Completion Report

For cVMS (D4, D5 & D6) Restriction Monitoring in EU Program

ERM Study Number: 0383878
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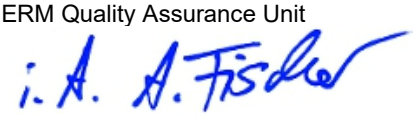

25th of April 2023

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25th of April 2023

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CONTENTS

List of Tables

List of Figures

Acronyms and Abbreviations

Name	Description
CoC	chain of custody
cVMS	cyclic volatile methyl siloxanes
D4	octamethylcyclotetrasiloxane
D4 ML	D4 mass loading
D5	decamethylcyclopentasiloxane
D5 ML	D5 mass loading
D6	dodecamethylcyclohexasiloxane
D6 ML	D6 mass loading
DE	Germany
DE1	WWTP Halle Nord (Halle an der Saale, Germany)
DE2	WWTP Wolfsburg-Brackstedt (Wolfsburg, DE)
DOC	dissolved organic carbon
EA	Environment Agency
ECHA	European Chemicals Agency
ERM	Environmental Resources Management
ERM GmbH	Environmental Resources Management Germany
ES	Spain
ES1	Lleida Wastewater Treatment Works (Lleida, Spain)
EU	European Union
GCMS	gas chromatography/mass spectrometry
GLP	good laboratory practice
HASP	health and safety plan
MDL	method detection limit
ML	mass loading
MOC	TOC/DOC matrix characterization parameters
MPC	measurement performance criterion
MS	mass spectrometry
MSS	TSS/VSS matrix characterization parameters
OC	organic carbon
PB	data qualifier: value > 0.5 * limit of quantification
PCA	Principal Component Analysis
PCP	personal care product
PL	Poland
PL1	Stalowa Wola Wastewater Treatment Works (Stalowa Wola, Poland)
QA	quality assurance
QAPP	quality assurance project plan
PCAMPCQC	quality control
RPD	relative percent difference
SE	Sweden
SE1	Norrköping Vatten och Avfall (Norrköping, Sweden)
SOP	standard operating procedure
TOC	total organic carbon
TSS	total suspended solids
UK	United Kingdom
UK1	Bury Wastewater Treatment Works (Bury, Greater Manchester, United Kingdom)
VSS	volatile suspended solids
WO	Wash-off personal care product
WWTP	wastewater treatment plant
yr	year
µg/mg-OC	micrograms per milligram organic carbon

1. INTRODUCTION

1.1 Context

The present document is the Completion Report of the cVMS (D4, D5 & D6) Restriction Monitoring in EU program. In the framework of the study, an Initial, Transitional and Termination Report have been prepared. The cVMS (D4, D5 & D6) Restriction Monitoring in EU program was terminated after the Transitional Period. This completion report is an abstract of the 0383878 - Termination Report from February 15th, 2022.

The Post-restriction Period was originally planned to run from Summer 2020 through Spring 2021. As the COVID-19 pandemic has clearly impacted the habits of many consumers, this would impede our ability to make meaningful comparisons between any new data collected in Period 3 (Post-restriction Period) with the results from Periods 1 and 2 (Initial and Transitional Period). The Study Sponsor has therefore decided to forgo the Post-restriction Period and to terminate the cVMS (D4, D5 & D6) Restriction Monitoring in EU program.

1.2 Content

This Completion Report contains:

- a summary of key findings of the Initial and Transitional Periods; and

The report does not contain any new data or interpretations compared to the Transitional and Termination Report.

2. PROJECT SUMMARY

ERM GmbH (ERM) was retained by Silicones Europe to conduct a monitoring study to assess the efficacy of the EU product use restriction of octamethylcyclotetrasiloxane (D4) and decamethylcyclopentasiloxane (D5) in wash-off personal care products (PCPs). The restriction on D4 and D5 in wash-off personal care products (PCPs) was published in the Official Journal on 11 January 2018 and is referred to as the “2018 Restriction.” The goal of the restriction was to reduce the concern for aquatic environments receiving discharges from WWTPs, while permitting other uses that do not pose a concern. At the request of the European Commission (EC), the European Chemical Agency (ECHA) proposed a new restriction for D4, D5, and dodecamethylcyclohexasiloxane (D6) in uses of leave-on personal care products and other consumer and professional products. The proposal also included a restriction of D6 in wash-off PCPs. The proposed restriction was published on 11 January 2019 and is referred to as the “2019 Proposed Restriction.”

A multi-year monitoring program was implemented to assess the efficacy of the 2018 Restriction. The program measured D4 and D5 influent concentrations at a variety of wastewater treatment plants (WWTPs). The first period of the study (Initial Period) was conducted from the Fall of 2017 through the Summer of 2018. The results of the Initial Period were reported in February of 2019. Following a laboratory validation and field pilot study, D6 was included in the study plan in the Transitional Period to evaluate the pre-restriction level of D6 in wastewater influent samples. The Transitional Period of the study was conducted from the Spring of 2019 through the Winter of 2020. The results are presented in the Transitional Report in October 2020.

The Post-restriction Period was originally planned to run from Summer 2020 through Spring 2021. In May 2020, the Study Sponsor decided to postpone the Post-restriction Period due to the COVID-19 pandemic. In April 2021, the Study Sponsor re-evaluated if the cVMS (D4, D5 & D6) Restriction Monitoring in EU program should continue and decided to forgo the Post-restriction Period and to terminate the study (as explained in section 1.1).

3. EXECUTIVE SUMMARY OF THE TRANSITIONAL REPORT

3.1 Most essential information

The main findings from the Initial and Transitional Periods are:

1. The materials, sampling procedures, and analytical methods detailed in the Study Plan (ERM 2017) and this report provide reliable monitoring results for D4, D5, and D6 in wastewater influent samples and modelled EU-wide mass loadings that can be used to assess the efficacy of the product use restrictions;
2. The D4 and D5 EU-wide mass loadings extrapolated from the monitoring results are well below the pre-restriction mass loadings presented in the 2018 Restriction[1];
3. The D4 EU-wide mass loadings extrapolated from the monitoring results are consistent with the post-restriction goal presented in the 2018 Restriction[2];
4. The D5 EU-wide mass loadings extrapolated from the monitoring results are approaching the post-restriction goal presented in the 2018 Restriction and fall within the range of the post-restriction goals presented in the 2019 Proposed Restriction[3], which includes other consumer and professional uses;
5. The distribution of the modelled D4 and D5 EU-wide mass loadings in the Transitional Period are statistically different than the Initial Period (i.e., lower and more tightly grouped); and
6. The D6 EU-wide mass loadings extrapolated from the monitoring results are well below the post-restriction goal presented in the 2019 Proposed Restriction.
7. It has to be noted, that from day 1 of the project that samples taken have always reflected both, wash-off PCPs as well as Leave-On PCPs since samples were taken at the intake to the WWTPs.

3.2 Methods and Procedures[4]

3.2.1 Study Design

The Study Plan calls for samples to be collected from six representative WWTPs across the European Union (EU) and the United Kingdom (UK) receiving wastewater from primarily residential sources (Table 3.1). However, in some unavoidable cases the selected WWTP also received parts of industrial effluents. Plants were selected in five countries to provide a broad geographical representation of the EU and the UK. Other selection criteria included size of service area, land use, demographics, separation of storm and sewer water, industrial and commercial activity, and willingness to participate in the study.

[1] EA, 2015a. Risk Management Options Analysis Report: Octamethylcyclotetrasiloxane.Table 1 “Summary of environmental release estimates for D4” – continental emission for “Personal care products – use”, Environment Agency June 2015
EA, 2015b. Risk Management Options Analysis Report: Decamethylcyclopentasiloxane. Table 1 “Summary of environmental release estimates for D5” – continental emission for “Personal care products – use”, Environment Agency June 2015.

[2] ECHA´s Annex XV Restriction Report, Proposal for a Restriction, Substance Name: octamethylcyclotetrasiloxane, Substance Name: decamethylcyclopentasiloxane, Version 1.1, section E.2.1.1.1.2 “Changes in the environmental risks/impacts”, June 2015.

[3] ECHA, 2019. Annex XV Restriction Report, Proposal for a Restriction, Substance Name: octamethylcyclotetrasiloxane, Substance Name: decamethylcyclopentasiloxane, Substance Name: dodecamethylcyclohexasiloxane, Table 14 “Tonnage and release estimates per use after restriction” and text below, Version 1.0, January 2019.

[4] This section is an executive summary extract from the Transitional Report (ERM, 23 October 2020).

Table 3.1 WWTPs selected for the monitoring study

Country	*Site Name and Location*
Germany	Wastewater Treatment Works Halle Nord (Halle an der Saale, DE)
Germany	Wolfsburg-Brackstedt (Wolfsburg, DE)
Spain	Lleida Wastewater Treatment Works (Lleida, ES)
Poland	Stalowa Wola Wastewater Treatment Works (Stalowa Wola, PL)
Sweden	Norrköping Vatten och Avfall (Norrköping, SE)
United Kingdom	Bury Wastewater Treatment Works (Bury, Greater Manchester, UK)

Plants were sampled eight times during each period, using a stratified random sampling design that considered potential seasonal, weekly, and diurnal factors. In addition to D4, D5, and D6 samples were analysed for temperature, conductivity, total and volatile suspended solids, and total and dissolved organic carbon. The mass loadings of D4, D5, and D6 to these plants are based on influent concentrations and flow measured at the time of sampling and reported on a per capita basis based on population data for each WWTP service area.

Statistical analyses were performed to assess the efficacy of the targeted D4 and D5 product use restriction (e.g., test of significant difference between the Initial and Transitional Periods). In addition, a probabilistic regression model was developed to account for regional, cultural, socio-economic, and demographic factors that may influence the mass loadings of PCPs. This model accounts for uncertainty and variability in the input parameters and generates a distribution of EU-wide mass loadings of D4, D5, and D6 to wastewater based on the monitoring results and available census data.

3.2.2 Quality Assurance/Quality Control Measures

The sampling of wastewater influent for D4, D5, and D6 presents some unique challenges from a health and safety and quality assurance/quality control (QA/QC) perspective. The Study Plan details the materials, sampling procedures, and analytical methods used to collect and analyse influent samples for D4, D5, and D6. The quality assurance project plan (QAPP) specifies QA/QC samples (e.g., field blanks, field duplicates, field spikes, lab procedural blanks, and calibration standards) and procedures to ensure the data collected are reliable (i.e., consistent, repeatable and credible). Following each sampling event, field documentation was reviewed for accuracy and completeness and the laboratory reports were reviewed to verify the results are consistent with the method performance criteria defined in the QAPP and are useable for the purposes of the study.

A health and safety plan was prepared for each sampling location to identify potential hazards (e.g., physical, chemical and biological) that may be encountered during sampling and best practices to ensure the safety of the workers involved in collecting and processing the samples. No reportable health and safety incidents were recorded during the first two periods of this study.

3.2.3 Laboratory Analytical Methods

After a rigorous pre-qualification process, Synlab (Rotterdam, NL) was selected as the laboratory for this program. Subsequently, a laboratory method was developed and reviewed by the ERM team and analytical chemists from Silicones Europe member companies. The method was verified by conducting a validation study, including the determination of the method detection limit, demonstration of clean procedural blank samples, and an inter-laboratory calibration study. Prior to the inclusion of D6 in the Transitional Period of this study, an additional method validation study was performed to verify D6 could be appropriately analysed using the analytical method previously established for D4 and D5. The method developed is the basis of the Analytical Standard ISO 20596-2 published in 2021.

3.2.4 Pilot Studies

Before the start of the Initial Period, a pilot study for D4 and D5 was conducted over a five-day period between July 13, 2017 and July 17, 2017 at the Bury, UK WWTP. Eight sampling events were conducted, including all combinations of weekday/weekend and time of day (morning, afternoon, evening, and night). The D4/D5 pilot study demonstrated that the Study Plan, including the experimental design and the sampling and analytical methods, could provide reliable data and accomplish the objectives of the D4 and D5 monitoring program.

Prior to adding D6 in the Transitional Period, another pilot study was conducted between September 28, 2018 and October 2, 2018 at the Bury, UK WWTP. The D6 pilot study was conducted in the same manner as the D4/D5 pilot study. The D6 pilot study verified that D6 could be collected and analysed in wastewater influent samples with a high level of confidence.

3.3 Initial and Transitional Period Results[5]

3.3.1 Initial Period Results

During the Initial Period, samples were collected from Fall 2017 through Summer 2018. Eight sampling events were completed at each of the six selected WWTPs. During each event, 24 samples were collected, including investigative, QA/QC, and retained samples. A total of 1,152 samples were collected during the Initial Period. Two samples for Total Suspended Solids (TSS) and Volatile Suspended Solids (VSS) were discarded inadvertently by the laboratory prior to analysis. All other planned analyses were completed. In addition to laboratory analyses, conductivity, temperature and influent flow were recorded during each sampling event.

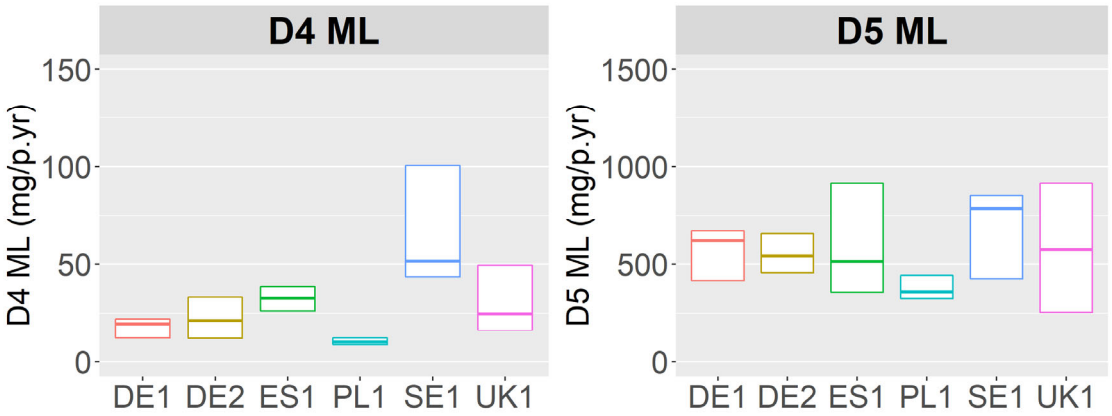
Analytical results were evaluated against method performance criteria and verified according to the QA/QC procedures specified in the QAPP. An outlier analysis was performed according to the procedure presented in the Study Plan. Samples greater than 1.5 times the inter-quartile range were identified as suspected outliers (Tukey 1977). In addition, suspected outliers were compared to the median value to account for narrow inter-quartile ranges. Samples with a relative percent difference between the suspected outlier and the median greater than 85% were confirmed as statistical outliers. This screening value corresponds to a difference of approximately three standard deviations, assuming a normal distribution and a relative standard deviation of 50%. This evaluation resulted in the exclusion of a single D5 result from Wolfsburg, DE. For each sampling event, the median of the measured results for each analyte was used for further evaluation.

Each WWTP was sampled eight times throughout the period resulting in 12 values for each season, 12 values for each time of day and 24 values for each day of week (i.e., weekday and weekend). For each sampling event, a per capita mass loading was calculated for D4 and D5, expressed in mg/person/year. These per capita mass loadings are based on the event-specific median concentration for D4 and D5, the influent flow rate, and the population served by the WWTP. Statistically significant differences were observed between some of the WWTPs, indicating the selected WWTPs captured a good spread of conditions throughout the EU and the UK (Figure 3.1). Additionally, statistically significant differences were observed in per capita mass loadings by time of day and season, reflecting different use patterns of PCPs throughout the day and year. No differences were detected between days of the week (i.e., weekdays and weekend).

Samples were stored by the laboratory for maximum 100 days.

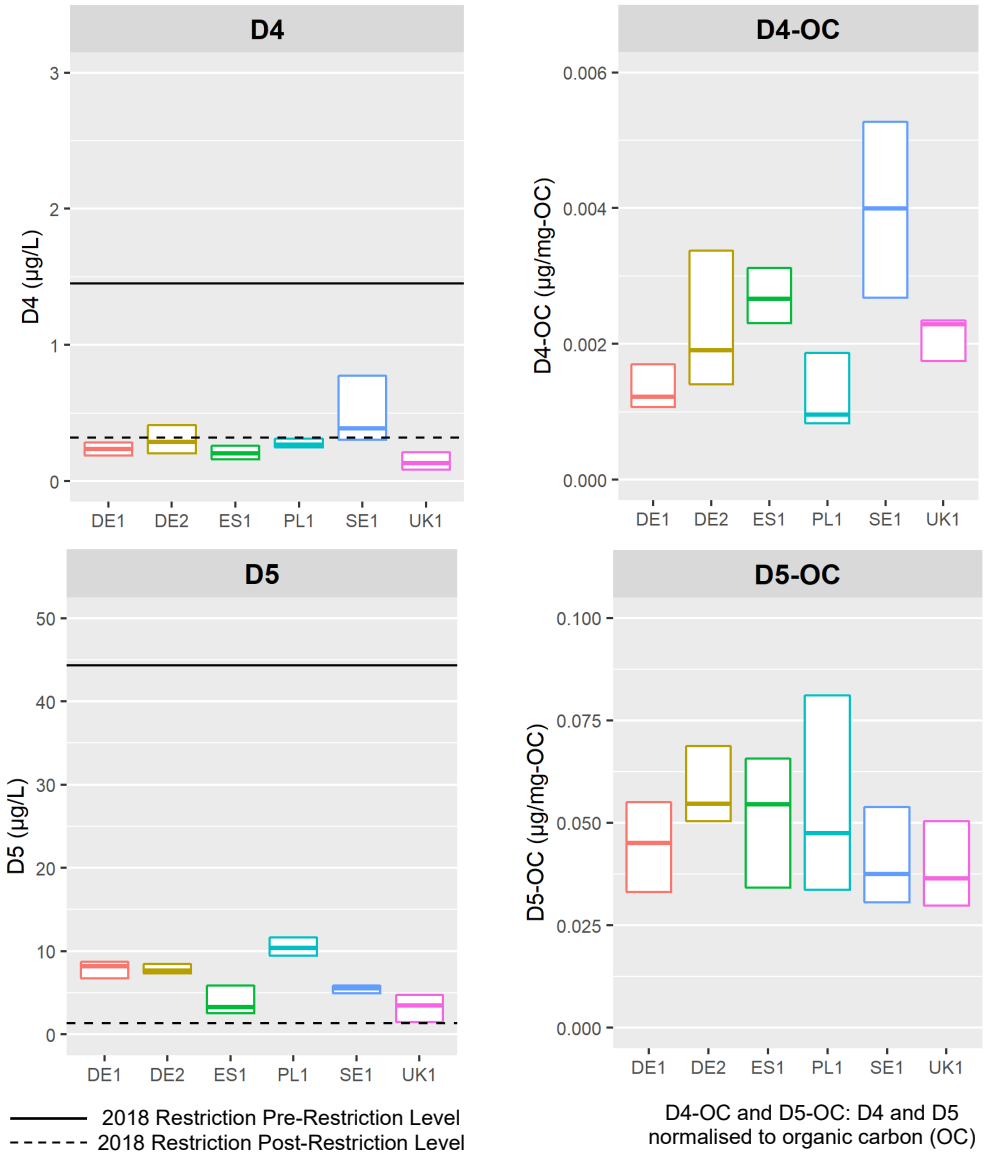
[5] This section is an executive summary extract from the Transitional Report (ERM, 23 October 2020).

Figure 3.1 Initial Period - D4/D5 Per Capita Mass Loadings (ML) by Location



DE1 – Halle, Germany **DE2** – Wolfsburg, Germany **ES1** – Lleida, Spain **PL1** – Stalowa Wola, Poland **SE1 -** Norrköping, Sweden **UK1** – Bury, United Kingdom

Figure 3.2 Initial Period - D4, D4-OC, D5, and D5-OC by Locations



DE1 – Halle, Germany **DE2** – Wolfsburg, Germany **ES1** – Lleida, Spain **PL1** – Stalowa Wola, Poland **SE1 -** Norrköping, Sweden **UK1** – Bury, United Kingdom

3.3.2 Transitional Period Results

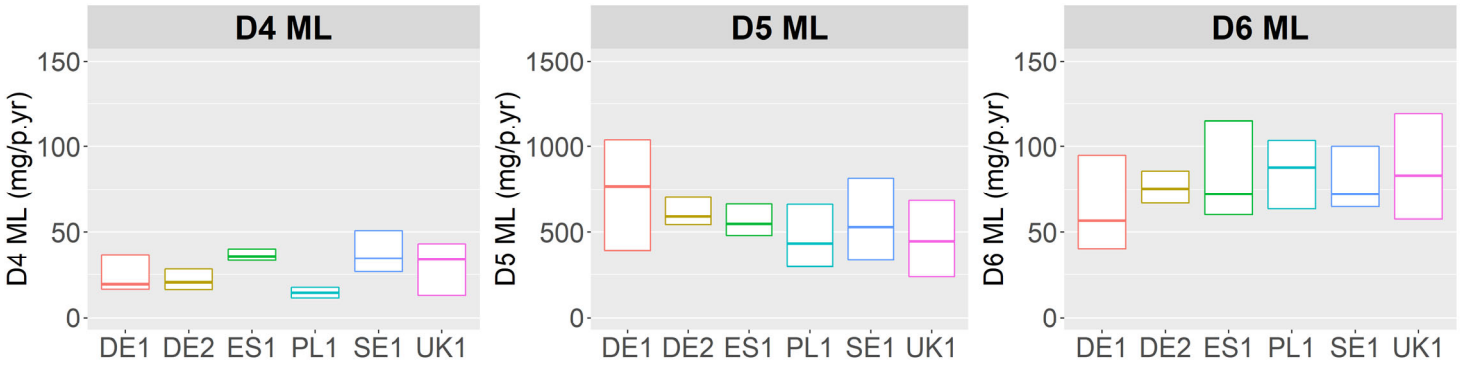
During the Transitional Period, samples were collected from Spring 2019 through Winter 2020. Eight sampling events were completed at each of the six WWTPs. During each event, 24 samples were collected, including investigative, QA/QC, and retained samples. A total of 1,152 samples were collected during the Transitional Period. Six samples for Total Organic Carbon (TOC) and Dissolved Organic Carbon (DOC) were discarded inadvertently by the laboratory prior to analysis and an erroneous result for TOC was reported by the laboratory. All other planned analyses were completed. In addition to laboratory analyses, conductivity, temperature and influent flow were recorded during each sampling event.

Analytical results were evaluated against method performance criteria and verified according to the QA/QC procedures specified in the QAPP. An updated outlier analysis, including the analysis of retained samples, was performed according to the amended Study Plan. This evaluation resulted in the exclusion of one D4, two D5, and one D6 result. For each event, the median of the measured results for each analyte was used for further evaluation.

Each WWTP was sampled eight times throughout the Transitional Period resulting in 12 values for each season, 12 values for each time of day and 24 values for each day of week. For each sampling event, a per capita mass loading was calculated for D4, D5 and D6, expressed in mg/person/year. These per capita mass loadings are based on the event-specific median concentration for D4, D5, and D6, the influent flow rate, and the population served by the WWTP (Figure 3.3). Statistical differences were observed in per capita mass loadings by time of day and between some locations. No differences were detected between days of the week and seasons.

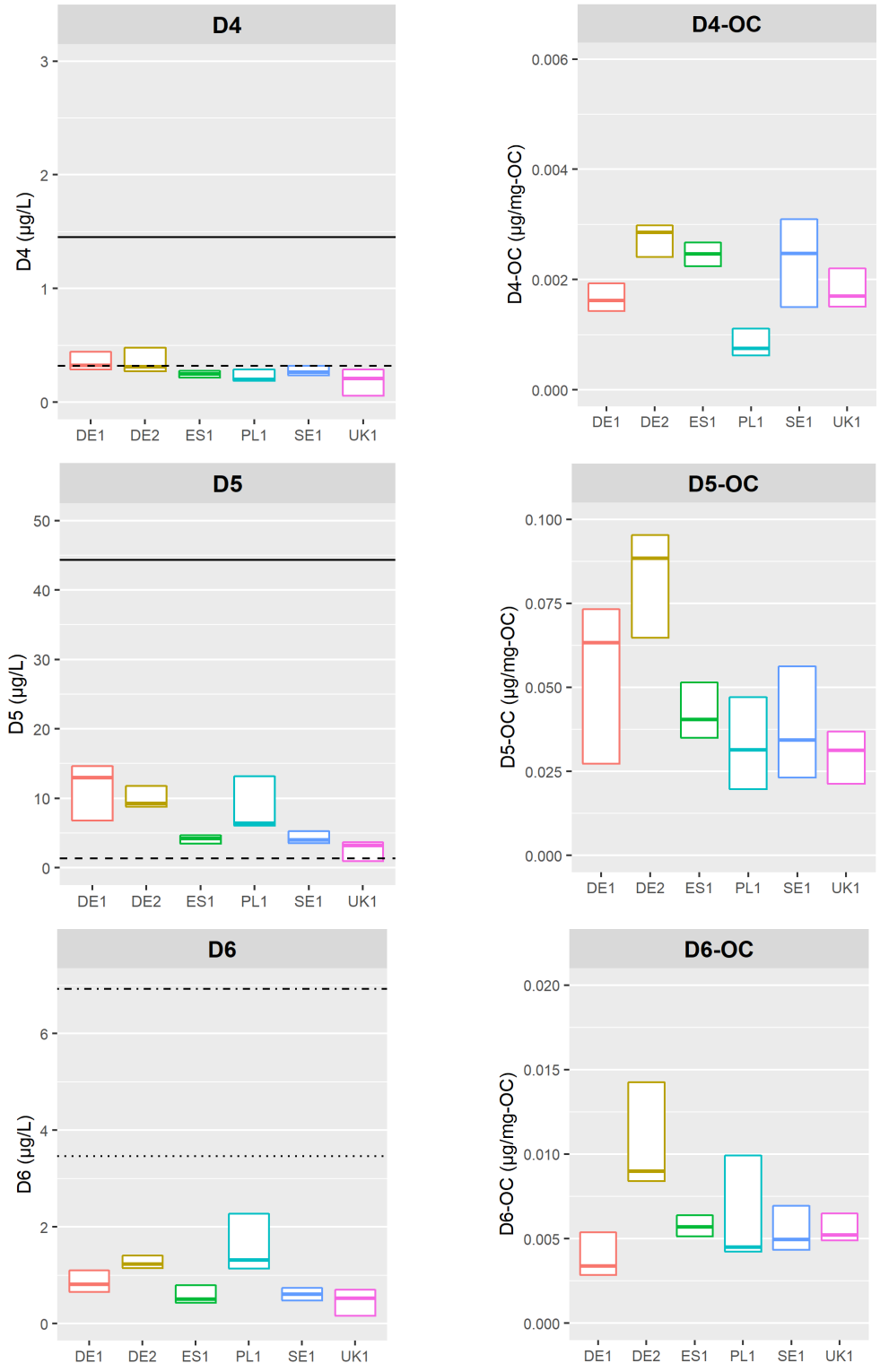
Samples were stored by the laboratory for maximum 100 days.

Figure 3.3 Transitional Period - D4, D5 and D6 Per Capita Mass Loadings (ML) by Location



DE1 – Halle, Germany **DE2** – Wolfsburg, Germany **ES1** – Lleida, Spain **PL1** – Stalowa Wola, Poland **SE1 -** Norrköping, Sweden **UK1** – Bury, United Kingdom

Figure 3.4 Transitional Period - D4, D4-OC, D5, D5-OC, D6, and D6-OC by Location



2018 Restriction Pre-Restriction Level
2018 Restriction Post-Restriction Level
2019 Restriction Pre-Restriction Level
2019 Restriction Post-Restriction Level

D4-OC, D5-OC and D6-OC: D4, D5 and D6 normalised to organic carbon (OC)

DE1 – Halle, Germany **DE2** – Wolfsburg, Germany **ES1** – Lleida, Spain **PL1** – Stalowa Wola, Poland **SE1** - Norrköping, Sweden **UK1** – Bury, United Kingdom

3.4 Correlation Analysis

The strength of association between the various analytes was evaluated. A low, positive correlation was observed between D4 and the matrix characterization parameters. This correlation was more pronounced for D5 and D6, where a moderate to high positive correlation was detected. To account for the relationship between D4, D5, and D6 and organic carbon, D4, D5, and D6 were normalized to TOC, resulting in the parameters D4-OC, D5-OC and D6-OC expressed in µg/mg-TOC.

3.5 Extrapolation to EU-wide Mass Loading to Wastewater

The per capita mass loadings measured at each WWTP were used to develop a probabilistic regression model to generate a distribution of the mass loadings for D4, D5, and D6 from all EU member states and the UK. The model was based on the relationship between per capita mass loadings of D4, D5, and D6 and regional, cultural, socio-economic, and demographic factors. Census data are available in the Eurostat database for more than 900 urban areas, defined as areas with populations greater than 10,000 people. Following each period, the most recent census data for each urban area was used to develop the model.

The Eurostat census data consists of 141 variables that capture a wide range of regional, cultural, socio-economic, and demographic factors (considered independent variables in the regression model). Each census variable was assessed for transformations to reduce outlier and non-linearity issues. Principal Component Analysis (PCA) was used to derive a reduced set of summary variables (known as principal components) that capture the majority of the information from all the census data. K-means cluster analysis was used to assess regional differences between urban areas and confirm that the sampled locations provided a good representation of the urban areas across the EU member states and the UK.

The regression model considered the uncertainty and variability of the input parameters using Monte Carlo analysis. The most likely (median), low-end (5th Percentile), and high-end (95th Percentile) mass loadings for D4, D5, and D6 determined from the monitoring results are presented in Table 3.2. The mass loadings determined in this study include D4, D5, and D6 from all sources to wastewater, including all remaining direct uses on the market and traces from other sources.

Table 3.2 Modelled EU-wide Mass Loadings for D4 and D5 in the Initial and Transitional Periods and D6 in the Transitional Period

Analyte	Period	Mass Loading (tonnes/yr)
D4	Initial	14.1 (10.3 – 21.1)
	Transitional	13.0 (10.5 – 17.6)
D5	Initial	288 (250 – 335)
	Transitional	271 (249 – 296)
D6	Transitional[6]	39.3 (36.7 – 42.2)

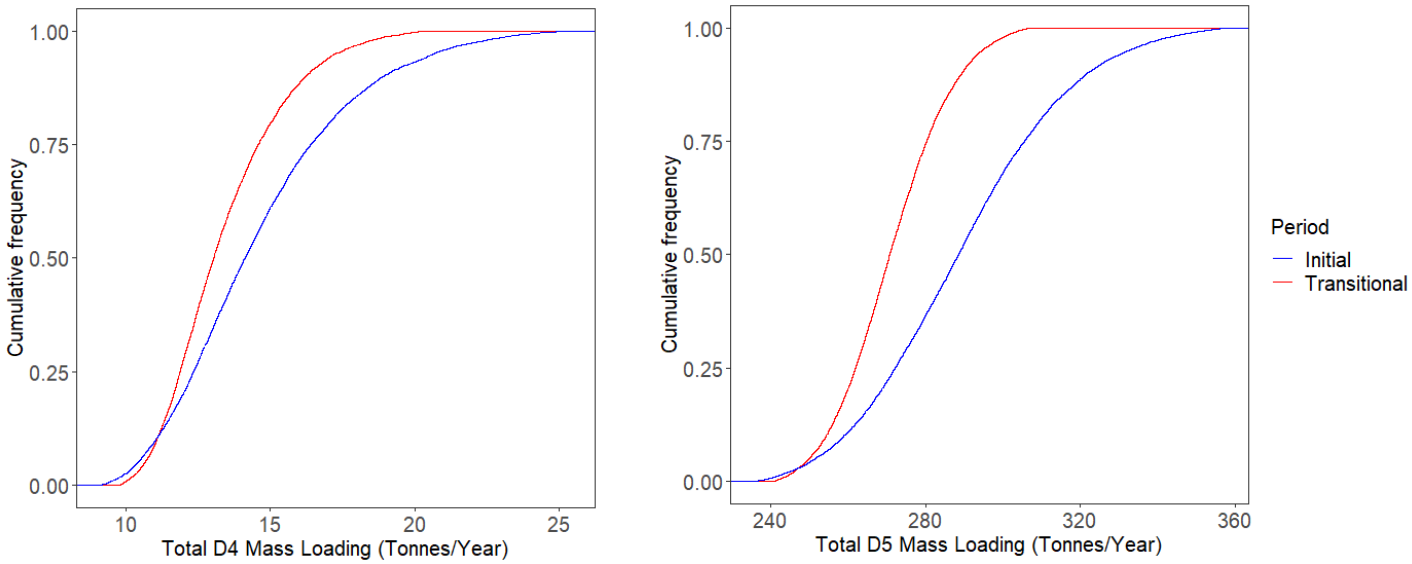
3.6 D4 and D5 Mass Loading Comparison between Periods

The mass loadings determined in this study include D4, D5, and D6 from all sources to wastewater as the underlying monitoring results reflect the D4, D5, and D6 concentrations in the wastewater influent no matter the source. The distributions of the D4 and D5 EU-wide mass loadings, based on the

[6] D6 was included in the Study Plan in the Transitional Period to evaluate the level of D6 in wastewater influent samples over a period prior to the potential implementation of the 2019 Proposed Restriction.

monitoring results and regional, cultural, socio-economic, and demographic factors were compared between the Initial and Transitional Periods and determined to be statistically different (Figure 3.5). Cumulative frequency plots show the mass loadings were lower (i.e., shifted to the left) and more tightly grouped (i.e., steeper slope) in the Transitional Period than in the Initial Period.

Figure 3.5 Cumulative Frequency Plots of the Modelled EU-wide Mass Loadings for D4 and D5 in the Initial and Transitional Periods



3.7 Comparison of Mass Loadings to Pre- and Post-Restriction Goals

The pre-restriction mass loadings of D4 and D5 in wash-off and leave-on PCPs were reported in the 2018 Restriction as 53.9 and 1,646 tonnes/yr, respectively. The goal of the 2018 Restriction is to reduce the levels of D4 and D5 to WWTPs by 78% and 97%, respectively, by restricting D4 and D5 content in wash-off PCPs (ECHA 2015). The post-restriction goals for the mass loading of D4 and D5 from PCPs to wastewater were 11.9 and 49.4 tonnes/yr, respectively. The data generated in this study indicate the total D4 mass loading to wastewater from all sources is below the post-restriction goal set for PCPs only, and the mass loading of D5 from all sources is below the pre-restriction level and is approaching the post-restriction goal for PCPs presented in the 2018 Restriction.

The 2019 Proposed Restriction presented updated mass loading from all sources, including all remaining direct uses on the market and traces from other sources, and assumed the goals of the 2018 Restriction were fully met (i.e., restriction of D4 and D5 content in wash-off PCPs). The mid-points of the proposed 2019 pre-restriction mass loadings to wastewater for D4, D5, and D6 are 141, 436, and 259 tonnes/yr, respectively. The goal of the 2019 Proposed Restriction is to reduce mass loadings of D4, D5, and D6 to wastewater from all remaining sources by an additional 50% (ECHA 2019). The EU-wide mass loadings of D4, D5, and D6 to wastewater determined in this study are compared to the pre- and post-restriction levels presented in the 2019 Proposed Restriction (Table 3.3). The mass loadings of D4, D5, and D6 determined in this study are well below the pre-restriction goals presented in the 2019 Proposed Restriction. Furthermore, the mass loadings of D4 and D6 determined in this study are well below the post-restriction goals presented in the 2019 Proposed Restriction and the mass loading of D5 falls within the range of the post-restriction goals presented in the 2019 Proposed Restriction.

Table 3.3 EU-wide Mass Loadings for D4, D5 and D6 from all Sources Compared to the Mass Loadings from the 2019 Proposed Restriction

Analyte	Period	Mass Loading (tonnes/yr)		
		Determined from the Results of This Study	Pre-restriction Level Presented by the Authorities	Post-restriction Goal Presented by the Authorities
D4	Initial	14.1 (10.3 – 21.1)	141 (98 – 184)	70.5 (49 – 92)
	Transitional	13.0 (10.5 – 17.6)		
D5	Initial	288 (250 – 335)	436 (221 – 651)	218 (111 – 326)
	Transitional	271 (249 – 296)		
D6	Transitional[7]	39.3 (36.7 – 42.2)	259 (175 – 343)	130 (87.5 – 172)

The monitoring results from this study are consistent with the goals of the 2018 Restriction and the current mass loadings of D4, D5, and D6 from all sources are below or consistent with the proposed post-restriction goals presented in the 2019 Proposed Restriction.

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[7] D6 was included in the Study Plan in the Transitional Period to evaluate the level of D6 in wastewater influent samples over a period prior to the potential implementation of the 2019 Proposed Restriction.