concentrate to quality criteria

3.4.1 Criteria pilot MC

$NH_4\text{-N}/\text{Total-N}$

Figures 3.4 and 3.5 show $NH_4\text{-N}/\text{Total-N}$ ratio values of RO-concentrates measured from 17 pilot installations.

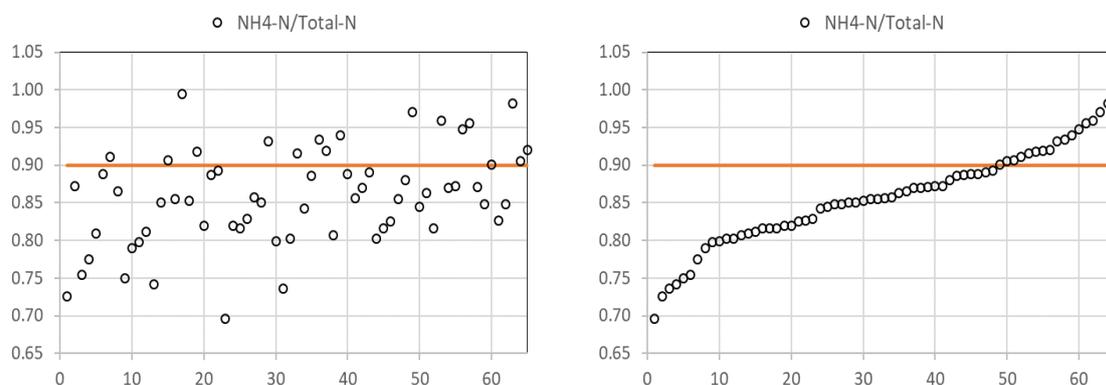


Figure 3.4 Values of $NH_4\text{-N}/\text{Total-N}$ ratio of RO-concentrates from 17 pilot installations (left) and cumulative (right) and threshold.

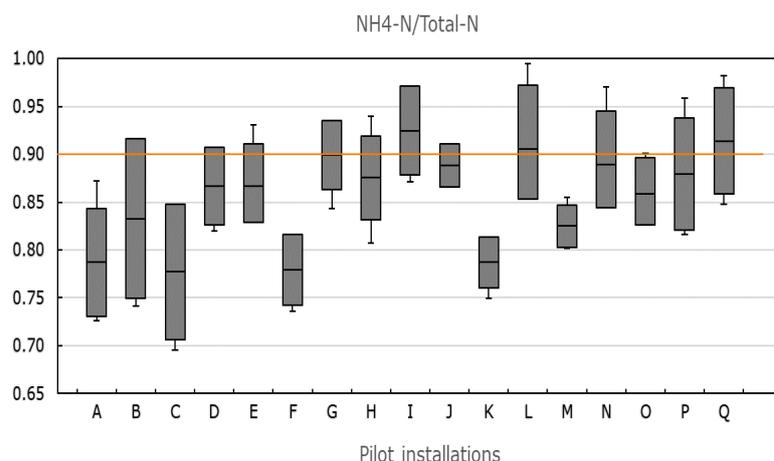


Figure 3.5 Variability of $NH_4\text{-N}/\text{Total-N}$ ratio in RO-concentrate per pilot installation. The orange line indicates the threshold. The mid-point of each box equals the mean; the lower and upper edges of each box represent the mean \pm SD, and the whiskers extend to the minimum and maximum.

Figure 3.4 shows that in the minority of the measured RO-concentrates the threshold of the $TAN/\text{Total-N}$ ratio was met; 26% of the observed $TAN/\text{Total-N}$ ratios met the threshold and the overall average $TAN/\text{Total-N}$ ratio was 0.86. The results of the monitoring in 2015 showed an average $TAN/\text{Total-N}$ ratio of 0.90 (Hoeksma and Buisonjé, 2015). A large variation in $TAN/\text{Total-N}$ ratio within and between pilot installations is reflected in Figure 3.2.

The results regarding the $TAN/\text{Total-N}$ ratio also show:

- 3 pilot installations with an average $>$ threshold,
- 12 pilot installations with one or more observations $>$ threshold,
- 5 pilot installations with all observations $<$ threshold,
- 0 pilot installations with all observations $>$ threshold.

Total-N/P₂O₅

Figures 3.6 and 3.7 show Total-N/P₂O₅ ratio values of RO-concentrates measured from 17 pilot installations.

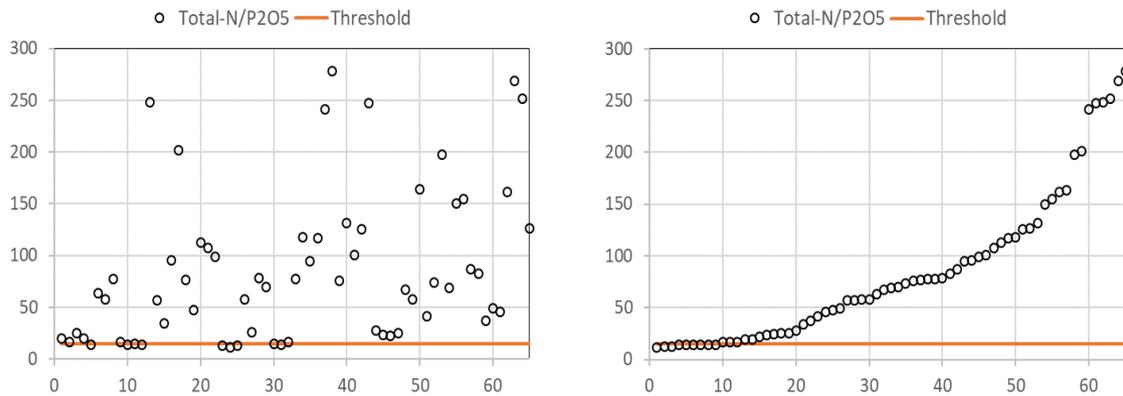


Figure 3.6 Values of Total-N/P₂O₅ ratio of RO-concentrates from 17 pilot installations(left) and cumulative (right) and threshold

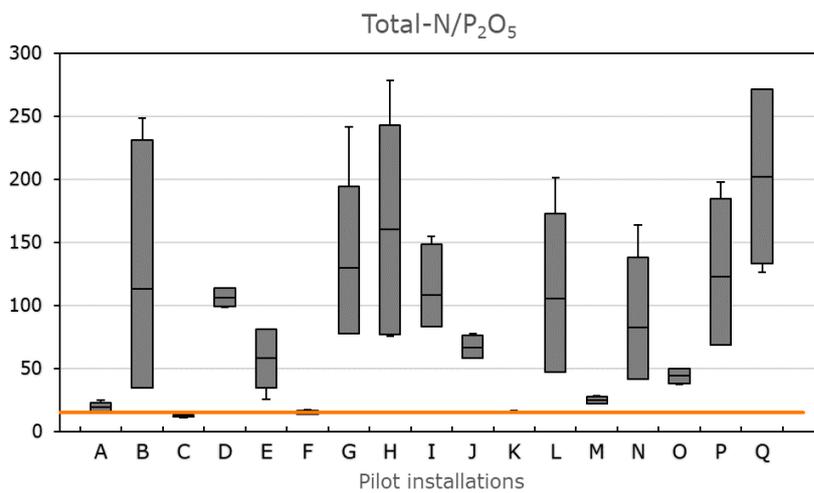


Figure 3.7 Variability of Total-N/P₂O₅ ratio in RO-concentrate per pilot installation. The orange line indicates the threshold. The mid-point of each box equals the mean; the lower and upper edges of each box represent the mean±SD, and the whiskers extend to the minimum and maximum.

Figure 3.6 shows that the majority (86%) of the observed Total-N/P₂O₅ ratios was above the threshold. The RO-concentrates of two pilot installations (C and F) in average did not meet the threshold of 15, as is shown in Figure 3.7., which can be attributed to relatively high concentrations of P₂O₅. In one sample of both RO-concentrates the threshold was just met.

EC

Figures 3.8 and 3.9 show EC values of RO-concentrates measured from 17 pilot installations.

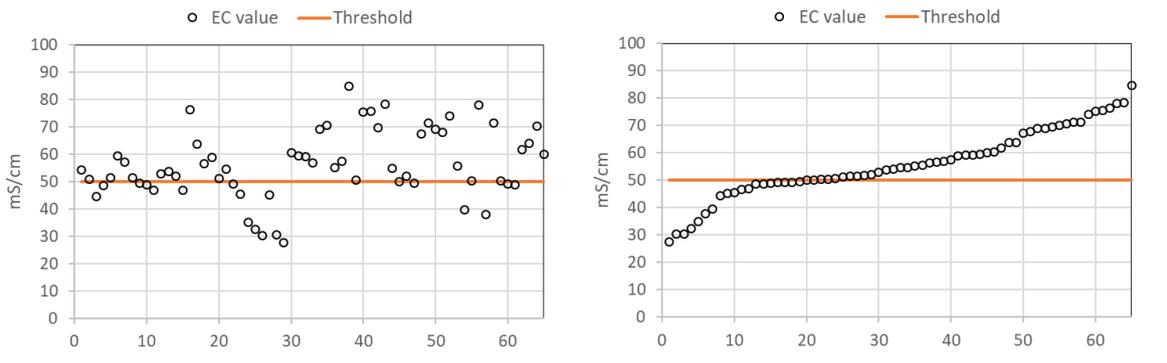


Figure 3.8 Measured values of EC of RO-concentrates from 17 pilot installations (left) and cumulative (right) and threshold

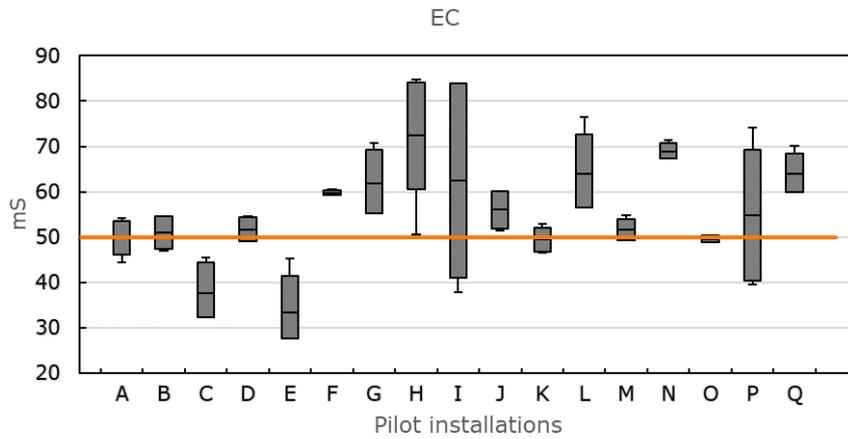


Figure 3.9 Variability of EC in RO-concentrate per pilot installation. The orange line indicates the threshold. The mid-point of each box equals the mean; the lower and upper edges of each box represent the mean \pm SD, and the whiskers extend to the minimum and maximum.

Figure 3.8 shows that the EC value in the majority (71%) of the RO-concentrate samples was above 50 mS/cm. Pilot installations C, E and K show an average EC value < 50 mS/cm (Figure 3.9).

3.4.2 Criteria RENURE

TOC/Total-N

Figures 3.10 and 3.11 show TOC/Total-N ratio values of RO-concentrates measured from 17 pilot installations.

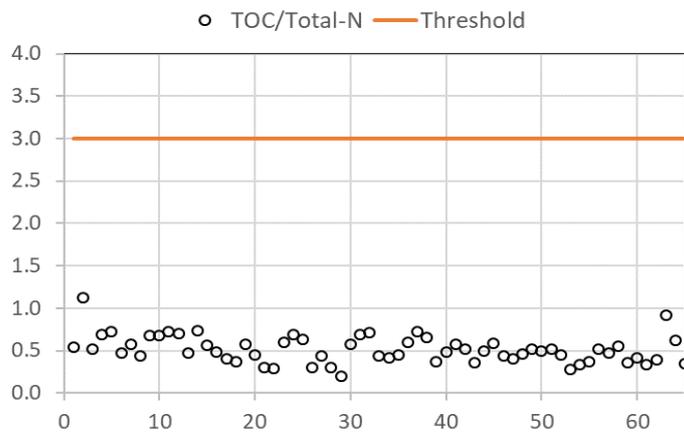


Figure 3.10 Values of TOC/Total-N ratio of RO-concentrates from 17 pilot installations and threshold

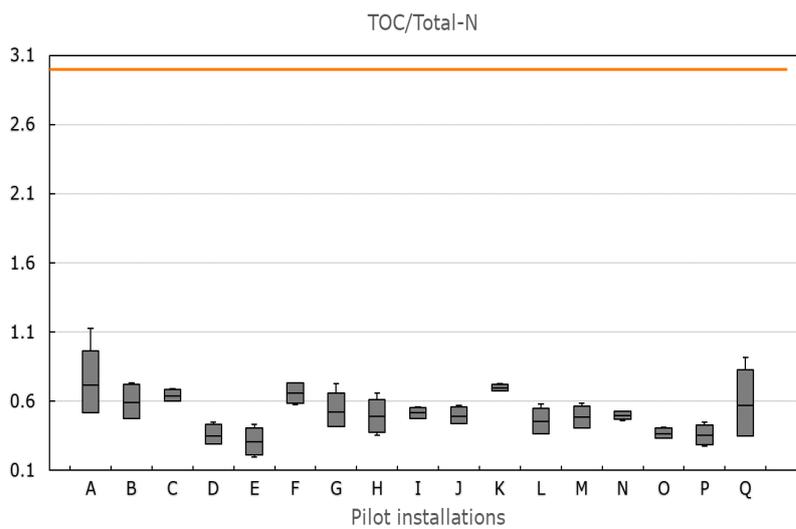


Figure 3.11 Variability of TOC/Total-N ratio of RO-concentrate per pilot installation. The orange line indicates the threshold. The mid-point of each box equals the means; the lower and upper edges of each box represent the mean \pm SD, and the whiskers extend to the minimum and maximum.

Figures 3.10 and 3.11 reveal that the RO-concentrates of all pilot installations amply met the criterion TOC/Total-N \leq 3.

Cu

Figures 3.12 and 3.13 show Cu concentrations measured in RO-concentrates measured from 17 pilot installations.

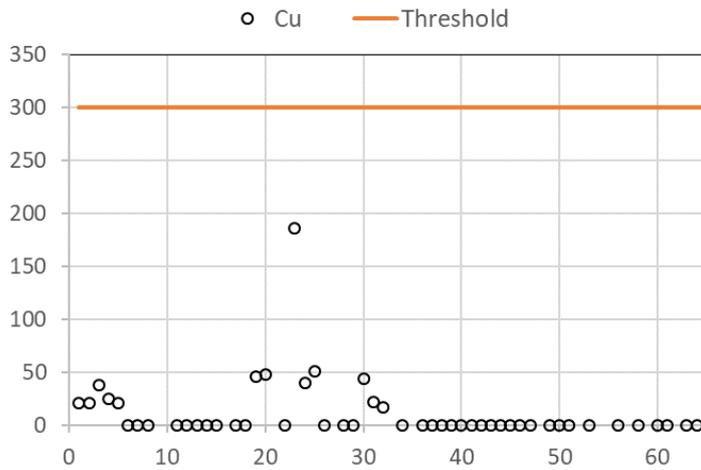


Figure 3.12 Measured concentrations (mg/kg DM) of Cu in RO-concentrates from 17 pilot installations and threshold. Values < detection limit (0.57 mg/kg DM) are shown as zero.

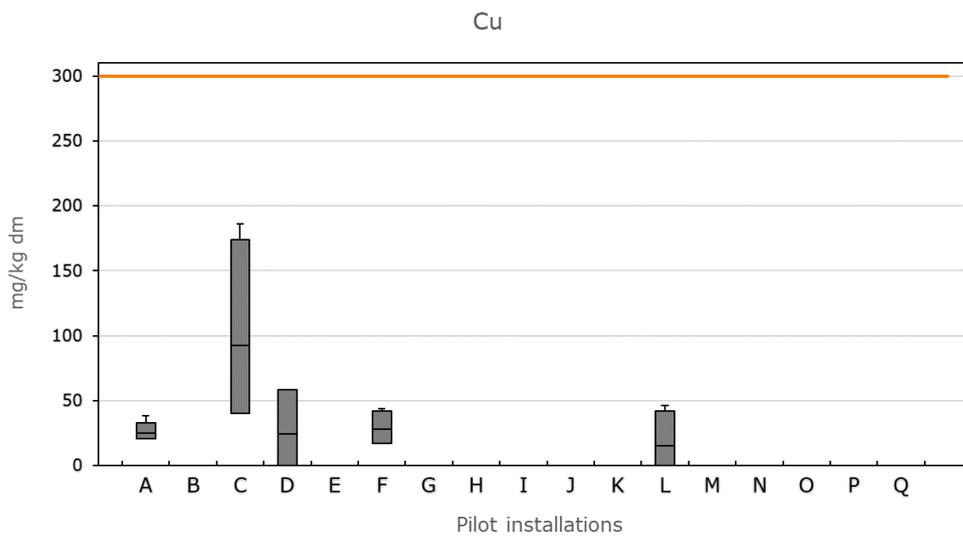


Figure 3.13 Variability of Cu in RO-concentrate per pilot installation. No box displayed means that only concentrations < detection limit (0.57 mg/kg DM) were measured. The orange line indicates the threshold. The mid-point of each box equals the mean; the lower and upper edges of each box represent the mean \pm SD, and the whiskers extend to the minimum and maximum.

The results show that RO-concentrates of all pilot installations met the threshold of Cu. Cu-values > detection limit were found in the RO-concentrates of 5 pilot installations. Highest concentrations of Cu were measured in the RO-concentrate of installation C.

Zn

Figures 3.14 and 3.15 show Zn concentrations measured in RO-concentrates from 17 pilot installations.

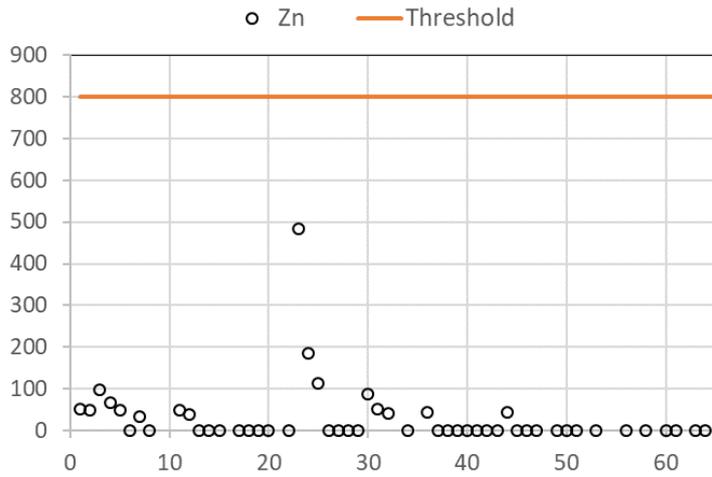


Figure 3.14 Measured concentrations (mg/kg DM) of Zn in RO-concentrates from 17 pilot installations and threshold. Values < detection limit (1.1 mg/kg DM) are shown as zero.

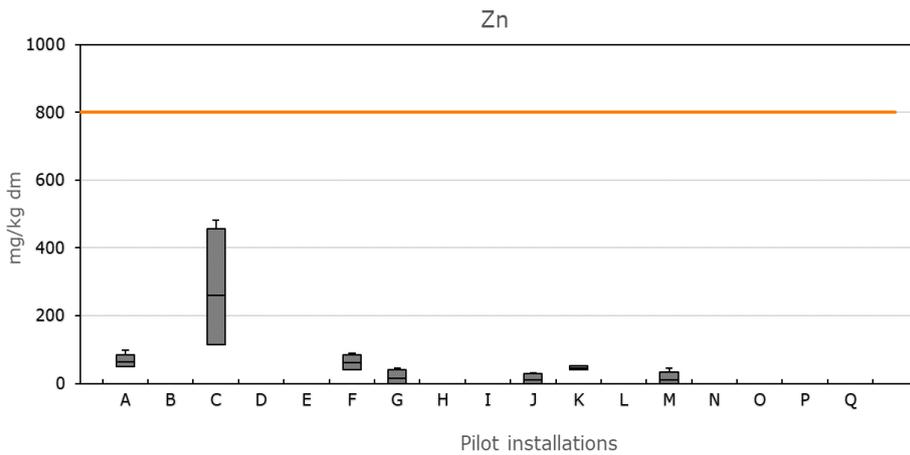


Figure 3.15 Variability of Zn in RO-concentrate per pilot installation. No box displayed means that only concentrations < detection limit (1.1 mg/kg DM) were measured. The orange line indicates the threshold. The mid-point of each box equals the mean; the lower and upper edges of each box represent the mean \pm SD, and the whiskers extend to the minimum and maximum.

The results show that RO-concentrates of all pilot installations met the threshold of Zn. Zn-values > detection limit were found in the RO-concentrates of 7 pilot installations. Highest concentrations of Zn, with a maximum value of 483 mg/kg DM, were measured in the RO-concentrate of installation C.

Table 14 shows the test results of pilot installations to quality criterions.

Table 14 Percentage of measurements and number of pilot installations that met quality criterions set for pilot MC and RENURE

	Criterion	Percentage of measurements that meet criterion	Number of pilot installations that meet criterion		
			Type 1	Type 2	Total
Pilot MC	NH ₄ -N / Total-N ¹ ≥ 0.90	26	3	0	3
	Total-N / P ₂ O ₅ ¹ ≥ 15	86	12	3	15
	EC ≥ 50 mS/cm	71	11	3	14
RENURE	NH ₄ -N / Total-N ≥ 0.90	26	3	0	3
	TOC / Total-N ≤ 3	100	12	5	17
	Cu ≤ 300 mg/kg DM	100	12	5	17
	Zn ≤ 800 mg/kg DM	100	12	5	17

¹ measured in wet product (w/w)

3.5 Composition of process flows

In Tables 15, 16, 17 and 18 the average concentrations are given of general parameters, heavy metal, antibiotics and biological parameters respectively, in the input, solid fraction, RO-input, RO-concentrate and RO-permeate of pilot installations A-I.

3.5.1 Input

Concerning general parameters the ingoing raw pig slurry of the pilot installations, originating from a great number of pig farms, on average can be considered as representative for the pig slurry produced at Dutch pig farms. The input is characterised by high moisture content and relatively high concentrations of primary nutrients Total-N, TAN, P and K and of secondary nutrient Ca. Approx. 60% of the Total-N content is TAN (Table 14). Today in the Netherlands the greater part of the produced raw pig slurry is used as a fertiliser on land, a minor part as feedstock for biogas production. For most soil types and crops P limits the amount of slurry that can be applied, regarding the legal usage standards. For most crops an additional gift of nitrogen is needed.

Trace elements Zn and Cu and Ba are present in concentrations >1mg/kg, other heavy metals are below detection limit (Hg) or <1 mg/kg. Of the measured antibiotics, oxytetracycline, doxycycline and sulfadiazine are generally found in the input of all pilot installations. The monitored bacteria and viruses are all found in the input, with concentrations of log 4-5.

3.5.2 Solid fraction

The solid fraction shows high concentrations of valuable constituents, especially primary nutrients Total-N, TAN, P and K and secondary nutrients Ca, Mg and S. Together with a high content of VS the solid fraction represents a valuable organic fertiliser/soil improver. It also finds application as a feed stock for aerobic and anaerobic fermentation processes. Concentrations of heavy metals and antibiotics in the solid fraction are 5-fold compared with the input. The solid fraction is a source of trace elements. Most of them do not contribute to the fertilizing value of the solid fraction. The concentrations of As, Cd, Cr, Ni and Pb are not of environmental concern at responsible agricultural use of the solid fraction according Good Agricultural Practices. The concentrations of Cu and Zn on the other hand have to be taken into account in the fertilization plan (Ehlert and Hoeksma, 2011). Concentrations of biological parameters in the solid fraction are slightly different from the input; only the concentration of spore forming Clostridia is significantly higher in the solid fraction.

3.5.3 RO-input

TAN, K, Na and Cl⁻ in the RO-input have similar concentration levels as in the raw slurry and solid fraction. The other general parameters are found in significantly lower concentrations in the RO-input as compared with the raw slurry and solid fraction. The concentration of VS of 319 g/kg DM in the RO-input is about half from the concentration in the raw slurry but high considering possible fouling of the RO-membranes. The concentrations of heavy metals and antibiotics in the RO-input are slightly but not significantly different from the input. No significant change is observed between the RO-input and the raw pig slurry regarding concentrations of *E.coli* and bacteriophages; the concentrations of enterococci and Clostridia are significantly lower in the RO-input.

3.5.4 RO-concentrate

The concentrations of the general parameters in the RO-concentrate on average are approximately 2.4 times higher compared with the RO-input, corresponding with the findings in former monitoring campaigns.

Concentrations of primary nutrients Total-N, TAN and K in the RO-concentrate are higher than in the input raw slurry. 86% of Total-N is TAN. RO-concentrate is a valuable mineral N and K fertiliser with N-efficiency close to the efficiency of chemical N fertilisers (Ehlert and Hoeksma, 2011). Secondary nutrients Na and S add to the fertilizing value. Please note the high concentrations of S in the RO-input and RO-concentrate compared with the raw slurry; S, in the form of iron(III)sulphate, was added in the process of the majority of the installations to support solid/liquid separation. Detectable

concentrations of heavy metals in the RO-concentrate are all <1 mg/l. Micro elements Cu and Zn contribute to the fertilising value of the RO-concentrate. The concentration of Ni (found in almost all samples) is not to be neglected from an environmental point of view. The antibiotics oxytetracycline, doxycycline and sulfadiazine are frequently found in the RO-concentrate in much lower concentrations compared with the raw slurry. The concentrations of biological parameters in the RO-concentrate are similar to the RO-input.

3.5.5 RO-permeate

All general parameters are present in measurable concentrations in the RO-permeate. C-total, Total-N and TAN are the main components. TAN is a key parameter in controlling the quality of surface water. Although there is no general legal threshold set for TAN, several water boards require a limiting moving average of 10-15 mg/l to permit the RO-permeate to be put on surface water. On average the RO-permeate does not meet this requirement. Heavy metals and antibiotic residues are sporadically found in the RO-permeate.

Concentrations of most heavy metals in the permeate are above the Dutch standards for surface water (see table below in this section). As and Hg, with values below detection limits, cannot accurately be tested against the standards because the detection limits of the method of analysis were higher than the standards.

PNEC's (= Predicted No Effect Concentrations) were found for some of the monitored antibiotics (see table below in this section). Concentrations in the RO-permeate of oxytetracycline, doxycycline and tilmicosine are above the PNEC values.

Microorganisms are present in part of the samples, in concentrations close to the limit of detection.

Standards of heavy metals for surface water (annual average concentration for long-term exposure):

Parameter	Standard (µg/l)	Remark
Cd	3.4	
Hg	0.08 – 0.25	Depending on water lime content
Pb	1.2	
As	0.5	
Cr	3.4	
Ni	4	
Cu	2.4	
Zn	7.8	
Ba	73	
V	3.5	
U	0.17	
Co	0.2	
Se	0.052	

Source: <https://rvs.rivm.nl>

PNEC's (= Predicted No Effect Concentrations) for antibiotics in surface water (Lahr et al, 2019):

Parameter	PNEC (µg/l)
Oxytetracycline	0.31
Doxycycline	1.39
Sulfadiazine	50
Sulfamethoxine	0.12
Trimethoprim	16
Tilmicosine	0.16
Florfenicol	4.81

Table 15 Average concentrations of general parameters in 5 process flows of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

parameter	Unit	Input		Solid fraction		RO-input		RO-concentrate		RO Permeaat	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
TS	g/kg	61 ^a	(35)	283 ^b	(35)	16 ^c	(35)	35 ^{ac}	(35)	NA	0
VS	g/kg dm	654 ^a	(35)	753 ^b	(35)	319 ^{cd}	(35)	296 ^d	(35)	NA	0
C-total	g/kg dm	340 ^a	(33)	378 ^b	(33)	127 ^c	(33)	123 ^c	(33)	136 ^c	(33)
TOC	mg/kg	2343 ^a	(35)	2776 ^b	(35)	1463 ^c	(35)	3357 ^b	(35)	16 ^d	(35)
Total-N	mg/kg	4687 ^a	(35)	11972 ^b	(35)	2872 ^c	(35)	6356 ^d	(35)	62 ^e	(35)
TAN	mg/kg	2763 ^a	(35)	2589 ^a	(35)	2438 ^a	(35)	5439 ^b	(35)	59 ^c	(35)
P	mg/kg	1311 ^a	(35)	7140 ^b	(35)	52 ^c	(35)	72 ^c	(35)	3.9 ^c	(35)
P-PO ₄	mg/kg	198 ^a	(35)	351 ^b	(35)	28 ^c	(35)	40 ^c	(35)	0.71 ^c	(35)
K	mg/kg	3489 ^a	(35)	3846 ^a	(35)	3074 ^a	(35)	7183 ^b	(35)	11 ^c	(35)
EC	mS	24 ^a	(35)	4.2 ^b	(35)	28 ^a	(35)	56 ^c	(35)	2.7 ^b	(35)
pH		7.8 ^a	(35)	8.4 ^b	(35)	7.8 ^a	(35)	7.9 ^{ac}	(35)	7.5 ^c	(35)
NO ₂ -N + NO ₃ -N	mg/kg	0.26 ^a	(35)	8.4 ^a	(35)	1.4 ^a	(35)	4.4 ^a	(35)	0.29 ^a	(35)
Ca	mg/kg	1837 ^a	(35)	9113 ^b	(35)	149 ^c	(35)	214 ^c	(35)	6.9 ^c	(35)
Mg	mg/kg	921 ^a	(35)	4546 ^b	(35)	129 ^c	(35)	251 ^{ac}	(35)	1.1 ^c	(35)
Na	mg/kg	982 ^a	(35)	914 ^a	(35)	952 ^a	(35)	2169 ^b	(35)	17 ^c	(35)
S	mg/kg	748 ^a	(35)	4089 ^b	(35)	1692 ^c	(35)	3623 ^b	(35)	27 ^a	(35)
Cl-	mg/kg	1570 ^a	(35)	1023 ^b	(35)	1591 ^a	(35)	3527 ^c	(35)	22 ^d	(35)

Table 16 Average concentrations of **heavy metals** (mg/kg) in 5 process flows of pilot installations A - I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between process flows for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

Parameter	Input		Solid fraction		RO-input		RO-concentrate		RO Permeaat	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
As	0.00 ^a	(1/30)	0.06 ^a	(5/30)	<0.11 ^a	(0/30)	<0.11 ^a	(0/50)	<0.12 ^a	(0/30)
Ba	3.0 ^a	(30/30)	15 ^a	(29/30)	0.03 ^a	(4/30)	0.07 ^a	(14/50)	0.65 ^a	(2/30)
Cd	0.03 ^a	(23/30)	0.18 ^b	(29/30)	<0.03 ^a	(0/30)	<0.03 ^a	(0/50)	0.01 ^a	(1/30)
Cr	0.63 ^a	(28/30)	3.1 ^b	(29/30)	0.01 ^a	(2/30)	0.04 ^a	(16/50)	0.13 ^a	(1/30)
Cu	21 ^a	(30/30)	119 ^b	(29/30)	0.11 ^a	(2/30)	0.34 ^a	(13/50)	3.3 ^a	(1/30)
Hg	<0.01 ^a	(0/30)	<0.03 ^a	(0/30)	<0.01 ^a	(0/30)	<0.01 ^a	(0/50)	<0.01 ^a	(0/30)
Ni	0.68 ^a	(30/30)	3.2 ^b	(30/30)	0.21 ^a	(26/30)	0.44 ^a	(48/50)	0.15 ^a	(3/30)
Pb	0.16 ^a	(27/30)	0.53 ^b	(29/30)	<0.01 ^a	(0/30)	0.00 ^a	(3/50)	0.02 ^a	(2/30)
Zn	65 ^a	(30/30)	335 ^b	(29/30)	0.55 ^a	(7/30)	0.83 ^a	(16/50)	12 ^a	(1/30)

Table 17 Average concentrations of **antibiotics** ($\mu\text{g}/\text{kg}$) in 5 process flows of pilot installations A - I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between process flows for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

parameter	Input		Solid fraction		RO-input		RO-concentrate		RO Permeaat	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
Oxytetracycline	95 ^a	(19/26)	443 ^b	(23/27)	30 ^a	(9/29)	14 ^a	(9/29)	1.5 ^a	(1/29)
Doxycycline	289 ^b	(25/26)	1347 ^c	(25/27)	62 ^{ab}	(15/29)	34 ^{ab}	(18/29)	2.3 ^a	(1/29)
Sulfadiazine	5.4 ^a	(16/26)	3.9 ^a	(19/27)	3.3 ^a	(17/29)	8.4 ^b	(21/29)	0.17 ^a	(1/29)
Sulfadimidine	<1.0 ^a	(0/26)	<1.2 ^a	(0/27)	<1.1 ^a	(0/29)	<1.1 ^a	(0/29)	<1.0 ^a	(0/29)
Trimethoprim	0.12 ^a	(1/26)	<1.9 ^a	(0/27)	<1.2 ^a	(0/29)	<1.5 ^a	(0/29)	<1.0 ^a	(0/29)
Ciprofloxacin	<3.5 ^a	(0/26)	<16 ^a	(0/27)	<2.5 ^a	(0/29)	<3.8 ^a	(0/29)	<1.7 ^a	(0/29)
Enrofloxacin	0.08 ^a	(1/26)	<25 ^a	(0/27)	<3.3 ^a	(0/29)	<4.7 ^a	(0/29)	0.12 ^a	(2/29)
Flumequine	0.20 ^a	(4/26)	0.80 ^a	(4/27)	0.07 ^a	(1/29)	0.11 ^a	(2/29)	<1.0 ^a	(0/29)
Lincomycine	<1.3 ^a	(0/26)	0.20 ^a	(1/27)	<1.4 ^a	(0/29)	0.07 ^a	(1/29)	0.03 ^a	(1/29)
Tilmicosine	0.95 ^a	(4/26)	5.0 ^a	(10/27)	0.23 ^a	(2/29)	0.25 ^a	(2/29)	9.7 ^a	(2/20)
Florfenicol	<1.5 ^a	(0/26)	<1.4 ^a	(0/27)	<1.3 ^a	(0/29)	<1.6 ^a	(0/29)	<1.0 ^a	(0/29)

Table 18 Average concentrations of **microorganisms** in 5 process flows collected from 10 pilot installations, based on the geometric mean. Values are shown in two units: the decadic logarithm of the concentration, and the concentration. As example: $+4 \log_{10}(\text{CFU/g})$ equals 10^4 CFU/g, and $0 \log_{10}(\text{CFU/g})$ equals 10^0 or 1 CFU/g. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter, calculated for the geometric mean ($p < 0.05$). When calculating the average of a process step for which a part of the measurements were smaller than the detection limit, the detection limit was used. The numbers between brackets represent the number of positive values and the total number of measurements.

	Unit	Input		Solid fraction		RO-input		RO-concentrate		RO-permeate	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
<i>E. coli</i>	$\log_{10}(\text{CFU/g})$	4.04 ^a	(19/19)	4.09 ^a	(19/19)	3.67 ^a	(19/19)	3.67 ^a	(18/19)	-2.92 ^b	(5/20)
Enterococci	$\log_{10}(\text{CFU/g})$	4.07 ^a	(19/19)	4.50 ^a	(19/19)	2.70 ^b	(17/19)	2.96 ^b	(16/19)	-2.74 ^c	(6/20)
Clostridia	$\log_{10}(\text{CFU/g})$	4.64 ^a	(19/19)	5.48 ^b	(19/19)	2.99 ^c	(19/19)	3.15 ^c	(17/19)	-2.46 ^d	(10/20)
Bacteriophages	$\log_{10}(\text{pfu/g})$	5.45 ^a	(3/3)	4.89 ^a	(3/3)	4.73 ^a	(4/4)	4.95 ^a	(4/4)	-2.21 ^b	(4/4)
Hepatitis E virus		positive	(3/3)	positive	(3/3)	positive	(3/4)	positive	(4/4)	negative	0/4)
<i>E. coli</i>	CFU/g	10972		12219		4697		4683		0.00122	
Enterococci	CFU/g	11844		31445		505		916		0.00183	
Clostridia	CFU/g	43535		303940		974		1418		0.00348	
Bacteriophages	pfu/g	281050		76970		53730		89790		0.00611	

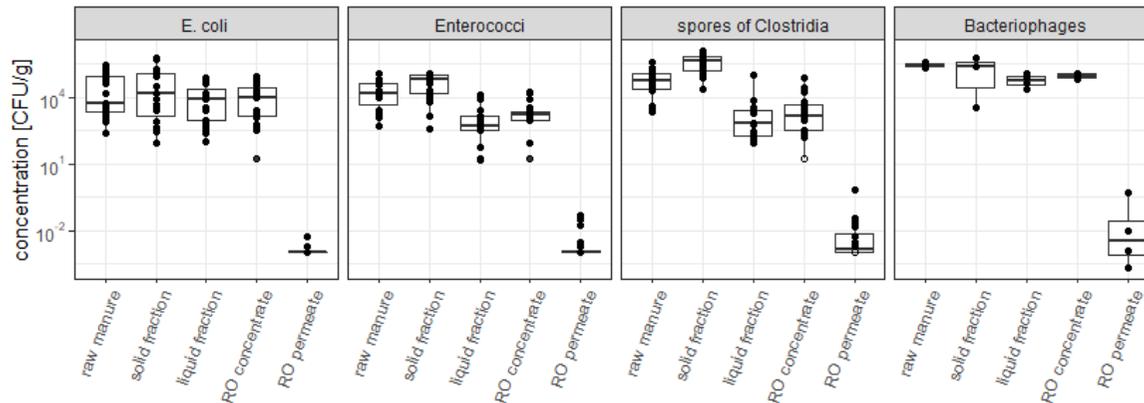


Figure 3.16 Bacteria and viruses in process flows of 10 pilot installations. Solid circles indicate measured concentrations, open circles indicate the limit of quantification in case measurements were below limit of quantification.

3.6 Comparison of type 1 and type 2 processes

Table 19 gives the average concentrations of the measured parameters in the input and RO-concentrate of the pilot installations that are classified into two types: Type 1 installations had a process set-up starting with separation by sedimentation or mechanical solid/liquid separation followed by dissolved air flotation (DAF) and using polyacrylamide (PAM) and iron(III)sulphate as additives. Type 2 installations (A, C, F, K and M) had a process set-up starting with DAF followed by mechanical solid/liquid separation (belt press or screw press) and using PAM only as an additive (see Table 1).

Table 19 shows no significant differences in chemical composition between the input of type 1 and type 2 installations, except TOC and Na which are significantly higher in the input of type 2 installations. The input of type 2 installations had lower concentrations of *E. coli* and enterococci. The RO-concentrates of type 1 and type 2 installations show significantly different concentrations of general parameters; concentrations of TS, VS, C-total and TOC are higher, the concentration of TAN is lower in the concentrate of type 2 installations. The TAN/Total-N ratios of type 1 and type 2 concentrates are 0.90 and 0.78 resp. Concentrations of heavy metals and antibiotics are higher in the RO-concentrates of type 2 installations. These findings indicate that the pre-treatment process of type 2 is less effective than the pre-treatment process of type 1.

The RO-concentrates of type 2 installations show higher concentrations of *E.coli* and enterococci and lower concentration of spores of sulphite reducing Clostridia.

Table 19 Average concentrations of measured parameters in raw slurry and in RO-concentrate of type 1 and type 2 installations. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between membrane filtration types for the designated parameter ($p < 0.05$). A single value between brackets represents the number of measurements, the double values between brackets represent the number of measurements > detection limit and the total number of measurements.

	Unit	Input				RO-concentrate			
		Type 1		Type 2		Type 1		Type 2	
		Mean	n	Mean	n	Mean	n	Mean	n
General parameters									
TS	g/kg	56 ^a	(24)	72 ^a	(11)	37 ^a	(46)	28 ^b	(19)
VS	g/kg ds	660 ^a	(24)	641 ^a	(11)	270 ^a	(46)	368 ^b	(19)
C-total	g/kg ds	346 ^a	(23)	341 ^a	(9)	84 ^a	(45)	220 ^b	(18)
TOC	mg/kg	1963 ^a	(24)	3171 ^b	(11)	3028 ^a	(46)	4154 ^b	(19)
Total-N	mg/kg	4285 ^a	(24)	5563 ^a	(11)	6485 ^a	(46)	6462 ^a	(19)
TAN	mg/kg	2535 ^a	(24)	3240 ^a	(11)	5811 ^a	(46)	5052 ^b	(19)
P-total	mg/kg	1225 ^a	(24)	1499 ^a	(11)	33 ^a	(46)	169 ^b	(19)
P-PO ₄	mg/kg	188 ^a	(24)	220 ^a	(11)	11 ^a	(46)	110 ^b	(19)
K	mg/kg	3183 ^a	(24)	4155 ^a	(11)	7372 ^a	(46)	6726 ^a	(19)
EC	mS	23 ^a	(24)	27 ^a	(11)	59 ^a	(46)	50 ^b	(19)
pH		7.7 ^a	(24)	7.9 ^b	(11)	7.8 ^a	(46)	8.1 ^b	(19)
NO ₂ -N + NO ₃ -N	mg/kg	0.29 ^a	(24)	0.22 ^a	(11)	6.0 ^a	(46)	0.74 ^a	(19)
Ca	mg/kg	1798 ^a	(24)	1923 ^a	(11)	237 ^a	(46)	159 ^b	(19)
Mg	mg/kg	827 ^a	(24)	1126 ^a	(11)	342 ^a	(46)	32 ^b	(19)
Na	mg/kg	906 ^a	(24)	1131 ^b	(11)	2341 ^a	(46)	1752 ^b	(19)
S	mg/kg	736 ^a	(24)	775 ^a	(11)	4631 ^a	(46)	481 ^b	(19)
Cl-	mg/kg	1454 ^a	(24)	1824 ^a	(11)	3691 ^a	(46)	3131 ^a	(19)
Heavy metals									
Cd	mg/kg	0.03 ^a	(13/19)	0.04 ^a	(10/11)	<0.03	(0/33)	<0.03	(0/17)
Hg	mg/kg	<0.01	(0/19)	<0.01	(0/11)	<0.01	(0/33)	<0.01	(0/17)
Pb	mg/kg	0.07 ^a	(17/19)	0.32 ^a	(10/11)	0.00 ^a	(2/33)	0.00 ^a	(1/17)
As	mg/kg	<0.11 ^a	(0/19)	0.01 ^a	(1/11)	<0.11	(0/33)	<0.11	(0/17)
Cr	mg/kg	0.56 ^a	(17/19)	0.76 ^a	(11/11)	0.03 ^a	(5/33)	0.07 ^a	(11/17)
Ni	mg/kg	0.57 ^a	(19/19)	0.89 ^b	(11/11)	0.41 ^a	(31/33)	0.51 ^a	(17/17)
Cu	mg/kg	16 ^a	(19/19)	29 ^a	(11/11)	0.10 ^a	(2/33)	0.80 ^a	(11/17)
Zn	mg/kg	54 ^a	(19/19)	82 ^a	(11/11)	0.09 ^a	(2/33)	2.3 ^b	(14/17)
Ba	mg/kg	2.5 ^a	(19/19)	3.8 ^a	(11/11)	<0.16 ^a	(0/33)	0.22 ^b	(13/17)
Antibiotics									
Oxytetracycline	µg/kg	114 ^a	(13/19)	48 ^a	(6/7)	2.4 ^a	(2/18)	37 ^b	(7/9)
Doxycycline	µg/kg	275 ^a	(19/19)	328 ^a	(6/7)	7.1 ^a	(9/18)	90 ^b	(9/9)
Sulfadiazine	µg/kg	3.2 ^a	(11/19)	11 ^a	(5/7)	8.7 ^a	(13/18)	7.8 ^a	(8/9)
Sulfadimidine	µg/kg	<1.0	(0/19)	<1	(0/7)	<1.2	(0/18)	<1.0	(0/9)
Trimethoprim	µg/kg	0.16	(1/19)	<1.4	(0/7)	<1.6	(0/18)	<1.4	(0/9)
Ciprofloxacin	µg/kg	<3.2	(0/19)	<4.4	(0/7)	<3.4	(0/18)	<4.8	(0/9)
Enrofloxacin	µg/kg	0.11	(1/19)	<3.6	(0/7)	<4.8	(0/18)	<4.4	(0/9)
Flumequine	µg/kg	0.06 ^a	(1/19)	0.57 ^b	(3/7)	<2.1	(0/18)	0.33	(2/9)
Lincomycine	µg/kg	<1.3	(0/19)	<1.4	(0/7)	<2.0	(0/18)	0.25	(1/9)
Tilmicosine	µg/kg	0.94 ^a	(3/19)	1.0 ^a	(1/7)	0.26	(1/18)	0.22	(1/9)
Florfenicol	µg/kg	<1.4	(0/19)	<1.6	(0/7)	<1.7	(0/18)	<1.4	(0/9)
Bacteria and viruses									
<i>E. coli</i>	log ₁₀ (CFU/g)	4.44 ^a	(12/12)	3.30 ^b	(6/6)	4.07 ^a	(12/12)	3.02 ^b	(5/6)
Enterococci	log ₁₀ (CFU/g)	4.39 ^a	(12/12)	3.66 ^b	(6/6)	2.55 ^a	(9/12)	3.73 ^b	(6/6)
Clostridia	log ₁₀ (CFU/g)	4.79 ^a	(12/12)	4.52 ^a	(6/6)	2.81 ^a	(10/12)	3.98 ^b	(6/6)
Bacteriophages	CFU/g	5.52 ^a	(2/2)	5.30 ^a	(1/1)	4.97 ^a	(2/2)	4.94 ^a	(2/2)

4 Discussion and conclusions

The results of the monitoring program provide a quality update of the end products from the manure processing plants that participate in the pilot Mineral concentrate of the Netherlands. Given the goals of the monitoring, the survey is focussed on the products that remain after reverse osmosis: RO-concentrate and RO-permeate. RO-concentrate is intended (desired) to be used as a mineral fertiliser, with characteristics of chemical inorganic (mineral) N fertiliser, above the threshold of the Nitrates Directive (91/676/EEC). The RO-permeate is intended to be discharged to surface water. Both products have to meet specific quality standards related to these uses, that are or will be introduced at national and/or European level to promote nutrient use from renewable resources without a negative impact to the environment and human health.

RO-concentrate and Fertilising Products Regulation (FPR)

To qualify for free trade as a fertiliser the RO-concentrate has to meet the quality standards of the Fertiliser Act of the Netherlands given in the Fertiliser Decree and the new facultative European Fertilising Products Regulation (EC/2019/1009) which will come into force from July 17th, 2022 onwards. Currently EC/2019/1009 is being implemented in the Fertiliser Decree of the Netherlands. This process of implementation will take place this year up to July 2022. The use of RO-concentrate will depend on the result of negotiation and adaptation of the RENURE proposals of JRC. The results of this process will be known most likely at the end of this year. EU member states have to implement the outcome in their directives for use of nitrogen fertilising products made from animal manure. The situation of January 2021 is given in Table 20a (nutrients) and Table 20b (contaminants).

Although RO-concentrates have an elevated content of nitrogen and potassium, their concentrations are too low to meet the requirements of FPR (Table 20a). Therefore RO-concentrates have no perspective yet for free trade between member states using a CE marking. Free trade has to follow national legislation and rules for mutual recognition. RO-concentrates however are produced for trade within the region where the manure is processed. For free trade with a CE marking a further increase in concentration is required. New types of membranes are currently tested and do show a perspective to reach higher concentrations levels.

Regional trade is promoted if nitrogen from RO-concentrates can be used on an equal basis as synthetic nitrogen from mineral nitrogen fertilisers. On average or mean values of mineral concentrates can meet RENURE criteria as proposed by JRC.

Table 20a Median and mean of measured chemical parameters in RO-concentrate from 17 pilot installations compared with quality standards of the EU fertilising products regulation EC/2019/1009 (FPR) and by JRC proposed RENURE criterions (RENURE); situation January 2021).

Parameters	Unit	Median	Mean	FPR	RENURE
				PFC 1 (C)(I)(b)(ii) Compound liquid macro-nutrient fertiliser	proposal JRC
General parameters					
C organic	%	0.3	0.3	1	*
Total-N	%	0.6	0.6	1.5	*
Mineral-N/Total-N	%	88	86	*	≥90
TOC/Total-N		0.5	0.5	*	≤3
N-forms other than NO ₂ -N + NO ₃ -N	%	88	86		>60% obligation to inject into the soil
P ₂ O ₅	%	0.004	0.007	1.5	
K ₂ O	%	0.9	0.9	1.5	
CaO	%	0.03	0.03	0.75	
MgO	%	0.04	0.04	0.75	
Na ₂ O	%	0.23	0.26	0.5	
SO ₃	%	0.76	0.91	0.75	
sum of declared nutrients	%	1.7	1.9	7	
pH		7.9	7.9		
pH-H ₂ O					>5.5 obligation to inject into the soil

Table 20b Median and mean of measured chemical parameters in RO-concentrate from 17 pilot installations compared with quality standards of the EU fertilising products regulation EC/2019/1009 (FPR) and by JRC proposed RENURE criteria (RENURE); situation January 2021).

Parameters	Unit	Median	Mean	FPR	RENURE
				PFC 1 (C)(I)(b)(ii) Compound liquid macronutrient fertiliser	proposal JRC
Heavy metals					
As	mg/kg DM	<3.6	<3.5	40	
Cd	mg/kg DM	<0.89	<0.85	3	
Cr	mg/kg DM	3.3	4.5	*	
Cr6+	mg/kg DM			2	
Cu	mg/kg DM	38	45	600 (*)	300
Hg	mg/kg DM	<0.40	<0.38	1	
Ni	mg/kg DM	13	14	100	
Pb	mg/kg DM	0.05	0.02	120	
Zn	mg/kg DM	51	93	1500 (*)	800
Pathogens					
Salmonella				not for inorganic fertilisers	
E. coli				not for inorganic fertilisers	
Other				biuret. perchlorate	Generic requirements for Good Agricultural Practice to control nutrient application rates and emission of GHG and NH ₃ during application and storage.

(*) not intentionally added

RO-concentrate vs criteria of the pilot MC

A minority of the pilot installations produce on average an RO-concentrate that meets the criterion $TAN/Total-N \geq 0.90$. This result is a setback compared with the average RO-concentrate that was measured in 2012-2014 when the majority of the 9 pilot installations met this criterion. The RO-concentrates of all type 2 installations do not meet the criterion. The observation regarding the TAN/Total-N ratio means that the current RO-concentrate cannot be qualified as an N-fertiliser with synthetic fertiliser characteristics.

Two installations (of type 2) do not meet the criterion $Total-N/P_2O_5 \geq 15$ which is due to high concentrations of Total-P (and P_2O_5). Three installations (two of type 2) do not meet the criterion $EC \geq 50$ mS/cm. From these findings it can be concluded that on average the type 1 pre-treatment process is more effective than the type 2 pre-treatment process and that the RO-concentrate of type 1 installations is qualitatively better than of type 2 installations. Hjorth et al. (2010) showed that the use of a coagulant is indispensable to ensure effective solid/liquid separation. But the addition of a coagulant is not the only condition for meeting the quality standards of the pilot MC. The results of this monitoring campaign have shown that most of the installations are able to produce an RO-concentrate that meet the quality requirements but also that between installations there is a great variation in the composition of the RO-concentrate. This indicates that the way the installation is designed and operated and the way it is maintained are essential for good performance and to achieve a high quality RO-concentrate. Producers have to make an extra effort to meet the quality requirements and realize a product with constant quality and moreover to make it a more valuable fertiliser with higher nutrient concentrations. Possible leads for improvement are (1) choice of separation and filtering techniques and dosage of flocculants/coagulants to increase the effectivity of pre-treatment, (2) adaptation of the cleaning procedure to improve performance and increase durability of RO-membranes and (3) application of higher quality RO-membranes. Research and test programs in this context are currently being executed.

Cost effective technologies for realising a quality upgrade are scarce however. In general they come with high energy inputs. To be profitable cheap energy must be available, such as thermal energy from a biogas fuelled heat and power generator or energy recovered from ventilation air from a pig house.

RO-concentrate vs criteria of RENURE

RO-concentrate should desirably comply with the quality criteria for N containing fertilisers derived from manure processing, referred to as RENURE (Recovered Nitrogen from manURE), so it can be used above the ceiling of 170 kg N/ha/year as prescribed in the Nitrates Directive. The results of the monitoring campaign have shown that the RO-concentrates (and also the RO-inputs) of all pilot installations meet the RENURE criteria on TOC/Total-N, Cu, Hg and Zn as proposed by JRC (Huygens et al., 2020). This proves that the RENURE criteria are easier to meet than the pilot MC criteria. The generated data from the monitoring campaign on the composition of RO-concentrate could support establishing the final RENURE criteria.

The study of JRC shows that apart from RO-concentrate scrubbing salts also can meet the RENURE criteria. The study also concludes that no criteria on contaminations of emerging concern (CEC) e.g. antibiotic residues and veterinary drugs, are necessary because the proposed criteria on TOC:Total-N or TAN:Total-N will effectively limit the CEC levels in RENURE N fertilisers.

RO-permeate vs requirements for discharge to surface water

RO-permeate, representing approx. 50% of the original volume of the incoming pig slurry, is the end product of RO without fertiliser value. This water is preferably discharged to surface water. To qualify for discharge to surface water the permeate has to meet high quality standards, on classical parameters as well as on so called precautionary parameters, such as antibiotic residues and pathogens, to prevent damage to the aquatic environment and risk to human health.

Previous studies have revealed that reverse osmosis in combination with other techniques is suitable for producing an effluent that meets the requirements on classical parameters and pathogens (Hoeksma et al., 2011 and 2015; Hoeksma and Buissonjé, 2015).

Results from this monitoring campaign show that heavy metals and antibiotic residues are effectively but not completely removed from the RO-input. The concentration of some parameters is above the threshold for surface water (see section 3.5.5). Weather discharge of RO-permeate to the surface water is harmful to the water environment depends on the dilution and the resulting concentrations in

the receiving water. The competent authority will judge, based on the situation in question, if additional treatment is necessary to achieve an acceptable quality of the permeate.

Based on the indications from previous studies and expert judgement reverse osmosis is for the time being classified as best available technic (BAT) for producing dischargeable water (Hoeksma et al., 2021).

However, the results from this monitoring campaign show that the RO-permeate (after ion exchange) is not clean water as could be expected from RO filtration. Normally an RO filter, also used for making drinking water from seawater, holds back all ions; at most small gaseous molecules such as ammonia can pass through an RO membrane. The fact that in the RO-permeate of a majority of the pilot installations several parameters in detectable concentrations are found can possibly be explained by wear or damage of the RO membranes. The presence of the observed parameters in the permeate could indicate suboptimal pre-treatment of the RO-input, deficient construction of the RO-unit and inadequate cleaning procedure of the RO membranes (Schwinge et al., 2004). All are critical success factors for manufacturing high quality RO-concentrate and a 'clean' RO-permeate.

The following is recommended:

- improve pre-treatment technology and operation,
- aim for a constant and 'clean' polished RO-input, to minimise fouling and damage and to extend the durability of the RO-membranes,
- carefully choose the right type and dosage of flocculants and supporting additives, in tune with the raw slurry, to improve the first separation step; the use of PAM only is discouraged,
- optimize the cleaning procedure of the RO-unit, to improve the performance RO-membranes,
- use high quality RO-membranes that are designed for this input from animal manure.

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Appendices

Appendix 1 Composition of feed stock

Table A1.1 Average concentrations of general parameters in the feed stock of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). The number between brackets represents the number of measurements.

Parameter	Unit	A		B		C		D		E		F		G		H		I	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
TS	g/kg	86 ^c	(5)	30 ^a	(3)	36 ^{ab}	(3)	58 ^{abc}	(3)	37 ^a	(4)	85 ^c	(3)	76 ^c	(5)	68 ^{bc}	(5)	55 ^{abc}	(3)
VS	g/kg dm	706 ^c	(5)	634 ^{bc}	(3)	470 ^a	(3)	664 ^c	(3)	554 ^{ab}	(4)	703 ^c	(3)	712 ^c	(5)	695 ^c	(5)	676 ^c	(3)
C-total	g/kg dm	364 ^c	(4)	341 ^{bc}	(3)	252 ^a	(3)	334 ^{bc}	(3)	309 ^b	(3/3)	352 ^{bc}	(3)	367 ^c	(5)	359 ^c	(5)	344 ^{bc}	(3)
TOC	mg/kg dm	40644 ^a	(5)	42824 ^a	(3)	82967 ^b	(3)	35269 ^a	(3)	42792 ^a	(4)	46008 ^a	(3)	34370 ^a	(5)	35844 ^a	(6)	48020 ^{ab}	(3)
Total-N	mg/kg dm	75134 ^a	(5)	94804 ^a	(3)	149109 ^b	(3)	85806 ^a	(3)	100464 ^a	(4)	71811 ^a	(3)	72004 ^a	(5)	74735 ^a	(6)	95084 ^a	(3)
TAN	mg/kg dm	43297 ^a	(5)	55366 ^a	(3)	109260 ^b	(3)	48510 ^a	(3)	64468 ^a	(4)	41614 ^a	(3)	39807 ^a	(5)	41914 ^a	(5)	58631 ^a	(3)
P	mg/kg dm	20042 ^a	(5)	24883 ^a	(3)	22551 ^a	(3)	22796 ^a	(3)	23818 ^a	(4)	18799 ^a	(3)	22104 ^a	(5)	21231 ^a	(6)	22819 ^a	(3)
P-PO ₄	mg/kg dm	2817 ^a	(5)	6761 ^c	(3)	6042 ^{bc}	(3)	2743 ^a	(3)	4323 ^{ab}	(4)	3176 ^a	(3)	3049 ^a	(5)	3555 ^a	(6)	3780 ^{ab}	(3)
K	mg/kg dm	54374 ^a	(5)	54694 ^a	(3)	138410 ^e	(3)	70079 ^{cd}	(3)	106277 ^{de}	(4)	55895 ^{ab}	(3)	50870 ^a	(5)	57796 ^{abc}	(6)	65903 ^{bc}	(3)
EC	mS/cm	29 ^c	(5)	17 ^a	(3)	24 ^b	(3)	25 ^{bc}	(3)	16 ^a	(4)	26 ^{bc}	(3)	25 ^b	(5)	24 ^b	(5)	27 ^{bc}	(3)
pH		7.8 ^{bc}	(5)	7.5 ^a	(3)	8.2 ^d	(3)	7.8 ^{bc}	(3)	7.7 ^{ab}	(4)	7.9 ^c	(3)	7.7 ^{bc}	(5)	7.7 ^b	(5)	7.8 ^{bc}	(3)
NO ₂ -N+NO ₃ -N	mg/kg dm	0 ^a	(5)	0 ^a	(3)	17 ^a	(3)	5.7 ^a	(3)	13 ^a	(4)	3.4 ^a	(3)	2.7 ^a	(5)	6.4 ^a	(5)	0 ^a	(3)
Ca	mg/kg dm	24204 ^a	(5)	35665 ^c	(3)	26540 ^{ab}	(3)	24613 ^a	(3)	75279 ^d	(4)	29983 ^{bc}	(3)	29118 ^{bc}	(5)	29940 ^{bc}	(6)	29389 ^{bc}	(3)
Mg	mg/kg dm	14958 ^a	(5)	16930 ^a	(3)	13360 ^a	(3)	14473 ^a	(3)	17170 ^a	(4)	15669 ^a	(3)	15076 ^a	(5)	15515 ^a	(6)	15820 ^a	(3)
Na	mg/kg dm	13370 ^a	(5)	22410 ^a	(3)	44033 ^b	(3)	17317 ^a	(3)	26960 ^{ab}	(4)	14879 ^a	(3)	13567 ^a	(5)	16176 ^a	(5)	20128 ^a	(3)
S	mg/kg dm	11310 ^{abc}	(5)	8457 ^a	(3)	12684 ^{abc}	(3)	24489 ^d	(3)	13255 ^{bc}	(4)	10066 ^{ab}	(3)	13638 ^c	(5)	11291 ^{abc}	(6)	12002 ^{abc}	(3)
Cl-	mg/kg dm	23446 ^{ab}	(5)	41917 ^{bc}	(3)	50070 ^c	(3)	27624 ^{ab}	(3)	27556 ^{ab}	(4)	28579 ^{ab}	(3)	22117 ^a	(5)	24032 ^{ab}	(6)	34079 ^{abc}	(3)

Table A1.2 Average concentrations of heavy metals (mg/kg dm) in the feed stock of pilot installations A – I. Values > 1 mg/kg are shown in bold. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). Positive values are presented in bold. For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of measurements. Values < detection limit are presented in mg/kg.

	A		B		C		D		E		F		G		H		I	
	Mean	n	Mean	n	Mean	n												
As	<0.11 ^a	(0/5)	<0.11 ^a	(0/3)	0.50 ^a	(1/3)	<0.11 ^a	(0/2)	<0.11 ^a	(0/3)	<0.11 ^a	(0/3)	<0.11 ^a	(0/3)	<0.11 ^{ab}	(0/6)	<0.11 ^a	(0/2)
Ba	47 ^a	(5/5)	46 ^a	(3/3)	133 ^b	(3/3)	52 ^{ab}	(2/2)	39 ^a	(3/3)	38 ^a	(3/3)	38 ^a	(3/3)	48 ^a	(6/6)	40 ^a	(2/2)
Cd	0.49 ^{ab}	(5/5)	<0.03 ^a	(0/3)	1.1 ^b	(2/3)	0.64 ^{ab}	(2/2)	0.20 ^a	(1/3)	0.50 ^{ab}	(3/3)	0.49 ^{ab}	(3/3)	0.53 ^{ab}	(6/6)	0.27 ^{ab}	(1/2)
Cr	9.0 ^{ab}	(5/5)	14 ^{bc}	(3/3)	18 ^c	(3/3)	8.3 ^{ab}	(2/2)	2.2 ^a	(1/3)	10 ^{abc}	(3/3)	9.7 ^{abc}	(3/3)	11 ^{bc}	(6/6)	9.1 ^{abc}	(2/2)
Cu	269 ^a	(5/5)	347 ^a	(3/3)	763 ^b	(3/3)	431 ^{ab}	(2/2)	131 ^a	(3/3)	473 ^{ab}	(3/3)	245 ^a	(3/3)	290 ^a	(6/6)	445 ^{ab}	(2/2)
Hg	<0.01 ^a	(0/5)	<0.01 ^a	(0/3)	<0.01 ^a	(0/3)	<0.01 ^a	(0/2)	<0.01 ^a	(0/3)	<0.01 ^a	(0/3)	<0.01 ^a	(0/3)	<0.01 ^a	(0/6)	<0.01 ^a	(0/2)
Ni	9.5 ^a	(5/5)	11 ^a	(3/3)	33 ^b	(3/3)	13 ^a	(2/2)	8.5 ^a	(3/3)	12 ^a	(3/3)	10 ^a	(3/3)	10 ^a	(6/6)	10 ^a	(2/2)
Pb	2.5 ^{ab}	(5/5)	1.9 ^{ab}	(3/3)	3.1 ^{ab}	(2/3)	1.8 ^{ab}	(2/2)	0.39 ^a	(1/3)	9.6 ^b	(3/3)	1.1 ^{ab}	(3/3)	1.3 ^a	(6/6)	1.6 ^{ab}	(2/2)
Zn	935 ^a	(5/5)	1121 ^a	(3/3)	2791 ^b	(3/3)	1364 ^{ab}	(2/2)	424 ^a	(3/3)	944 ^a	(3/3)	851 ^a	(3/3)	975 ^a	(6/6)	1143 ^{ab}	(2/2)

Table A1.3 Average concentrations of antibiotics ($\mu\text{g}/\text{kg dm}$) in the feed stock of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). Values above detection limit are presented in bold. For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of values > detection limit and the total number of measurements. Values < detection limit are presented in $\mu\text{g}/\text{kg}$.

	A		B		C		D		E		F		G		H		I	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
Oxytetracycline	385 ^a	(3/3)	8112 ^b	(2/2)	2005 ^{ab}	(2/2)	1198 ^{ab}	(1/2)	269 ^a	(1/2)	801 ^a	(1/2)	872 ^{ab}	(2/2)	3474 ^{ab}	(5/5)	666 ^{ab}	(2/2)
Doxycycline	3075 ^a	(3/3)	11496 ^{ab}	(2/2)	13228 ^b	(2/2)	1725 ^{ab}	(2/2)	762 ^a	(2/2)	982 ^a	(1/2)	6826 ^{ab}	(2/2)	5512 ^{ab}	(5/5)	6314 ^{ab}	(2/2)
Sulfadiazine	160 ^a	(2/3)	<1.0 ^a	(0/2)	115 ^a	(2/2)	21 ^a	(1/2)	192 ^a	(1/2)	250 ^a	(1/2)	13 ^a	(1/2)	21 ^a	(3/5)	15 ^a	(1/2)
Sulfadimidine	<1.0	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/5)	<1.0	(0/2)
Trimethoprim	<1.7	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<1.3	(0/5)	<1.5	(0/2)
Ciprofloxacin	<5.7	(0/3)	<3.0	(0/2)	<3.0	(0/2)	<5.5	(0/2)	<1.5	(0/2)	<4.0	(0/2)	<1.5	(0/2)	<5.8	(0/5)	<2.0	(0/2)
Enrofloxacin	<4.7 ^a	(0/3)	<1.0 ^a	(0/2)	<2.0 ^a	(0/2)	<1.5 ^a	(0/2)	60 ^b	(1/2)	<3.5 ^a	(0/2)	<3.5 ^a	(0/2)	<6.0 ^a	(0/5)	<3.0 ^a	(0/2)
Flumequine	7.4 ^a	(1/3)	<1.0 ^a	(0/2)	24 ^a	(1/2)	<1.0 ^a	(0/1)	<1.0 ^a	(0/2)	7.0 ^a	(1/2)	<1.5 ^a	(0/2)	4.3 ^a	(1/5)	<1.5 ^a	(0/2)
Lincomycin	<1.7	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<1.5	(0/5)	<1.5	(0/2)
Tilmicosin	23 ^a	(1/3)	177 ^b	(2/2)	<10 ^a	(0/1)	<5.5 ^a	(0/2)	<5.0 ^a	(0/1)	<6.0 ^a	(0/2)	<7.5 ^a	(0/2)	<1.5 ^a	(0/3)	38 ^a	(1/2)
Florfenicol	<2.3	(0/3)	<1.0	(0/2)	<1.0	(0/2)	<3.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<2.0	(0/5)	<1.0	(0/2)

Appendix 2 Composition of RO-Concentrate

Table A2.1a Average concentrations of general parameters in the RO-concentrates of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

Parameter	Unit	A		B		C		D		E		F		G		H		I		n
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	
TS	g/kg	29 ^{bc}	(5)	22 ^{ab}	(3)	22 ^{ab}	(3)	22 ^{bcd}	(3)	17 ^a	(4)	35 ^{bcde}	(3)	44 ^{ef}	(5)	44 ^f	(6)	47 ^{ef}	(3)	
VS	g/kg dm	422 ^f	(5)	301 ^{cde}	(3)	357 ^{ef}	(3)	357 ^{bcd}	(3)	131 ^a	(4)	373 ^{ef}	(3)	354 ^{ef}	(5)	354 ^{ef}	(6)	291 ^{cde}	(3)	
C-total	g/kg dm	255 ^g	(4/4)	132 ^d	(3)	214 ^f	(3)	214 ^a	(3)	80 ^{ab}	(3)	221 ^f	(3)	77 ^a	(5)	77 ^a	(6)	66 ^a	(3)	
TOC	mg/kg dm	172128 ^e	(5)	145953 ^{de}	(3)	129682 ^{cd}	(3)	129682 ^a	(3)	56303 ^a	(4)	156516 ^{de}	(3)	80809 ^{ab}	(5)	80809 ^{ab}	(6)	74725 ^{ab}	(3)	
Total-N	mg/kg dm	249919 ^g	(5)	244966 ^g	(3)	202545 ^{def}	(3)	202545 ^{abcd}	(3)	181417 ^{abcd}	(4)	238159 ^{fg}	(3)	157466 ^{ab}	(5)	157466 ^a	(6)	145770 ^a	(3)	
TAN	mg/kg dm	195133 ^{fgh}	(5)	204146 ^{gh}	(3)	157866 ^{abcde}	(3)	157866 ^{ab}	(3)	157568 ^{abcde}	(4)	185679 ^{efgh}	(3)	141929 ^{abc}	(5)	141929 ^a	(6)	134871 ^a	(3)	
P	mg/kg dm	5796 ^f	(5)	1812 ^{cd}	(3)	7183 ^g	(3)	7183 ^{ab}	(3)	1693 ^{cd}	(4)	6957 ^g	(3)	646 ^{ab}	(5)	646 ^{ab}	(6)	662 ^{ab}	(3)	
P-PO ₄	mg/kg dm	3755 ^e	(5)	384 ^{ab}	(3)	4953 ^{fg}	(3)	4953 ^a	(3)	1033 ^{bc}	(4)	4986 ^g	(3)	148 ^a	(5)	148 ^a	(6)	267 ^a	(3)	
K	mg/kg dm	223131 ^{bc}	(5)	193570 ^{ab}	(3)	216063 ^{bc}	(3)	216063 ^{ab}	(3)	257608 ^{de}	(4)	242657 ^{cde}	(3)	175560 ^a	(5)	175560 ^a	(6)	161815 ^{bcd}	(3)	
EC	mS/cm	50 ^{cd}	(5)	51 ^{cde}	(3)	38 ^{ab}	(3)	38 ^{cde}	(3)	33 ^a	(4)	60 ^{cdefg}	(3)	62 ^{efgh}	(5)	62 ^h	(6)	62 ^{cefg}	(3)	
pH		8.4 ^e	(5)	7.8 ^{ab}	(3)	8.2 ^{de}	(3)	8.2 ^{abc}	(3)	7.8 ^{abc}	(4)	8.1 ^{cd}	(3)	7.7 ^a	(5)	7.7 ^a	(6)	7.7 ^a	(3)	
NO ₂ -N + NO ₃ -N	mg/kg dm	6.2 ^a	(5)	139 ^a	(3)	18 ^a	(3)	18 ^a	(3)	44 ^a	(4)	11 ^a	(3)	6.4 ^a	(5)	6.4 ^a	(6)	21 ^a	(3)	
Ca	mg/kg dm	4433 ^{abc}	(5)	9763 ^f	(3)	6584 ^{bcdef}	(3)	6584 ^{abcd}	(3)	9202 ^{ef}	(4)	3979 ^{abc}	(3)	6724 ^{bcdef}	(5)	6724 ^{def}	(6)	6337 ^{abcdef}	(3)	
Mg	mg/kg dm	1087 ^a	(5)	15142 ^g	(3)	1238 ^a	(3)	1238 ^{fg}	(3)	10281 ^{def}	(4)	572 ^a	(3)	9605 ^{de}	(5)	9605 ^{de}	(6)	5758 ^{bc}	(3)	
Na	mg/kg dm	58179 ^{bcd}	(5)	84157 ^{hi}	(3)	70805 ^{efg}	(3)	70805 ^{abc}	(3)	81571 ^{ghi}	(4)	59052 ^{bcde}	(3)	48104 ^a	(5)	48104 ^{ab}	(6)	87929 ⁱ	(3)	
S	mg/kg dm	16453 ^{ab}	(5)	4722 ^a	(3)	12045 ^{ab}	(3)	12045 ⁱ	(3)	109169 ^{de}	(4)	29749 ^b	(3)	146538 ^{ghi}	(5)	146538 ⁱ	(6)	153196 ^{hi}	(3)	
Cl-	mg/kg dm	106554 ^{abc}	(5)	313909 ^e	(3)	89444 ^{ab}	(3)	89444 ^{ab}	(3)	105027 ^{abc}	(4)	110082 ^{abcd}	(3)	81918 ^a	(5)	81918 ^a	(6)	83929 ^{ab}	(3)	

Table A2.1b Average concentrations of general parameters in the RO-concentrates of pilot installations J –Q. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

Parameter	Unit	J		K		L		M		N		O		P		Q	
		Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
TS	g/kg	31	bcd (3)	28	abc (4)	39	cde (4)	27	ab (4)	42	de (4)	24	ab (3)	33	bcd (4)	42	de (4)
VS	g/kg DM	217	bc (3)	366	ef (4)	190	ab (4)	309	de (4)	259	bcd (4)	242	bcd (3)	217	bc (4)	324	de (4)
C-total	g/kg DM	120	cd (3)	227	f (4)	78	ab (4)	181	e (4)	101	bc (4)	138	d (3)	56	a (4)	79	ab (4)
TOC	mg/kg DM	96216	bc (3)	154666	de (4)	73698	ab (4)	106611	bc (4)	96066	bc (4)	89830	ab (3)	59772	a (4)	99925	bc (4)
Total-N	mg/kg DM	197516	cde (3)	222621	efg (4)	161751	abc (4)	220769	efg (4)	192978	bcd (4)	247487	g (3)	168314	abcd (4)	172965	abcd (4)
TAN	mg/kg DM	175215	bdefg (3)	175329	defg (4)	145955	abcd (4)	182080	efgh (4)	171242	bcd (4)	211999	h (3)	147955	abcd (4)	158854	abcde (4)
P	mg/kg DM	1314	abc (3)	6590	fg (4)	882	abc (4)	3938	e (4)	1288	bc (4)	2518	d (3)	731	ab (4)	414	a (4)
P-PO ₄	mg/kg DM	297	a (3)	4242	ef (4)	364	a (4)	2048	d (4)	314	a (4)	1191	c (3)	255	a (4)	45	a (4)
K	mg/kg DM	261238	de (3)	235337	cd (4)	244565	cde (4)	271552	bc (4)	212526	cd (4)	231405	a (3)	233091	a (4)	167346	e (4)
EC	mS/cm	56	cdef (3)	49	bcd (4)	64	fgh (4)	52	cde (4)	69	gh (4)	49	bc (3)	55	cdef (4)	64	fgh (4)
pH		7.8	abc (3)	8.1	bcd (4)	7.9	abcd (4)	7.9	abc (4)	7.9	abcd (4)	8.0	abcd (3)	8.0	bcd (4)	7.8	ab (4)
NO ₂ -N + NO ₃ -N	mg/kg DM	13	a (3)	69	a (4)	27	a (4)	30	a (4)	25	a (4)	0	a (3)	1638	b (4)	16	a (4)
Ca	mg/kg DM	2313	a (3)	6938	cdef (4)	4590	abc (4)	6967	cdef (4)	5738	abcde (4)	9308	ef (3)	3214	ab (4)	5911	abcde (4)
Mg	mg/kg DM	2935	ab (3)	903	a (4)	7748	cd (4)	2024	a (4)	7874	cd (4)	8134	cde (3)	9327	de (4)	10553	ef (4)
Na	mg/kg DM	66877	def (3)	61563	cde (4)	73230	fgh (4)	65789	def (4)	57982	bcd (4)	79958	ghi (3)	63265	cdef (4)	67362	def (4)
S	mg/kg DM	94782	d (3)	14250	ab (4)	129901	fg (4)	11972	ab (4)	122896	ef (4)	68314	c (3)	137825	fgh (4)	140896	fghi (4)
Cl-	mg/kg DM	111209	abcd (3)	112683	bcd (4)	91329	ab (4)	127910	cd (4)	97899	abc (4)	140702	d (3)	104894	abc (4)	86610	ab (4)

Table A2.2 Average concentrations of heavy metals (mg/kg dm) in the RO-concentrates of pilot installations A –Q. Positive values are shown in bold. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). Positive values are presented in bold. For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of measurements. All the values < detection limit are presented in mg/kg.

	A		B		C		D		E		F		G		H		I	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
As	<0.11	(0/5)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/2)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/3)	<0.11	(0/6)	<0.11	(0/2)
Ba	7.1 ^c	(4/5)	<0.16	(0/3)	15 ^d	(3/3)	<0.16	(0/2)	<0.16	(0/3)	6.9 ^c	(3/3)	<0.16	(0/3)	<0.16	(0/6)	<0.16	(0/2)
Cd	<0.03	(0/5)	<0.03	(0/3)	<0.03	(0/3)	<0.03	(0/2)	<0.03	(0/3)	<0.03	(0/3)	<0.03	(0/3)	<0.03	(0/6)	<0.03	(0/2)
Cr	3.3 ^{ab}	(5/5)	1.4 ^a	(1/3)	6.4 ^b	(3/3)	<0.07	(0/2)	<0.07	(0/3)	2.9 ^{ab}	(3/3)	<0.07	(0/3)	0.20 ^a	(1/6)	<0.07	(0/2)
Cu	25 ^a	(5/5)	<0.57	(0/3)	92 ^b	(3/3)	24 ^a	(1/2)	<0.57	(0/3)	28 ^a	(3/3)	<0.57	(0/3)	<0.57	(0/6)	<0.57	(0/2)
Hg	<0.01	(0/5)	<0.01	(0/3)	<0.01	(0/3)	<0.01	(0/2)	<0.01	(0/3)	<0.01	(0/3)	<0.01	(0/3)	<0.01	(0/6)	<0.01	(0/2)
Ni	16 ^f	(5/5)	11 ^{bcd}	(3/3)	28 ⁱ	(3/3)	12 ^{bcd}	(2/2)	9.4 ^b	(3/3)	17 ^{fgh}	(3/3)	10 ^{bc}	(3/3)	8.6 ^b	(5/6)	3.8 ^a	(1/2)
Pb	<0.01 ^a	(0/5)	<0.01	(0/3)	<0.01	(0/3)	0.34 ^{ab}	(1/2)	<0.01	(0/3)	0.17 ^{ab}	(1/3)	<0.01	(0/3)	<0.01	(0/6)	<0.01	(0/2)
Zn	63 ^a	(5/5)	<1.1	(0/3)	261 ^b	(3/3)	<1.1	(0/2)	<1.1	(0/3)	60 ^a	(3/3)	15 ^a	(1/3)	<1.1 ^a	(0/6)	<1.1	(0/2)

	J		K		L		M		N		O		P		Q	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
As	<0.11	(0/3)	<0.11	(0/2)	<0.11	(0/3)	<0.11	(0/4)	<0.11	(0/3)	<0.11	(0/2)	<0.11	(0/1)	<0.11	(0/2)
Ba	<0.16	(0/3)	8.5 ^c	(2/2)	<0.16 ^a	(0/3)	3.2 ^b	(2/4)	<0.16	(0/3)	<0.16	(0/2)	<0.16 ^a	(0/1)	<0.16	(0/2)
Cd	<0.03	(0/3)	<0.03	(0/2)	<0.03	(0/3)	<0.03	(0/4)	<0.03	(0/3)	<0.03	(0/2)	<0.03	(0/1)	<0.03	(0/2)
Cr	6.6 ^b	(2/3)	<0.07	(0/2)	<0.07 ^a	(0/3)	<0.07	(0/4)	<0.07	(0/3)	<0.07	(0/2)	2.8 ^{ab}	(1/1)	<0.07	(0/2)
Cu	<0.57	(0/3)	<0.57	(0/2)	15 ^a	(1/3)	<0.57	(0/4)	<0.57	(0/3)	<0.57	(0/2)	<0.57	(0/1)	<0.57	(0/2)
Hg	<0.01	(0/3)	<0.01	(0/2)	<0.01	(0/3)	<0.01	(0/4)	<0.01	(0/3)	<0.01	(0/2)	<0.01	(0/1)	<0.01	(0/2)
Ni	19 ^g	(3/3)	16 ^{ef}	(2/2)	13 ^{de}	(3/3)	16 ^f	(4/4)	13 ^{cde}	(3/3)	10 ^{bcd}	(2/2)	16 ^{efg}	(1/1)	10 ^{bc}	(2/2)
Pb	<0.01	(0/3)	<0.01	(0/2)	0.49 ^b	(1/3)	<0.01	(0/4)	<0.01	(0/3)	<0.01	(0/2)	<0.01	(0/1)	<0.01	(0/2)
Zn	11 ^a	(1/3)	45 ^a	(2/2)	<1.1	(0/3)	11 ^a	(1/4)	<1.1	(0/3)	<1.1	(0/2)	<1.1	(0/1)	<1.1	(0/2)

Table A2.3 Average concentrations of antibiotics ($\mu\text{g}/\text{kg dm}$) in the RO-concentrates of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). Positive values are presented in bold. For values < than detection limit the average detection limit is given. The numbers between brackets represent the number of positive values and the total number of measurements. Values < detection limit are presented in $\mu\text{g}/\text{kg}$.

	A		B		C		D		E		F		G		H		I	
	Mean	n																
Oxytetracycline	1515 ^e	(5/5)	1391 ^d	(1/1)	<15 ^a	(0/1)	<2.0 ^a	(0/2)	<5.5 ^a	(0/2)	1258 ^c	(2/2)	<10 ^a	(0/1)	29 ^b	(1/6)	<10 ^a	(0/1)
Doxycycline	3147 ^f	(5/5)	1597 ^d	(2/2)	3484 ^g	(2/2)	<3.0 ^a	(0/2)	<3.0 ^a	(0/2)	2749 ^e	(2/2)	157 ^c	(1/1)	59 ^b	(2/6)	83 ^b	(1/2)
Sulfadiazine	333 ^f	(4/5)	<1.0 ^a	(0/2)	274 ^e	(2/2)	82 ^c	(2/2)	265 ^e	(1/2)	126 ^d	(2/2)	<5.5 ^a	(0/2)	122 ^d	(5/6)	38 ^b	(1/2)
Sulfadimidine	<1.0	(0/5)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.2	(0/6)	<1.0	(0/2)
Trimethoprim	<1.4	(0/5)	<1.0	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<2.0	(0/2)	<1.5	(0/2)	<2.0	(0/6)	<2.0	(0/2)
Ciprofloxacin	<4.4	(0/5)	<1.5	(0/2)	<3.0	(0/2)	<3.0	(0/2)	<1.0	(0/2)	<7.5	(0/2)	<1.5	(0/2)	<5.2	(0/6)	<5.5	(0/2)
Enrofloxacin	<5.0	(0/5)	<1.0	(0/2)	<1.5	(0/2)	<1.0	(0/2)	<1.5	(0/2)	<6.0	(0/2)	<3.5	(0/2)	<6.5	(0/6)	<6.0	(0/2)
Flumequine	13 ^a	(1/5)	<1.0 ^a	(0/2)	<1.0 ^a	(0/2)	<1.0 ^a	(0/1)	<1.5 ^a	(0/2)	14 ^a	(1/2)	<1.5 ^a	(0/2)	<2.3 ^a	(0/6)	<1.5 ^a	(0/2)
Lincomycin	17 ^a	(1/5)	<1.0 ^a	(0/2)	<1.0 ^a	(0/1)	<1.5 ^a	(0/2)	<2.2 ^a	(0/6)	<1.5 ^a	(0/2)						
Tilmicosin	17 ^a	(1/5)	<10 ^a	(0/1)	<10 ^a	(0/2)	<5.5 ^a	(0/2)	<7.5 ^a	(0/2)	<6.0 ^a	(0/2)	<7.5 ^a	(0/2)	<3.5 ^a	(0/6)	38 ^a	(1/2)
Florfenicol	<1.8	(0/5)	<1.0	(0/2)	<1.0	(0/2)	<3.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<1.0	(0/2)	<2.7	(0/6)	<1.0	(0/2)

Appendix 3 Composition of process flows

Table A3.1 Average concentrations of general parameters in 5 process flows of pilot installations A – I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between installations for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements.

	Unit	Raw slurry			Solid fraction		RO-input		RO-concentrate		RO Permeaat	
		Mean		n	Mean	n	Mean	n	Mean	n	Mean	n
TS	g/kg	61	^a	(35)	283	^b (35)	16	^c (35)	35	^{ac} (35)	NA	0
VS	g/kg DM	654	^a	(35)	753	^b (35)	319	^{cd} (35)	296	^d (35)	NA	0
C-total	g/kg DM	340	^a	(33)	378	^b (33)	127	^c (33)	123	^c (33)	136	^c (33)
TOC	mg/kg DM	43845	^b	(35)	10232	^a (35)	90655	^c (35)	100653	^c (35)	NA	0
Total-N	mg/kg DM	88267	^c	(35)	43457	^b (35)	188132	^d (35)	192839	^d (35)	NA	0
TAN	mg/kg DM	53621	^b	(35)	10050	^a (35)	160253	^c (35)	163976	^c (35)	NA	0
P	mg/kg DM	21991	^c	(35)	25222	^d (35)	3712	^b (35)	2473	^b (35)	NA	0
P-PO ₄	mg/kg DM	3880	^a	(35)	1236	^b (35)	2080	^c (35)	1383	^{bc} (35)	NA	0
K	mg/kg DM	70449	^b	(35)	14691	^a (35)	201273	^c (35)	216159	^c (35)	NA	0
EC	mS/cm	24	^a	(35)	4.2	^b (35)	28	^a (35)	56	^c (35)	2.7	^b (35)
pH		7.8	^{ac}	(35)	8.4	^b (35)	7.8	^c (35)	7.9	^b (35)	7.5	^a (35)
NO ₂ -N + NO ₃ -N	mg/kg DM	5.3	^a	(35)	27	^a (35)	167	^a (35)	128	^a (35)	NA	0
Ca	mg/kg DM	34000	^a	(35)	32157	^a (35)	10308	^b (35)	6214	^b (35)	NA	0
Mg	mg/kg DM	15447	^a	(35)	16187	^a (35)	8730	^b (35)	6878	^b (35)	NA	0
Na	mg/kg DM	19991	^b	(35)	3550	^a (35)	64012	^c (35)	64974	^c (35)	NA	0
S	mg/kg DM	12862	^a	(35)	14498	^a (35)	95477	^b (35)	92243	^b (35)	NA	0
Cl-	mg/kg DM	29559	^b	(35)	4088	^a (35)	111537	^c (35)	109977	^c (35)	NA	0

Table A3.2 Average concentrations of heavy metals (mg/kg dm) in 5 process flows of pilot installations A - I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between process flows for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements. Values < detection limit are presented in mg/kg.

	Raw slurry		Solid fraction		RO-input		RO-concentrate		RO Permeaat	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
Cd	0.47 ^a	(23/30)	0.71 ^b	(29/30)	<0.03	(0/30)	<0.03	(0/50)	NA	(1/30)
Hg	<0.01	(0/30)	<0.03	(0/30)	<0.01	(0/30)	<0.01	(0/50)	NA	(0/30)
Pb	2.6 ^a	(27/30)	2.0 ^{ab}	(29/30)	<0.01	(0/30)	0.05 ^b	(3/50)	NA	(2/30)
As	0.05 ^{ab}	(1/30)	0.19 ^a	(5/30)	<0.11	(0/30)	<0.11	(0/50)	NA	(0/30)
Cr	10 ^b	(28/30)	12 ^b	(29/30)	0.59 ^a	(2/30)	1.4 ^a	(16/50)	NA	(1/30)
Ni	13 ^a	(30/30)	12 ^a	(30/30)	13 ^a	(26/30)	14 ^a	(48/50)	NA	(3/30)
Cu	360 ^a	(30/30)	449 ^a	(29/30)	7.7 ^b	(2/30)	12 ^b	(13/50)	NA	(1/30)
Zn	1136 ^a	(30/30)	1299 ^a	(29/30)	35 ^b	(7/30)	30 ^b	(16/50)	NA	(1/30)
Ba	53 ^a	(30/30)	61 ^a	(29/30)	1.8 ^b	(4/30)	2.6 ^b	(14/50)	NA	(2/30)

Table A3.3 Average concentrations of antibiotics ($\mu\text{g}/\text{kg dm}$) in 5 process flows of pilot installations A - I. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between process flows for the designated parameter ($p < 0.05$). The value between brackets represents the number of measurements. Values < detection limit are presented in $\mu\text{g}/\text{kg}$.

	Raw slurry		Solid fraction		RO-input		RO-concentrate		RO Permeaat	
	Mean	n	Mean	n	Mean	n	Mean	n	Mean	n
Oxytetracycline	2043 ^b	(19/25)	2110 ^b	(23/27)	2373 ^b	(9/28)	507 ^a	(9/25)	NA	(1/29)
Doxycycline	5426	(25/26)	4657	(25/25)	4902	(15/29)	1245	(18/28)	NA	(1/29)
Sulfadiazine	85 ^{abc}	(16/26)	18 ^c	(19/27)	206 ^b	(17/29)	221 ^{abc}	(21/29)	NA	(1/29)
Sulfadimidine	<1.0	(0/26)	<1.2	(0/27)	<1.0	(0/29)	<1.1	(0/29)	NA	(0/29)
Trimethoprim	0	(1/26)	<1.8	(0/27)	<1.2	(0/29)	<1.6	(0/29)	NA	(0/29)
Ciprofloxacin	<3.9	(0/26)	<13	(0/27)	<2.7	(0/29)	<3.9	(0/29)	NA	(0/27)
Enrofloxacin	5.7 ^a	(1/26)	<20 ^a	(0/27)	<3.4 ^a	(0/29)	<4.4 ^a	(0/29)	NA	(2/25)
Flumequine	5.0 ^a	(4/25)	3.4 ^a	(4/25)	4.6 ^a	(1/29)	3.5 ^a	(2/28)	NA	(0/29)
Lincomycine	<1.3 ^a	(0/26)	0.91 ^a	(1/25)	<1.3 ^a	(0/28)	3.3 ^a	(1/28)	NA	(1/29)
Tilmicosine	29 ^a	(4/22)	21 ^a	(10/26)	13 ^a	(2/26)	6.2 ^a	(2/28)	NA	(2/18)
Florfenicol	<1.6	(0/26)	<1.4	(0/27)	<1.3	(0/29)	<1.7	(0/29)	NA	(0/29)

Appendix 4 Comparison of type 1 and type 2 process

Table A4.1 Average concentrations of measured parameters in raw slurry and in RO-concentrate of type 1 and type 2 installations. Meaning of letters in superscript: different letters per line means that there is a significant difference in concentration between membrane filtration types for the designated parameter ($p < 0.05$). A single value between brackets represents the number of measurements, the double values between brackets represent the number of measurements > detection limit and the total number of measurements. Values of heavy metals < detection limit are presented in mg/kg, values of antibiotics < detection limit are presented in µg/kg.

	Unit	Raw slurry				RO-concentrate				
		Type 1		Type 2		Type 1		Type 2		
		Mean	n	Mean	n	Mean	n	Mean	n	
General parameters										
TS	g/kg	56 ^a	(24)	72 ^a	(11)	37 ^a	(46)	28 ^b	(19)	
VS	g/kg DM	660 ^a	(24)	641 ^a	(11)	270 ^a	(46)	368 ^b	(19)	
C-total	g/kg DM	346 ^a	(23)	341 ^a	(9)	84 ^a	(45)	220 ^b	(18)	
TOC	mg/kg DM	39155 ^a	(24)	53650 ^b	(11)	82133 ^a	(46)	145492 ^b	(19)	
Total-N	mg/kg DM	85332 ^a	(24)	94403 ^b	(11)	178304 ^a	(46)	228698 ^b	(19)	
TAN	mg/kg DM	50174 ^a	(24)	60828 ^b	(11)	157290 ^a	(46)	180168 ^b	(19)	
P-total	mg/kg DM	22758 ^a	(24)	20387 ^a	(11)	1027 ^a	(46)	5974 ^b	(19)	
P-PO ₄	mg/kg DM	3920 ^a	(24)	3795 ^a	(11)	351 ^a	(46)	3882 ^b	(19)	
K	mg/kg DM	66977 ^a	(24)	77708 ^b	(11)	207195 ^a	(46)	237862 ^b	(19)	
EC	mS	23 ^a	(24)	27 ^a	(11)	59 ^a	(46)	50 ^b	(19)	
pH		7.7 ^a	(24)	7.9 ^b	(11)	7.8 ^a	(46)	8.1 ^b	(19)	
NO ₂ -N + NO ₃ -N	mg/kg DM	5.1 ^a	(24)	5.7 ^a	(11)	170 ^b	(46)	27 ^a	(19)	
Ca	mg/kg DM	37626 ^b	(24)	26417 ^a	(11)	6357 ^a	(46)	5762 ^a	(19)	
Mg	mg/kg DM	15796 ^a	(24)	14716 ^a	(11)	9228 ^b	(46)	1188 ^a	(19)	
Na	mg/kg DM	19329 ^a	(24)	23207 ^a	(11)	65945 ^b	(46)	62625 ^a	(19)	
S	mg/kg DM	13587 ^a	(24)	11345 ^a	(11)	121877 ^a	(46)	16449 ^b	(19)	
Cl-	mg/kg DM	28340 ^a	(24)	32107 ^b	(11)	109887 ^a	(46)	110196 ^a	(19)	
Heavy metals										
Cd	mg/kg DM	0.36 ^a	(13/19)	0.65 ^a	(10/11)	<0.03 ^a	(0/33)	<0.03 ^a	(0/17)	
Hg	mg/kg DM	<0.01 ^a	(0/19)	<0.01 ^a	(0/11)	<0.01 ^a	(0/33)	<0.01 ^a	(0/17)	

		Raw slurry				RO-concentrate							
Pb	mg/kg DM	1.3	^a	(17/19)	4.6	^b	(10/11)	0.06	^a	(2/33)	0.03	^a	(1/17)
As	mg/kg DM	<0.11	^a	(0/19)	0.14	^a	(1/11)	<0.11	^a	(0/33)	<0.11	^a	(0/17)
Cr	mg/kg DM	9.2	^a	(17/19)	12	^a	(11/11)	0.85	^a	(5/33)	2.6	^a	(11/17)
Ni	mg/kg DM	10	^a	(19/19)	17	^a	(11/11)	11	^a	(31/33)	18	^b	(17/17)
Cu	mg/kg DM	299	^a	(19/19)	459	^a	(11/11)	2.9	^a	(2/33)	29	^b	(11/17)
Zn	mg/kg DM	949	^a	(19/19)	1443	^a	(11/11)	2.3	^a	(2/33)	83	^b	(14/17)
Ba	mg/kg DM	44	^a	(19/19)	68	^a	(11/11)	<0.16	^a	(0/33)	8.1	^a	(13/17)
Antibiotics													
Oxytetracycline	µg/kg DM	2581	^b	(13/19)	48	^a	(6/7)	104	^a	(2/18)	1261	^b	(7/9)
Doxycycline	µg/kg DM	5449	^a	(19/19)	328	^a	(6/7)	245	^a	(9/18)	3134	^b	(9/9)
Sulfadiazine	µg/kg DM	41	^a	(11/19)	11	^b	(5/7)	195	^a	(13/18)	274	^a	(8/9)
Sulfadimidine	µg/kg DM	<1.0	^a	(0/19)	<1	^a	(0/7)	<1.1	^a	(0/18)	<1.0	^a	(0/9)
Trimethoprim	µg/kg DM	0	^a	(1/19)	<1.4	^a	(0/7)	<1.6	^a	(0/18)	<1.4	^a	(0/9)
Ciprofloxacin	µg/kg DM	<3.6	^a	(0/19)	<4.4	^a	(0/7)	<3.4	^a	(0/18)	<4.8	^a	(0/9)
Enrofloxacin	µg/kg DM	8.6	^b	(1/19)	<3.6	^a	(0/7)	<4.4	^a	(0/18)	<4.4	^a	(0/9)
Flumequine	µg/kg DM	1.3	^a	(1/19)	0.57	^b	(3/7)	<2.2	^a	(0/18)	10	^b	(2/9)
Lincomycin	µg/kg DM	<1.2	^a	(0/19)	<1.4	^a	(0/7)	<1.6	^a	(0/18)	11	^b	(1/9)
Tilmicosin	µg/kg DM	39	^b	(3/19)	1.0	^a	(1/7)	4.4	^a	(1/18)	9.6	^a	(1/9)
Florfenicol	µg/kg DM	<1.6	^a	(0/19)	<1.6	^a	(0/7)	<1.8	^a	(0/18)	<1.4	^a	(0/9)

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the potential
of nature to
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