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Screening human health risk of environmental and direct exposure to personal care products

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Lestremau, INERIS; M. Chevreuil, école pratique des hautes études UMR SISYPHE. Parabens are a group of alkyl esters of the p-hydroxybenzoic acid that are widely used as preservatives in pharmaceutical and personal care products. In order to gain knowledge about presence (or not) of parabens in different water categories, a screening study was performed in France and in 5 overseas territories in 2012. More than 1700 occurrence data were obtained in water and sediment samples for 3 parabens (methylparaben, ethylparaben and propylparaben). Samples were collected on more than 160 sites during 3 sampling campaigns (spring/summer/fall). Sampling sites representing different types of landcover were chosen. This screening study has been conducted on a larger datasets compared to previous reported literature data. For methylparaben and propylparaben, concentrations are similar in all samples, and no obvious effect could be identified associated to the type of anthropogenic sources. The occurrence of parabens as ubiquitous contaminants in almost 100% of the analysed samples can represent a risk. At the measured levels, acute and chronic toxicity is not expected with any of these compounds tested individually. However, currently available PNEC values do not take into account non-standard endpoints, such as endocrine disrupting effects which need to be considered in assessing the potential risks associated to occurrence of parabens in the aquatic environment. Parabens are common hygiene product ingredients and special care must be considered for sampling.

139 Field dissipation and risk assessment of typical personal care products in biosolid-amended soils G. Ying, Chinese Academy of Sciences / State Key Laboratory of Organic Geochemistry; F. Chen, Z. Chen, Chinese Academy of Sciences Guangzhou Institute of Geochemistry; Y. Ma, Chinese Academy of Agricultural Sciences. Antibacterial agents triclocarban (TCC) and triclosan (TCS), and synthetic musks AHTN (Tonalide) and HHCB (Galaxolide), and antifungal agents climbazole, clotrimazole and miconazole are extensively used in many household and personal care products. After use, they enter wastewater treatment plants and finally reach the receiving environment via discharge of effluent and disposal of sewage sludge. Due to their hydrophobic nature, these compounds have been detected in biosolids. Land application of biosolids has become a common practice throughout the world. However, concerns continue to be raised about the potential risks of this practice to the soil environment and public health. The aims of this study were to evaluate contamination levels of four typical personal care products (TCC, TCS, AHTN and HHCB as well as three antifungals) in biosolids and biosolid-amended soils of three field trial sites (Zhejiang, Hunan and Shandong), investigate their dissipation patterns in the biosolid-amended soils under different treatments (T1: single application; T2: repeated applications), and assess their potential risks to terrestrial organisms. The seven target compounds (TCC, TCS, AHTN and HHCB, as well as three antifungals) were detected in all biosolid-amended soils from the three trial sites (HN, SD and ZJ) in October 2010. The concentration levels for the seven compounds of both T1 and T2 in the biosolid-amended soils of the three sites were found to have the following order: TCC > TCS > AHTN > HHCB > miconazole > clotrimazole > climbazole. TCC was found to have concentrations ranging from 111 (ZJ) to 365 (SD) mg/kg for T1, and from 454 (ZJ) to 1584 (HN) mg/kg for T2. The presence of these personal care products in the soils applied with biosolid indicates the persistence of these compounds in the soils. One-year field monitoring at the SD site showed dissipation of these chemicals with their half-lives of 51-1000 days. Preliminary risk assessment suggests that TCC and TCS might pose high risks to soil organisms based on the limited toxicity data, while AHTN and HHCB showed low to medium ecological risks. The results from this study will help evaluate the environmental impacts of these personal care products associated with biosolid application on agricultural land.

140 Determination of toxicity data of ultraviolet filters toward selected aquatic organisms for a preliminary environmental risk assessment D. Molins Delgado, Environmental Chemistry; P. Gago, CSIC / Environmental Chemistry; S. Diaz-Cruz, IDAEACSIC / Environmental Chemistry; D. Barcelo, IIQABCSIC / Environmental

Chemistry. UV Filters (UV-F) constitute a heterogeneous group of chemicals used as a protection against the harmful effects of the UV solar radiation. We can find them in a wide range of compounds such as personal care products or as additives in polymeric materials that need protection against sunlight. Nowadays, these emerging pollutants are considered persistent due to their continuous release into the environment [1]. Some studies performed mostly *in vitro* but also in *in vivo* have shown that many of these compounds can produce adverse effects on reproduction, interfering the normal development on fishes and rodents [2,3]. For instance, 4MBC has similar estrogenic effects than those of 17- β -estradiol in mammals and amphibious species [4]. Despite that, the knowledge on the potential chronic or acute toxicity to biota of these compounds is still very scarce [5,6]. In order to fill the knowledge gap on the ecotoxicity of UV-F, this study aims to i) determine the EC₁₀, EC₂₀, EC₅₀ values of the target UV-F through a series of toxicological assays on three aquatic species i.e. *Daphnia Magna*, *Selenastrum Capricornutum* and *Vibrio Fischeri*, and ii) explore the potential synergetic effects in UV-F mixtures. Besides that, the ecotoxicological risks associated to UV-F and some of their known transformation products in selected case studies will be presented.

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141 Do insect repellents induce drift behaviour in aquatic non-target organisms? P. Fink, University of Cologne / Aquatic Chemical Ecology; K. Vasters, University of Cologne / Workgroup Aquatic Chemical Ecology; E. von Elert, University of Cologne / Biology. Synthetic insect repellents are substances applied to surfaces to discourage insects, mainly mosquitoes from landing on these surfaces. As some of these repellents have been detected in surface waters, they may also exert repellent effects on aquatic non-target organisms. In running water systems, aquatic invertebrates actively enter the drift in order to move out of locally malign environments. We thus tested the hypothesis that the widely used insect repellents DEET (N,N-Diethyl-m-toluaminide) and EBAAP (3-(n-n-butyl-n-acetyl-amino)-propionic acid ethyl ester) induce drift behaviour in the aquatic invertebrates *Gammarus pulex* (Crustacea, Amphipoda) and *Cloeon dipterum* (Insecta, Ephemeroptera), using a laboratory-scale drift assay. If the insect repellents cause drift behaviour in such non-target organisms at environmentally relevant concentrations, the resulting changes in resource consumption, but also in prey availability for predators, may influence ecosystem functioning in streams. Further, the effects of insect repellents on the behaviour of non-target organisms may also interfere with these organisms' natural behavioural responses to predator infochemicals with resulting detrimental effect of repellents in surface waters.

142 Screening human health risk of environmental and direct exposure to personal care products A. Ernstoff, University of Michigan / Environmental Health Science; S.A. Csiszar, University of Toronto / Dept of Chemical Engineering and Applied Chemistry; P. Fantke, Technical University of Denmark / IER; O. Jolliet, University of Michigan / School of Public Health. No existing assessment framework evaluates the uptake of product ingredients via both product use and via environmental exposure. To address this issue, this work presents a framework to flag potential risk and compare use and environmental exposures of chemicals within personal care products. The framework is based on the novel concept of the *product intake fraction (PiF)* defined

as: the chemical mass of substance *i* within a product *x* taken in by humans via both use-phase and environmentally related emissions, per unit of chemical mass within that product. Chemical mass within products may be taken in by individuals via several use-phase pathways including direct dermal contact, and via several environmentally related pathways, such as food chain bioaccumulation. Applying the *PiF* concept to evaluate all relevant exposure pathways for 25 chemicals within personal care products, we found use-phase *PiFs* ranged from 0.02 to 1 (kg intake/kg applied) and were mostly dominated by dermal intake, and environmental *PiFs* were generally several orders of magnitude lower and mostly dominated by intake of lipophilic chemicals with high bioaccumulation. *PiFs* may also be converted into daily population-scale intakes using production volume data. We demonstrated this framework using a paraben case-study with available NHANES biomarker data for US women, and found model predictions generally in-line with observed *in vivo* paraben concentrations. To flag potential risk, the *PiF* is combined with Oral Equivalency Doses (OED) back-calculated from e.g. ToxCast *in vitro* bioactivity assays to estimate a maximum allowable fraction of chemical within a product. Evaluating the maximum allowable fraction for 25 chemicals within personal care products and to known product concentration data for 11 of the compounds (documented by the Household Product Database), we find 4 out of 11 potential chemical/product combinations with known chemical fractions within products exceeding the maximum allowable chemical fractions and 9 out of 10 with margins of exposure lower than 100. Evaluating both use-phase and environmentally related intakes of chemicals due to consumer products, suggests risk evaluation of dermal application of products must be held in high priority due to intakes that may exceed oral equivalency dose estimates for bioactivity and use-phase intakes generally dominating over environmentally associated intakes.

143 Sulphidation kinetics of silver nanoparticles reacted with metal sulphides B.A. Thalmann, Eawag; A. Voegelin, Eawag Swiss Federal Institute of Aquatic Science and Technology; E. Morgenroth, ETH Zurich; R. Kaegi, . Silver nanoparticles (Ag-NP) are used in many consumer products, most importantly textiles and cosmetics, due to the well-known antimicrobial properties of Ag⁺. Eventually, these particles will be released from products and either reach surface waters or a wastewater treatment plant through the municipal sewer system. Ag-NP undergo either dissolution or other transformations, most importantly they react with sulphide to form Ag₂S. Because Ag as Ag₂S is several orders of magnitudes less toxic than ionic Ag⁺, the sulphidation of Ag-NP is of prime interest. Thermodynamically Ag₂S is not the stable form of Ag in aerobic waters but it has been shown to be rather resistant towards oxidation with dissolved O₂, due to the formation of extraordinarily strong sulphide bonds. Therefore, the speciation of Ag-NP in aerobic surface waters is determined by the kinetic rate laws rather than by the thermodynamic stabilities. However, little is known about the sulphidation kinetics of silver nanoparticles under conditions typical for surface and waste waters. Although sulphide is dominantly present as bisulphide (HS⁻, pH >7) in anaerobic environments, it is not stable under oxidizing conditions. Metal sulphides such as ZnS and CuS that resist rapid oxidation by O₂ may serve as sulphide sources for the sulphidation of Ag-NP under aerobic conditions. The goal of this study was, therefore, to assess the sulphidation kinetics of Ag-NP (10, 20, 40, 70 and 100 nm) reacting with ZnS and CuS. We measured the sulphidation kinetics of Ag-NP in oxygen saturated DI water buffered to a pH of 7.5. The reactions exhibited pseudo-first-order kinetics, with rate coefficients depending on the Ag-NP size and initial metal sulphide concentration. Analytical electron microscopy analysis revealed Ag-Ag₂S core-shell formation of Ag-NP. Reported concentrations of ZnS in aerobic surface waters range from 1.5 and 127 nM with corresponding half-life times of 5 days. By material-flow analysis predicted Ag-NP concentrations are 6.7 pM in combination with now experimentally determined half-life times of the Ag-NP ranging from 2 h to 2.4 days, therefore suggest a complete sulphidation of the Ag-NP also in aerobic surface waters.

144 Co-tolerance of microbial litter decomposers to silver nanoparticles and antibiotics A. Tlili, Eawag / Environmental Toxicology; M. Kuburic, Leibniz Institute of Freshwater Ecology and Inland Fisheries / Experimental Limnology; C. Canhoto, IMAR-CMA and Department of Life Sciences, University of Coimbra; R. Behra, Eawag / Department of Environmental Toxicology; M. Gessner, Leibniz Institute of Freshwater Ecology and Inland Fisheries. With the accelerated production and use of silver nanoparticles (AgNP) in commercial products, freshwater ecosystems will most likely serve as recipients of, and repositories for, these nanomaterials. In addition to their potential environmental toxicity, there is a growing concern that AgNP act as a selective agent in antibiotic resistance. The aim of this study was therefore to evaluate the consequences of a long-term exposure to silver nanoparticles (AgNP), AgNO₃ (as a source of Ag⁺) or a mixture of 5 antibiotics on the pollution-induced community tolerance (PICT) of microbial decomposers associated to leaf litter in freshwaters. Moreover, specificity of PICT was assessed by evaluating the co-tolerance between these toxicants. Bacterial production by ¹⁴C-leucine incorporation and fungal sporulation rate by microscopy were used in short-term inhibition bioassays with antibiotics, AgNP and AgNO₃ to assess sensitivities of pre-exposed microbial decomposers. Diversity profiles of the bacterial and fungal communities following the different treatments were determined by DNA fingerprint technique. The results demonstrated that chronic exposure to AgNP, AgNO₃ or ATB induced structural shifts in the community and led to tolerance enhancements in the bacterial communities. On the other hand, communities exposed to AgNP also increased their tolerance to Ag⁺ and ATB. Similarly, communities exposed to Ag⁺ showed an increase of their tolerance to AgNP and ATB. Finally, bacterial community exposed to ATB displayed higher tolerances to AgNP and Ag⁺. Overall, our findings support the fact that exposure to toxicants as diverse as ATB and metal-based nanoparticles may lead to similar effects when considering complex biological systems such as microbial litter decomposers raising a direct environmental and public health implications.

145 Fate and heteroaggregation of titanium dioxide nanoparticles in natural surface waters D.L. Slomberg, CNRS AixMarseille Université / CNRS AixMarseille Université; P. Ollivier, BRGM; J. Labille, CNRS. Despite continual progress in the development of engineered nanoparticles (ENPs), their fate and impact on the natural environment remain largely unknown due to challenges in detection and quantification. New strategies are necessary to more accurately assess ENP hazard and exposure. Titanium dioxide (TiO₂) NPs are of specific interest due to their use in a wide range of commercial products. The fate of TiO₂ NPs in aqueous environments depends on pH, ionic strength, and natural organic matter (NOM) concentration, however a lack of understanding regarding the behaviour of these NPs in natural systems remains, especially at relevant concentrations (µg/L range) where homoaggregation is less likely.¹ The heteroaggregation of TiO₂ NPs with natural suspended particulate matter (SPM) in the aqueous environment is also an important process in transport and fate.² Herein, we present the fate of TiO₂ NPs in natural waters from a lake with high NOM and low mineral SPM (Cholet, France), and a main river with high SPM and low NOM (Rhône River at Arles and Jons, France). The holistic approach presented here studies the fate of TiO₂ NPs under relevant concentrations in natural lake and river waters by characterization of their physical-chemical interactions and heteroaggregation with natural suspended matter (NSM). Homo- and hetero-aggregation kinetics were measured using light scattering, nanoparticle tracking analysis, and laser diffraction. The natural waters were evaluated for pH, ionic strength, NOM content, colloidal composition, and major and trace elements to elucidate the key contributors to NP fate. The kinetics of NP-NP and NP-NSM interactions were evaluated and the respective sticking efficiencies for homo- and hetero-aggregation were determined. They were influenced by the pH, ionic strength, and NSM composition of the waters. Nanoparticle concentration also played a role in the heteroaggregation mechanism, assuming binding between neighbour mineral colloids.