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SAFETY & ENVIRONMENTAL ASSURANCE CENTRE

Evaluation of a global multiscale multimedia fate model framework applied to home and personal care products in Europe

John Kilgallon[1], Cedric Wannaz[2], Olivier Jolliet[3], Lucy Speirs[1], Maria Blanco Rubio[1], Juliet Hodges[1]

[1]Safety & Environmental Assurance Centre (SEAC), Unilever, Sharnbrook, Bedfordshire, UK

[2]Eurisko Research, 3107 Village Circle, Ann Arbor, 48108, MI, USA

[3]Environmental Health Science, School of Public Health, University of Michigan, Ann Arbor MI, USA

E-mail contact: john.kilgallon@unilever.com

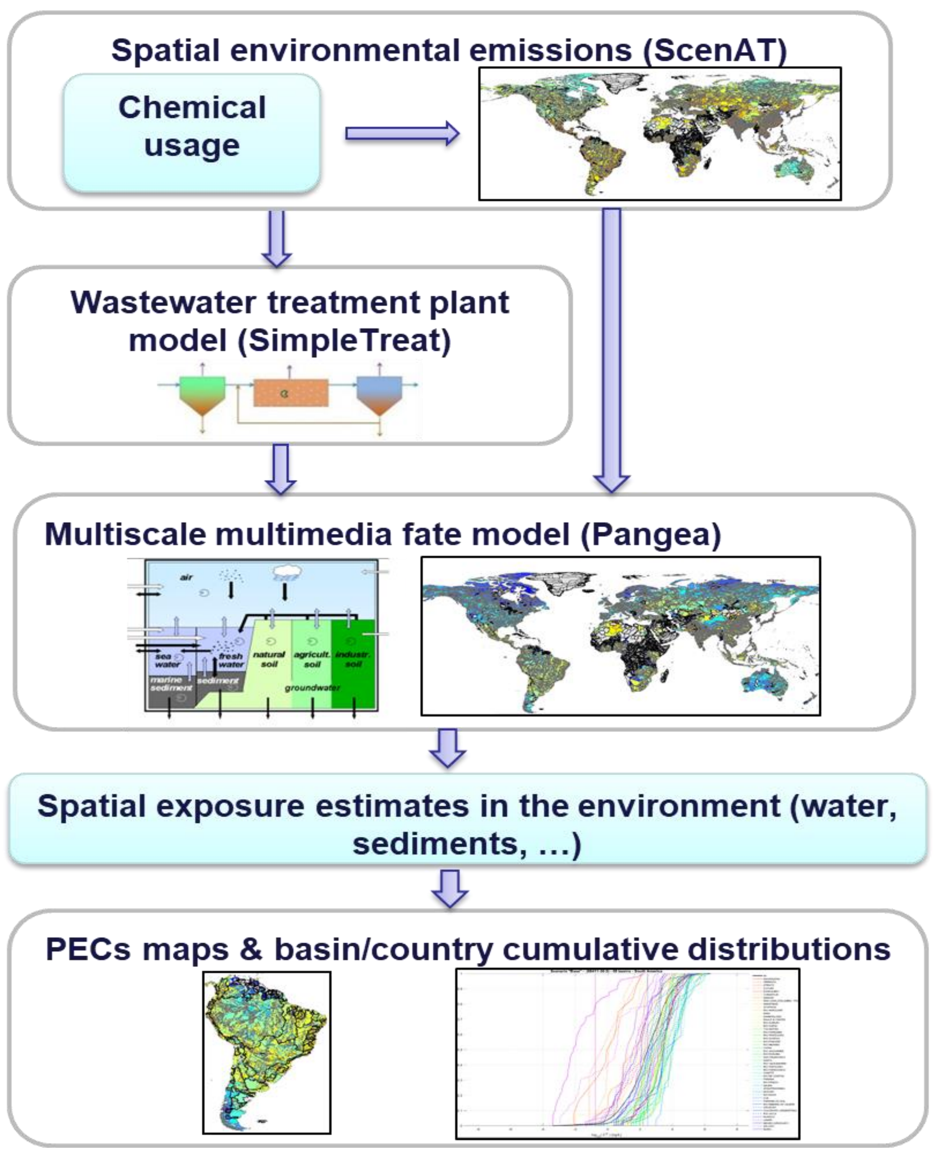

Figure 1 – Environmental exposure modelling framework

Introduction

Regulatory environmental exposure scenarios are typically conducted using coarse approaches and are often criticized for their lack of spatial and temporal resolution, which can create large variations in exposure for many ingredients. This is typical for home care and beauty and personal care (HC & BPC) products which have a wide dispersive use and are discharged into the sewer systems after use. An evaluation of the updated global Pangea/EcoHope multi-scale multimedia model with HydroBASINS hydrology (1,2] with Asia monitoring data demonstrated good agreement. Here we present an initial and provisional evaluation of the model focusing on European monitoring data.

Objectives:

- Predict freshwater concentrations of five case study HC & BPC industry ingredients: linear alkylbenzene sulfonates (LAS) (anionic surfactant), Benzophenone-3 (BP-3), Octocrylene (OC), ethylhexyl methoxycinnamate (OMC) (UV filters) and Triclosan (TCS) (antimicrobial) in Europe (EU) using the Pangea/EcoHope framework.
- Compare freshwater predicted environmental concentrations (PECs) to measured environmental concentrations collected from the literature and the Norman Database [3]

Methods – Monitoring

- Georeferenced monitoring data were collated from literature and the Norman database for the select case study ingredients across 15 European countries (Croatia, Denmark, Germany, Greece, Hungary, Italy, Moldova, Poland, Romania, Serbia, Slovakia, Spain, Switzerland, Ukraine and United Kingdom) (figure 2) (table 1).
- In total, 10,624 samples (freshwater: 10,409, 215 sediment) were collected (1996-2016). TCS was the most data rich ingredient (9,165 points, of which 8,960 were sampled in Germany). A specific search criteria under the Norman Network data search was used to select the data of the case study ingredients (i.e. from 2005, surface water, sediments, quality indicators), many of which were single time point grab samples.
- For freshwater LAS data, 1996-1998 UK monitoring from across the Aire, Calder, Went and Rother catchments [4] were used due to the lack of recent LAS monitoring campaigns.
- There is variation across the monitoring literature in the limit of detections (LoDs) of the chemistry methods used. Median LoDs were thus calculated to provide an indication for each ingredient (figure 4) at which it is no longer possible to analytically determine a test sample that can be reliably distinguished from zero.

Ingredient	Germany		Spain		United Kingdom
	Water	Sed	Water	Sed	Water
LAS				54	1171
TCS	8960	1	23	106	25
BP-3	4		53	6	5
OMC				6	
OC	12			20	

Table 1 – Number of measured LAS, TCS, BP-3, OMC and OC monitoring points in the top 3 countries

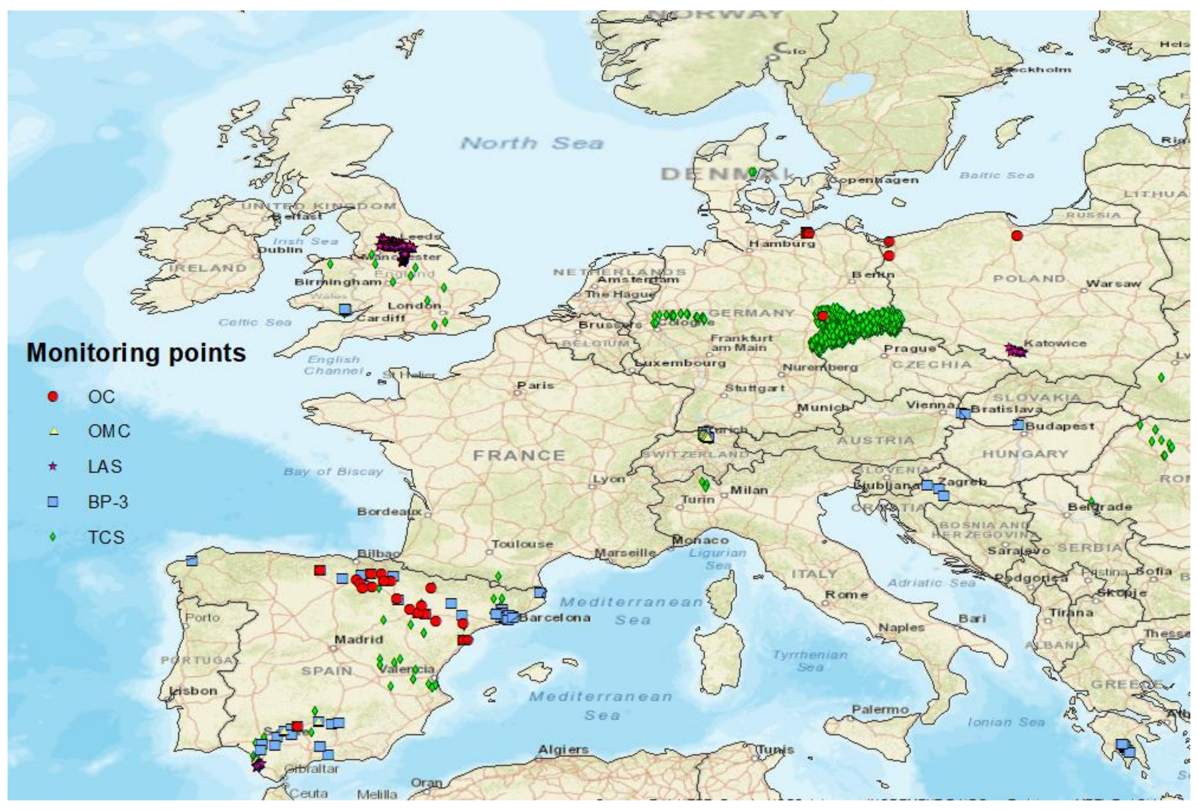

Figure 2 – Freshwater and sediment monitoring samples for LAS, BP-3, OC, OMC and TCS locations modelling framework

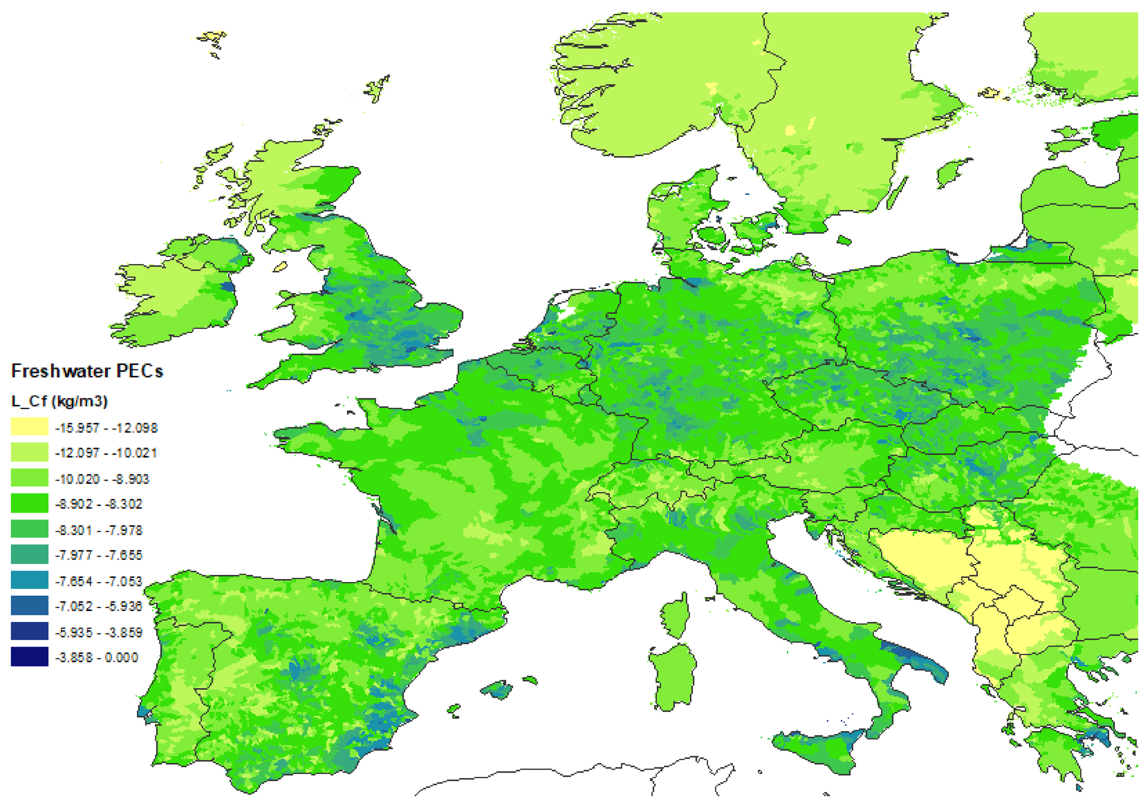

Figure 3 – TCS PEC concentration map across Europe

Methods – Modelling

- Following the EcoHope/Pangea model framework (figure 1), an emissions inventory for the five HC & BPC case study ingredients was completed globally. This was done by compiling 2016 total market annual sales tonnage data from the Euromonitor database for each country (Table 2).

Ingredient	Country (ScenAT-88) Tonnes / year		
	European Max	European Total	Total use (Globally)
LAS	69,818 (UK)	357,760	4,330,274
TCS	21 (Poland)	182	2,895
BP-3	72 (Germany)	221	1.273
OMC	317 (Germany)	1,341	6,886
OC	416 (Spain)	2,358	7,473

Table 2 – Estimated global annual tonnage emissions

- To address temporal discrepancy between the year of monitoring data (1996-1998) and measured tonnage estimates for the LAS case study, a per capita use value (3.28 grams/person/day) was used in combination with population from the year of monitoring (69,818 tonnes/year) [5].
- Tonnages were first run through ScenAT to account for population water use, connectivity to sewage treatment and removal mechanisms in sewage treatment plants (STP) based on the SimpleTreat model (figure 1) [1].
- Emission inventories were then run through Pangea to simulate the fate and transport of the ingredients in the environment (i.e. partitioning, hydrological processes) to determine environmental concentrations in the freshwater, sediment, soil and air compartments.
- Higher spatial resolution was assigned to areas with high water volume, high population and monitoring locations to cover areas of interest.

Results and Discussion

- Freshwater and sediment compartments were analysed by comparing simulated Pangea PECs (figure 3) in each HydroBASINS spatial unit with measured concentrations. It shows that LAS, TCS and BP-3 monitoring data are typically exceeding PECs (figure 4). OC and OMC PECs both overpredict monitoring data. However, for BP-3, OC and OMC sample numbers are relatively low so we focus on TCS and LAS here.
- TCS modelling data is underestimating monitored data to within half an order of magnitude (figure 4, figure 5). The top 10 rivers by monitoring data points have been added to the 1:1 plots (figure 5) for both freshwater and sediment compartments and show good correlation for sediment (r=0.72) but poor for freshwater (r=-0.09). As the emission and monitoring data across regions are comparable (within an order of magnitude) the model is unable to discriminate between regions and therefore its ability to rank rivers.
- Due to the nature of monitoring campaigns, sampling sites are primarily confined to limited areas due to logistical reasons and are usually designed for catchment based modelling (i.e. GREAT-ER) [6]. As a result, spatial variability of TCS freshwater PECs and monitoring data is not captured (figure 5) as the majority of sampling locations were collected in relatively close proximity (i.e. the Elbe catchment).
- From our previous analysis in Asia [2] it was identified that emissions are not a substantial source of uncertainty compared to model input parameters (i.e. half lives) and for substances which are rapidly degraded such as LAS. PECs tend to be limited by dilution (i.e. volume of water) on the main rivers, with highest PECs being identified on tributaries rather than on the main river streams. Due to this flushing effect in large rivers, research on how this is impacting PECs in Europe and subsequently the evaluation is required.
- In addition, to further understand LAS PECs, further analyses on loss processes in sewer, STPs and river are being conducted by completing a sensitivity analysis in Europe as the removal processes are highly complex to model accurately.
- Temporal variability between the model and monitoring data is expected to be a significant factor as the model is based on annual averages of emissions and river flow over time. In most circumstances monitoring data, however, are based on one single point in space and time (i.e. a grab sample) with no seasonal variation.-Including monthly flow data in the model is expected to improve the evaluation since river flow can be highly influenced by the seasons.

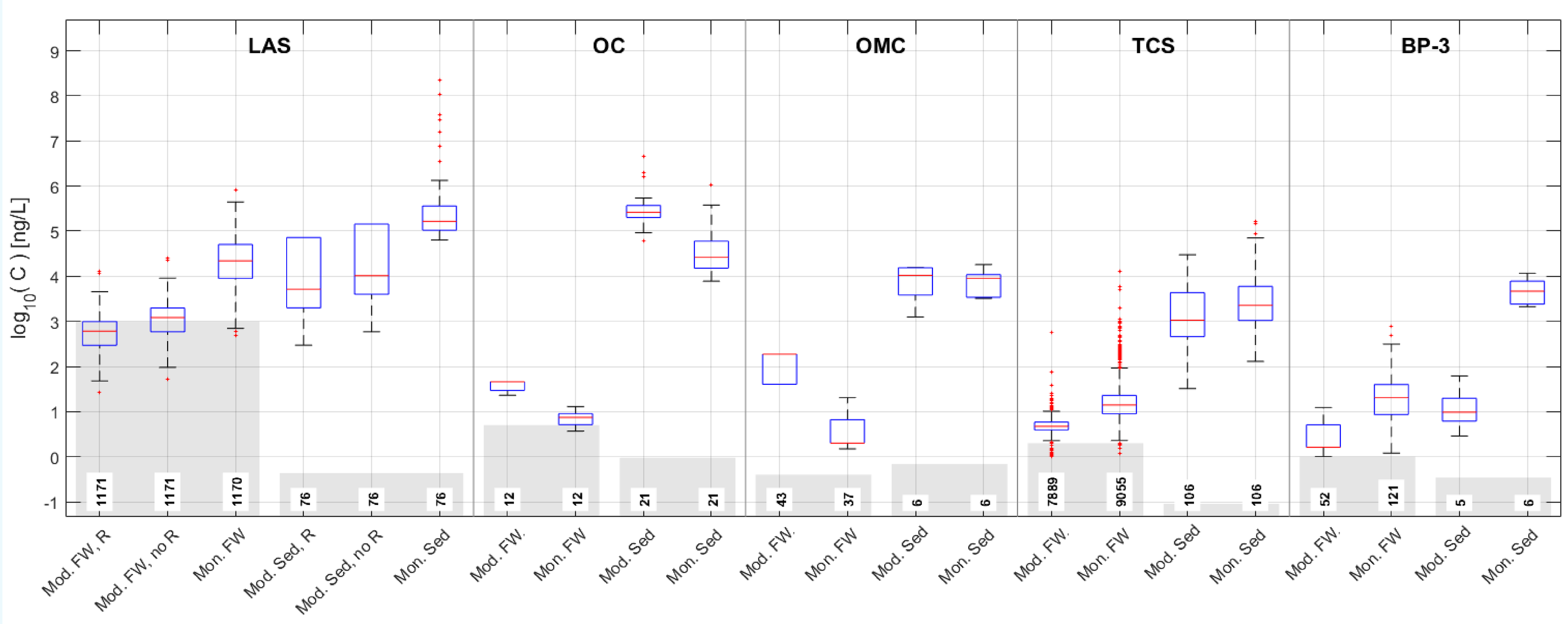

Figure 4 – Boxplot of fresh water (FW) and sediment (Sed) monitored concentrations (Mon) and modelled concentrations (Mod) by Pangea in Europe. Central mark - median (50th percentile), boxes - 25th and 75th percentiles. Whiskers – ca ±2.7σ and 99.3 percent coverage for normally distributed data. Red dots – statistical outliers. Grey - median limit of detection. Mod, FW/Sed, R = 50% in-sewer removal.

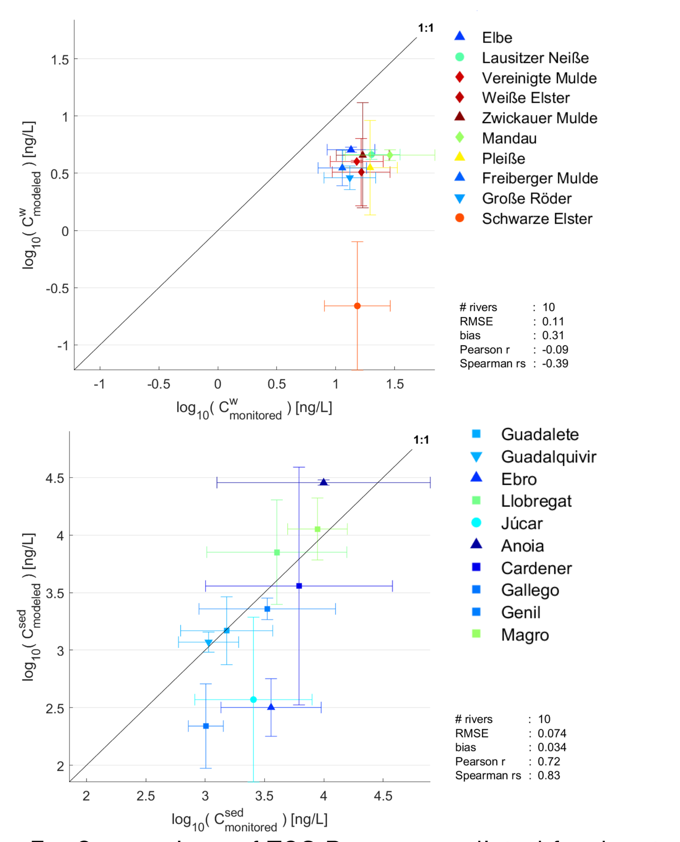

Figure 5 – Comparison of TCS Pangea predicted freshwater and sediment PECs with monitored concentrations. The top 10 rivers by number of monitoring points across Europe. Central mark – means, errors bars - one standard deviation

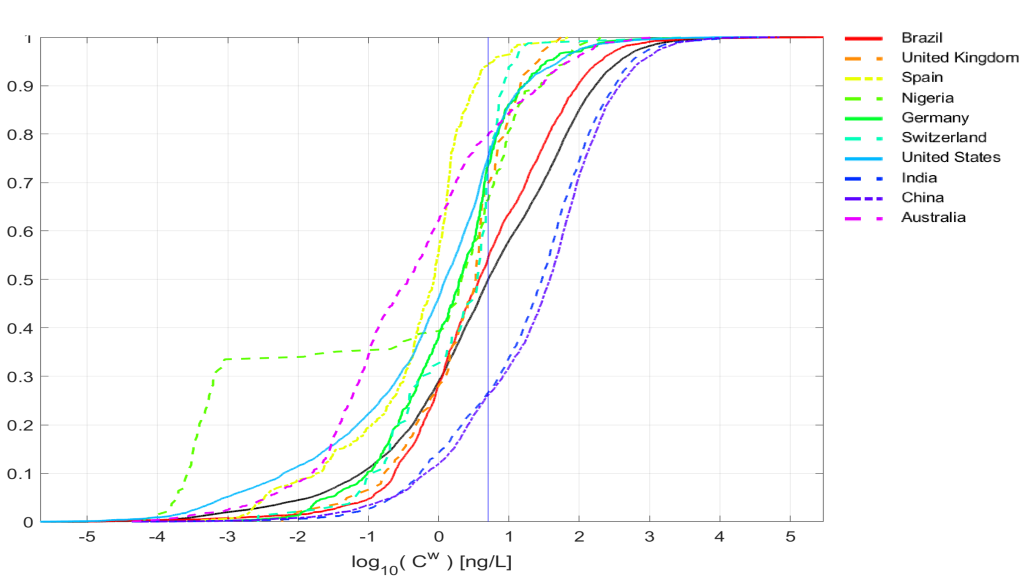

Figure 6 – Cumulative distribution of TCS PECs globally across 10 countries

Conclusion

The global Pangea model framework [1], allows a comparison of freshwater and sediment PECs across Europe. Initial and provisional comparisons of modelled versus monitored values for the five case study ingredients are typically to within an order of magnitude of monitored data (figure 4). This level of predictivity is not unexpected for this initial comparison given the spatial and temporal differences between the monitoring and modelled data used. However, further work to understand the uncertainties in both monitoring and modelling inputs, addressing in particular the reduced spatial and temporal availability of monitoring data, is in progress.

Next steps:

- Include monthly flows in the Pangea hydrological model;
- Complete sensitivity analysis in Europe to further understand model performance;
- Increase spatial resolution around areas where monitoring data exists;
- As Pangea is now global, it is possible to analyse continental variations in PECs (figure 6) compared to available monitoring data.

[1] Wannaz C, Franco A, Kilgallon J, Hodges J, Jolliet O. 2018b. A global framework to model spatial ecosystems exposure to home and personal care chemicals in Asia. Sci. Total Environ. 622–623, 410–420.

[2] Jolliet O, Wannaz C., Kilgallon J, Speirs L, Franco, A., Lehner, B., Veltman, K., Hodges, J., 2020 Spatial variability of ecosystem exposure to home and personal care chemicals in Asia.

[3] Norman Network. 2020. Available at: https://www.norman-network.net/?q=node/24 (Accessed Jan 2020)

[4] Holt MS, Fox KK, Burford M, Daniel M, Buckland H. 1998. UK monitoring study on the removal of linear alkylbenzene sulphonate in the trickling filter type sewage treatment plants. Contribution to GREAT-ER project #2. *Sci Total Environ*, 1998; 210-211: 255-269.

[5] Holt MS, KK Fox, M Daniel, H Buckland. 2003. LAS and Boron monitoring in four catchments in the UK contribution to GREAT-ER, The Science of the Total Environment 314-316: 271-288.

[6] Kehrein N, Berlekamp B, Klasmeier, J. 2015. Modelling the fate of down-the-drain chemicals in whole watersheds: New version of the GREAT-ER software. *Environ Modell Softw*, 2015: 64, 1-8.