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The importance of household pharmaceutical products disposal and its risk management: Example from Southwestern Europe

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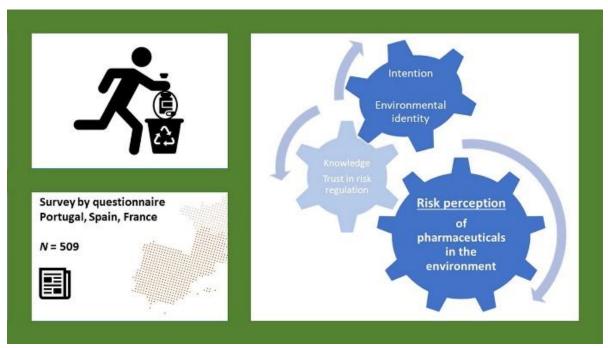
Highlights

- Pharmaceuticals in the environment is an emergent risk, often unknown by the public.
- Need to promote the proper disposal of household pharmaceutical leftovers.
- Stage models of behaviour change highlight the relevance of risk perception.
- Survey data shows that risk perception related to intention and disposal behaviour.
- Environmental identity promoted the translation of intention to disposal behaviour.

Abstract

The presence of pharmaceuticals in the environment is an emergent unknown environmental problem, linked to increased production and consumption of pharmaceuticals and, as such, understanding risk perception is fundamental. This study focuses on exploring causes (trust and knowledge) and effects (intention and proper individual disposal of pharmaceutical leftovers) of environmental and health risk perception. Survey data was collected in Portugal, Spain, and France (sample of 509 individuals). Data illustrated that in France, where the quantity of recycled pharmaceuticals is much higher, respondents reported a higher need for

knowledge, but not a higher risk perception nor self-reported behaviour. Although previous research illustrates that higher trust correlates with lower risk perception, we found a positive correlation, which highlights the need to comprehend these variables in emergent risks. Results further confirmed a hypothesized moderated mediation model to explain proper disposal behaviour. We found an indirect effect of risk perception on behaviour through intention, which was stronger for participants with higher environmental identity. Understanding the causes and effects of risk perception of pharmaceuticals in the environment thereby contributes to improve pharmaceutical waste management processes and to promote the proper disposal of pharmaceuticals.



Keywords

Pharmaceuticals in the environment; Risk perception; Knowledge; Trust; Pharmaceutical waste; Environmental identity

1. Introduction

Over the last few decades, the presence of pharmaceutical products in the environment has become a worldwide issue of increasing concern. This study contributes to understand one of the pathways by which pharmaceuticals reach the environment, i.e., the improper disposal of pharmaceutical leftovers. More concretely, we will analyse environmental and health risk perception as a product of individual trust in pharmaceutical waste management and as a determinant of proper disposal of pharmaceuticals in three Southwestern European countries (Portugal, Spain, and France). Pharmaceuticals in the environment is an emergent problem that has not been highly debated in the public arena, and most individuals might not be aware of the issue. In this context, risk perception is expected to be a paramount variable for understanding individual disposal of unused pharmaceuticals (Fatokun et al., 2011). More specifically, we will explore whether risk perception relates to trust in risk regulation, and whether it might influence proper disposal through intention, depending on individual environmental identity.

1.1. Pharmaceuticals in the environment

The presence of pharmaceutical residues in the environment has been an issue of concern since it was first reported in the 1970s (<u>Daughton, 2016</u>). Pharmaceuticals are produced and used in large quantities, and their production and consumption is expected to be amplified with the increase of life expectancy (<u>Lacorte et al., 2017</u>). The sources and pathways of pharmaceutical products in the environment have been well identified: metabolic excretion from individuals who ingest pharmaceuticals (wastewater effluents, septic tanks), agriculture and industrial activity (industrial wastewater and waste), livestock activities (waste lagoons, manure application to soil), as well as indirectly through improper disposal of pharmaceutical residues by individuals (e.g., <u>Adeel et al., 2017</u>). As such, pharmaceutical residues are present in water (both wastewater and superficial water, although in smaller concentrations in the latter) and in the soil, thus contaminating crops (<u>McEachran et al., 2017</u>).

Pharmaceuticals might be persistent, toxic, and have the potential for bioaccumulation in organisms belonging to different trophic levels, therefore becoming hazardous to wildlife (Brodin et al., 2014). Most of the pharmaceuticals that have been investigated up to now do not pose an acute threat to the environment, but the remaining pharmaceuticals do (World Health Organization, 2012). There is clear evidence for the negative effects of such pharmaceuticals in the environment, namely in some animal, fish, and plant species (e.g., Green et al., 2015). As far as human health is concerned, most research indicates that no pharmaceutical is likely to pose a significant threat via environmental exposure, in particular through the consumption of drinking water or crops irrigated with treated wastewater (World Health Organization, 2012, Wu et al., 2015). However, some researchers state otherwise. Adeel et al. (2017) suggest that there is enough published evidence to establish a causal relationship between the high prevalence of oestrogen in the environment worldwide (soil and water) and breast cancer (even though there are significant gaps in current knowledge about oestrogen levels in the environment). In addition, the presence of antibiotics in the environment is also linked to antibiotic resistance (Martinez, 2009), which leads to higher medical costs, prolonged hospital stays, and increased mortality. Furthermore, in its latest report, the World Health Organization (2012) stresses that risk assessment studies of most pharmaceuticals in the environment are not yet conclusive, and there is a lack of systematic assessments and multiple knowledge gaps, such as their effects in vulnerable populations. Consequently, in face of uncertain risks for the environment and for human health, the need for precautionary management action to reduce the release of pharmaceuticals into the environment has been highlighted (Kümmerer, 2010). Numerous initiatives have been launched to decrease the presence of pharmaceuticals in the environment. These include actions in the industry itself (green pharmacy), as well as medicine (sustainable prescription, control on behalf of healthcare establishments), technology (effluent treatment), and others aimed at individual behaviour (raising awareness about disposing of pharmaceuticals). This study focuses on the latter, by exploring risk perception of hazards related to the presence of pharmaceuticals in the environment.

1.2. Trust, knowledge and risk perception

Researchers have suggested that trust acts as a heuristic to assess risks when people do not have enough time, cognitive capacity, or motivation to deliberately evaluate risks (<u>Visschers and Siegrist, 2008</u>). People seldom possess the time, or an elaborate knowledge of risks, or the will to assess risk, and thus tend to rely only on other types of information. In fact, there is evidence that environmental risks are frequently estimated based on individual trust that

authorities are managing them, which results in an association between higher levels of trust and lower levels of perceived risk (Earle, 2010). This effect, however, does not appear to be universal. In a cross-national study, Viklund (2003) illustrated that the relation depended on both the country and the type of risk: the relation was much weaker for Spain and France (in comparison with Sweden and the United Kingdom) and appears to be stronger for dreadful risks, such as nuclear ones.

Most studies on the relations between knowledge, trust, and risk perception focus solely in perceived knowledge (Siegrist and Cvetkovich, 2000). However, we will analyse three types of knowledge: factual knowledge, perceived knowledge, and perceived need for knowledge. Factual knowledge, also referred to as objective knowledge, is the accurate or scientific-type of knowledge. Perceived knowledge, also referred to as subjective knowledge, corresponds to the individual's extent of their knowledge within a certain domain. There is evidence suggesting these two types of knowledge are positively related but might correspond to different constructs (Carlson et al., 2009). Following the model of risk information seeking developed by Griffin et al. (1999), we further explored the perceived amount of knowledge needed to deal with a hazard. Individuals appear to engage in information seeking and processing until they have reached the knowledge that they perceive to be comfortable and necessary, which highly depends on subjective norms about knowledge and information gathering about the risk. Higher perceived need for knowledge includes a motivational aspect and is associated with more systematic processing of information.

Pharmaceuticals in the environment pose an emerging problem that is still unknown for many people, as recently recognized by the European Union strategic approach to pharmaceuticals in the environment (European Commission, 2019). We will test whether people use their trust on how authorities deal with risk to estimate the magnitude of the risk, and whether this effect holds for all three countries. Understanding the relation between trust in risk regulation and risk perception is fundamental for the management of this environmental problem. In particular, we want to analyse if the individuals' trust in risk regulation of pharmaceuticals in the environment is negatively correlated to their risk perception, and, if so, whether it might be a s demotivating factor for them to take action.

1.3. Risk perception, intention, and behaviour

Psychological stage models of behaviour change, such as the stage model of self-regulated behavioural change (e.g., <u>Bamberg, 2013</u>), indicate risk perception as an important factor that promotes change from a pre-intentional phase to an intentional one. Recent studies show that consumers keep large amounts of unused medicines at home (<u>Dias-Ferreira et al., 2016</u>) and that only a small percentage of the consumers is aware of the importance of adequate medicine waste disposal (<u>Amaral and Fop, 2013</u>). As such, consumers are expected to be in a pre-intentional phase and increasing risk perceptions through risk communication should foster the intention to adequately dispose of pharmaceuticals leftovers.

Environmental identity refers to a part of the self-concept that is associated to the sense of connection to natural environment, that affects one's perception of the world and one's actions (<u>Clayton, 2003</u>). High environmental identity is related to more pro-environmental behaviours (<u>Lima and Branco, 2018</u>) and can be an important moderator of predictors of pro-environmental behaviour, such as intention (<u>Carfora et al., 2017</u>). Therefore environmental identity appears to be important in translating individual intentions into actions.

To sum up, our goals are 1) to explore if the perception of risks associated to pharmaceuticals in the environment might be built based on the trust that individuals have on authorities when their knowledge on this topic is limited and 2) to explore the predictive role of risk perception for proper disposal behaviour, taking into account intention and environmental identity. We will analyse if these effects are similar in three Southwestern European countries: Portugal, Spain, and France. We focused on these countries because this work is part of an Interreg-SUDOE project (Innovec'EAU) that aimed to promote transnational cooperation in southwestern Europe to help analyse and solve the presence of pharmaceuticals in the environment. These countries are under the same European strategy and directives, and share borders but, naturally, have different regulations and contexts.

1.4. Context of the current study

In the European Union, Directive 2004/27/EC (related to medicinal products for human use) introduced an obligation to implement appropriate collection schemes for unused pharmaceutical products destined for human consumption. However, it did not provide any guidelines on the implementation of those schemes, and a few studies have pointed out significant differences between countries. A survey by the European Environment Agency of the authorities responsible for pharmaceutical waste (Vollmer, 2010) illustrated a broad variety in information policy. Information channels ranged from doctors or pharmacists providing verbal information directly to a patient on the best way to dispose of these products to comprehensive information on websites, brochures, collection containers, and pharmaceuticals' packages. Back in 2010, in Spain, all routes of information were applied; in France, only three; and, in Portugal, merely one. Since then, most countries have been committed to amplifying their communication plans. In Portugal, for instance, information is being channelled by the traditional paths (websites, social media, brochures), but also through more creative channels, such as automated banking machines.

Regarding pharmaceutical collection, in all three countries, there is a nationwide level of organization and coverage, and the collection schemes are financially supported by the <u>pharmaceutical industry</u>, with pharmacies acting as collection sites. However, in France pharmacies are legally obligated to participate in these collection schemes (article 32 of law n°2007-248), whereas in Portugal and Spain pharmacies participate voluntarily. Nonetheless, in 2016, practically all pharmacies voluntarily participated (99.62% in Portugal, and 98.36% in Spain; indicators calculated based on information from VALORMED, 2016, SIGRE, 2016, and Health Market Research, 2017).

In terms of results, we searched for the latest statistics regarding the quantity of pharmaceutical residues and containers that were collected per capita in 2017, in order to have comparable indicators between countries. In Portugal and Spain, the average collection was 92 g (SIGRE, 2017, VALORMED, 2017). In France, the amount was much higher: 164 g (CYCLAMED, 2017).

To know if the results of pharmaceutical waste returned to pharmacies could be influenced by different patterns of consumption in these countries, we further searched for differences in pharmaceutical consumption. Eurostat data about self-reported use of medication was available for 2014, illustrating similar patterns in all three countries. The results were slightly below the European Union average consumption of 34.6% in non-prescribed pharmaceuticals (23.9% in Portugal, 21.9% in Spain, and 27.1% in France, Eurostat, 2014a), but above the European Union average consumption of 48.6% in prescribed pharmaceuticals (56.1% in

Portugal, 53.1% in Spain, and 52.4% in France, <u>Eurostat, 2014b</u>). This suggests that different amounts of unused pharmaceuticals returned to pharmacies should not be related with different consumption patterns.

1.5. Hypotheses

In this study we combined stage and environmental models of behaviour change to account for proper disposal of pharmaceutical products, and anticipated psychosocial effects based on country-level indicators of pharmaceutical waste separation. We expect to find:

- 1) Differences between countries in terms of knowledge, risk perception and usual discarding behaviour. Individuals from France were expected to score higher on these variables, because the amount per capita disposed in pharmacies is much higher and, therefore, knowledge, risk perception and behaviour should be congruent with this indicator (H1);
- 2) A moderation effect of knowledge in the relation between trust and risk perception, considering this is an emerging risk and people might base their assessments of risk on trust when the knowledge is limited (H2);
- 3) A moderated mediation effect, more specifically (a) an indirect effect of risk perception on behaviour through intention, as risk perception should be a highly relevant variable to motivate behaviour in the case of emerging risks, (b) with this effect being contingent on high environmental identity (H3).

2. Method

2.1. Participants

2.1.1. Sample determination and weighting

To decide the sample size adequate to test H1, we first conducted a priori power analysis that indicated that the required minimum sample size was 252 participants to have 95% power for detecting a medium sized effect, using the GPower software (Faul et al., 2007). We further ensured that participants represented the population in terms of gender (male and female), age groups, and educational level, by using nonproportionate quota sampling, a procedure that increased our sample size to 509. We collected samples with similar quotas and used a weighting adjustment based on the Eurostat information for each country. To test for H2 and H3, we used bootstrap methods that have relatively high power for detecting mediation.

2.1.2. Participants

Five hundred and nine participants from Portugal (30.8%), Spain (35.6%), and France (33.6%) responded to the survey. Qualtrics Panels was used to recruit participants in market research panels, through social media channels in the three countries. Participants provided their informed consent, were paid, and thanked for their participation. As previously mentioned, the sample was collected aiming for similar quotas for gender (50.7% were female), age groups (30.5% between 18 and 29 years old; 26.3% between 30 and 49 years old; 29.1% between 50 and 64 years old; but only 14.1% more than 65 years old), and educational level (33.6% low; 33.8% medium; and 32.6% high education).

2.2. Procedure and materials

2.2.1. Questionnaire

The questionnaire was developed in English, translated to Portuguese, Spanish, and French by native speakers, and back translated to English (see <u>supplementary material</u>). Questionnaire measured knowledge on pharmaceuticals in the environment, trust in the authorities to deal with the risk, health and environmental risk perception towards four hazards (pharmaceuticals in the environment in general, in treated wastewater, in drinking water, and in crops), environmental identity, intention to adequately discard of pharmaceuticals and usual discarding behaviour. Pharmaceuticals were presented in a general manner, we did not distinguish form of presentation (e.g., syrup, pill), or focused on classes of pharmaceuticals.

Three measures of knowledge were included. Factual knowledge was assessed by multiple response questions regarding the excretion of pharmaceuticals, how the pharmaceuticals are excreted, and the fate of excreted pharmaceuticals. This measure, adapted from Götz et al., 2019, ranges between 0, no selection of correct answers, and 10, selection of all correct answers. Respondents also had the option to respond "I do not know". Perceived knowledge was measured by the items "We would like you to rate your knowledge about pharmaceuticals in the environment / wastewater / drinking water / crops. Please use a scale of 0 to 100, where 0 means knowing nothing and 100 means knowing everything you could possibly know about this topic. Using this scale, how much do you currently know about pharmaceuticals in the environment / wastewater / fresh water / crops?". Items were averaged into a composite measure with an adequate level of internal consistency, $\alpha = 0.94$. Need for knowledge was assessed by a similar item, asking people how much knowledge they would need to adequately deal with the four risks, using the scale from 0 to 100. Items were averaged into a composite measure with an adequate level of internal consistency, $\alpha = 0.96$. Perceived knowledge and need for knowledge measures were adapted from Griffin et al. (1999) measure of information sufficiency.

Trust in risk regulation, i.e., trust that proper authorities and current regulations will manage the risk of pharmaceuticals in the environment was measured by two items created by Poortinga and Pidgeon, 2005 (e.g., "I feel confident that the responsible institutions are adequately regulating pharmaceutical waste."). The response scale ranged from 1, *strongly disagree*, to 7, *strongly agree*. Items were averaged into a composite measure with an adequate level of internal consistency, Spearman-Brown = 0.82.

Risk perception was measured by asking the level of risk of the four hazards both for the environment and health (e.g., "Do you consider that the risks of pharmaceuticals in drinking water are low or high for health?"), on a scale ranging from 1, *very low*, to 7, *very high*. The eight items were averaged into a composite measure with an adequate level of internal consistency, $\alpha = 0.90$.

Intention was adapted from adapted from Götz et al. (2019), to assess the individual's willingness to return unused or expired pharmaceutical products to the pharmacy, in order to reduce the presence of pharmaceuticals in the environment, water, and crops (e.g., "Would be willing to return unused or expired pharmaceutical products to the pharmacy at all times to reduce the presence of pharmaceuticals in water?"). The response scale ranged from 1, *yes*, to 5, *no*. These three items were averaged into a composite measure with an adequate level of internal consistency, $\alpha = 0.97$.

Environmental identity measure followed on Lima and Branco's (2018) short version of Clayton's scale (2003), which had already been validated by Olivos and Aragones (2011). The measure consisted of 5 items, such as "If I had enough time or money, I would certainly devote some of it to working for environmental causes". The response scale ranged from 1, $strongly\ disagree$, to 7, $strongly\ agree$. Items were averaged into a composite measure with an adequate level of internal consistency, $\alpha = 0.88$.

Pharmaceutical discarding behaviour was measured by asking participants about their usual behaviour of: (1) returning pharmaceuticals to the pharmacy (e.g., "I usually return my pharmaceutical residues to a pharmacy"), (2) throwing pharmaceuticals down the sink/drain, (3) throwing pharmaceuticals down the toilet, (4) throwing pharmaceuticals into undifferentiated garbage. The response scale ranged from 1, *strongly disagree*, to 7, *strongly agree*. The last three items were reverse-coded, and all items were averaged into a composite measure with an adequate level of internal consistency, $\alpha = 0.78$.

3. Results

3.1. Descriptive results and differences between countries

To test if there were differences between the three countries we used and Analysis of Variance test (ANOVA; see Mohanty and Misra, 2016). ANOVA compares the amount of variation between countries with the amount of variation within countries, generating an F value which allows to calculate the associated probability of getting a result at least as extreme as the one observed, given the null hypothesis that there are no differences between countries (the p value). The null hypothesis was rejected when the p value was smaller than 0.050 in all the conducted statistic tests. In case of differences between countries, we used the Bonferroni procedure to identify which pairs of means were statistically different. We further indicated the eta square value (η^2), which provides an estimate of the effect size of the differences between countries. In concrete, it indicates the proportion of the total variance of each study variable that is associated with the countries. The 95% Confidence Intervals [CI] for the mean of the three countries are described in the text below, to illustrate the range of values that contain the point estimates of the population. To simplify the interpretation of the differences between countries, we present the estimated mean and standard deviation of the sample, instead of CIs, in Table 1.

Table 1. Descriptive statistics and ANOVA results study variables.

M (SD)

Variable	Portugal	Spain	France
Factual knowledge	3.56(1.97) ^a	2.42(1.30)b	2.97(1.81) ^a
Perceived knowledge	38.99(23.89) ^{a,b}	36.76(28.70)a	43.05(23.28)b
Need for knowledge	40.26(31.23)a	48.47(30.78)a	61.18(26.29)b
Trust	4.54(1.40) ^{a,b}	5.11(1.57) ^a	3.94(1.55)b
Risk perception	5.62(1.08) ^a	4.95(1.55)b	5.02(1.00) ^{a,b}
Environmental risk perception	5.62(1.24) ^{a,b}	4.91(1.63) ^a	5.24(1.35)b
Health Risk perception	5.63(1.01) ^a	4.99(1.58) ^{a,b}	4.80(1.39)b
Intention	4.78 (0.74) ^a	3.88(1.45)b	4.33(0.94)a
Environmental identity	4.77(1.31) ^a	4.51(1.42) ^a	4.61(1.34) ^a
Usual behaviour	5.78 (1.47) ^{a,b}	5.37(1.53) ^a	5.75(1.39)b

Notes. Variables ranged from 1 to 7, except factual knowledge, that ranged 0 to 10, perceived knowledge and need for knowledge that ranged 0 to 100, and intention that ranged from 1 to 5. Means with different superscript letters within the same line are significantly different from each other (Bonferroni pairwise comparisons, p<.050 expect for Environmental risk perception where p=.080).

The hypothesis that respondents from France would score higher than Portugal and Spain in knowledge, risk perception and usual discarding behaviour (H1) was merely marginally corroborated. French respondents scored clearly higher than Portuguese and the Spanish respondents only in need for knowledge. For perceived knowledge, risk perception and usual behaviour, the scores of the French respondents were higher than the ones for Spanish respondents, but they were not different from the scores of the Portuguese respondents.

Individuals' factual knowledge on pharmaceuticals in the environment was low. On average, individuals selected 3 out of 10 correct answers. Factual knowledge mean was significantly lower in individuals from Spain than in individuals from Portugal and France, F(2,498) = 9.29, p < .001, 95% CI [2.74, 3.23], $\eta^2 = 0.04$. Perceived knowledge results were partially similar to factual knowledge results, with a mean result below the median point of the scale and significantly lower in individuals from Spain than in individuals from France, F(2,459) = 3.26, p = .039, 95% CI [35.74, 43.46], $\eta^2 = 0.01$. Need for knowledge mean was relatively higher than perceived knowledge but it was also below the medium point of the scale, except for the mean result of individuals from France. As stated previously, this was the only variable in which individuals from France scored higher than individuals from Portugal and Spain, F(2,459) = 13.96, p < .001, 95% CI [45.69, 54.25], $\eta^2 = 0.06$.

Trust in authorities to manage the risk of pharmaceuticals in the environment was medium. Respondents from Spain showed more trust than respondents from France, F(2,459) = 30.46, p < .001, 95% CI [4.30, 4.76], $\eta^2 = 0.12$.

Environmental and health risk perception of pharmaceuticals in the environment was medium / high. The mean of individuals from Portugal was marginally higher than the mean of individuals from Spain, F(2,442) = 2.94, p = .054, 95% CI [5.01, 5.39], $\eta^2 = 0.01$. However, when the risk perception measure was disaggregated, environmental risk perception mean was relatively higher in individuals from France than in individuals from Spain, F(2,437) = 3.83, p = .023, 95% CI [5.04, 5.48], $\eta^2 = 0.02$, and health risk perception was relatively lower in respondents from France than in respondents from Portugal, F(2,437) = 3.78, p = .023, 95% CI [4.92, 5.36], $\eta^2 = 0.02$.

Intention to adequately dispose of pharmaceuticals was high. The mean for intention was lower in respondents from Spain than in respondents from France and Portugal, F(2,430) = 10.58, p < .001, 95% CI [4.16, 4.51], $\eta^2 = 0.05$.

Environmental identity was medium and there were no differences between countries, 95% CI [4.42, 4.84].

Usual behaviour of disposal of pharmaceuticals was medium/high. This behaviour mean was lower in respondents from Spain than in those from France, F(2,442 = 3.66, p = .026, 95% CI [5.42, 5.85], $\eta^2 = 0.02$.

We further calculated the percentage of individuals that responded above the medium point of each variables to provide a deeper understanding of the differences between countries (Fig. 1). These results are consistent with Table 1. We highlight the low percentage of individuals with factual and perceived knowledge above the average, particularly in Spain. In Portugal and France, most respondents did not trust the current risk regulations, considered that pharmaceuticals in the environment had a medium-high risk to the environment and to health, intended to properly dispose of pharmaceuticals, and reported that they usually dispose of pharmaceutical products properly.

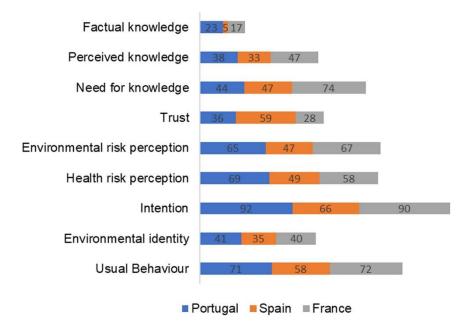


Fig. 1. Percentage of individuals that scored above the average point of the scales.

3.2. Moderation of knowledge on the relation between trust and risk perception

To test for moderation and conditional processes, we used the PROCESS macro for SPSS version 3 (<u>Hayes, 2018</u>), which is based on ordinary least square regression and path analysis. The number of bootstrap samples for percentile bootstrap confidence intervals was 5000. To reduce country-type influences, the country variable was dummy coded and entered in all analyses, as detailed below.

The interpretation of the models was based on a) the percentual amount of variability of risk perception explained by the model, b) the F value, which is the result of a test where the null hypothesis is the <u>regression coefficient</u> of the model (trust), indicated by b, is equal to zero, and c) the associated p value. The value of b, the unstandardized regression coefficient, is also presented and indicates the size and direction of the effect that trust is having on risk perception, as well as the t statistic that allows calculating the p value and the 95% CI, the range of values that likely contain the true value of b (Hayes, 2018).

For moderation analyses, all variables were mean-centered. To explore the nature of the moderation effects that were found, we used simple slope analysis that probe the effects of trust on risk perception within low values of knowledge (one standard deviation below its mean), average values of knowledge (mean), and high values of knowledge (one standard deviation above the mean knowledge).

We entered both knowledge and country as moderators of the relation between trust and risk perception. We ran three separate models, one for each knowledge variable (factual, perceived, needed), to explore differences due to the type of knowledge that was being assessed. We expected a moderation effect of knowledge in the relation between trust and risk perception (H2), but our hypothesis was not corroborated. The relation between risk perception and trust was always positive, and only need for knowledge about risks moderated this effect. Therefore, we will focus on this model.

The variable country did not interact with trust to influence risk perception. Hence, we removed this variable to preserve a maximum degree of freedom and facilitate the interpretation of the model. This model significantly explained a small amount of the variability of risk perception (11%; F(3,460) = 18.31; p < .001). The relation between trust and risk perception was positive, b = 0.18, t = 4.64, p < .001, 95% CI [0.10, 0.26], andthe moderation effect was negative, b = -0.00, t = -2.27, p = .023, 95% CI [-0.01, -0.00], meaning that increases in the need for knowledge decreased the positive effect of trust on risk perception. In this regard, simple slope analyses show that the correlation between trust and risk perception was stronger when need for knowledge was lower (in concrete, one standard deviation below average) b = 0.25, t = 4.89, p < .001, 95% CI [0.16, 0.36] and it was lower and marginally significant when need for knowledge was higher (one standard deviation above average) b = 0.10, t = 1.94, p = .052, 95% CI [-0.00, 0.20]. The three regression lines representing the correlation between trust and risk perception when the need for knowledge was high, average, and low are illustrated in Fig. 2.

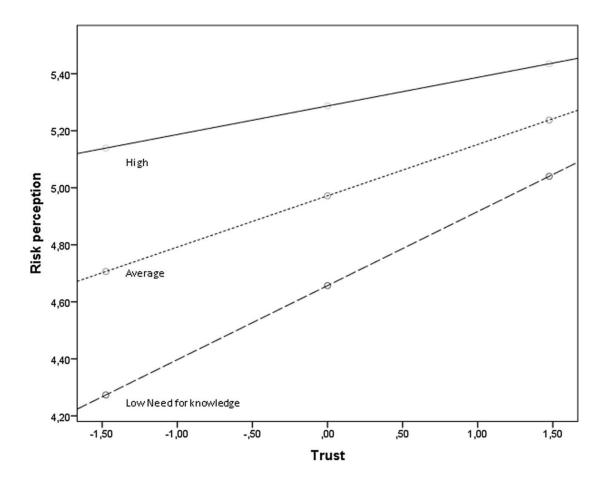


Fig. 2. Moderation effect of need for knowledge to deal with risks on the relation between trust in risk regulation and environmental and health risk perception of pharmaceuticals in the environment (mean-centered values).

As such, higher trust in the authorities to manage the risk of pharmaceuticals in the environment was related to higher risk perception of pharmaceuticals in the environment. The moderation of need for knowledge means that the relation was stronger when people had low need for knowledge to adequately deal with hazards. This result strengthens the concept that the relation between trust and risk perception is heuristically driven. In line with the model of risk information seeking (Griffin et al., 1999) low need for knowledge should be associated with less motivation and less systematic processing of information. Additional correlation analyses also illustrated that respondents with less need for knowledge had lower factual knowledge, r = 0.18, p < .001, and lower perceived knowledge, r = 0.56, p < .001.

3.3. Moderated mediation model of risk perception on behaviour

We tested if there was an indirect effect of risk perception on behaviour through intention, and if the effect of intention on behaviour was moderated by environmental identity (see this conceptual model in <u>Fig. 3</u>). We further controlled if this model held for all countries by adding dummy variables to the three mediation paths. No country effects emerged, hence we dropped the country dummy variables to preserve a maximum degree of freedom.

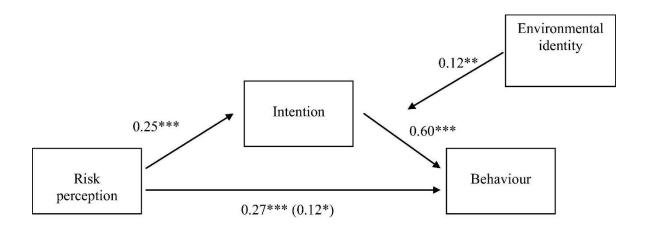


Fig. 3. Moderated mediation model: indirect effect of environmental and health risk perception on proper disposal behaviour through intention, moderated by environmental identity. *p < .050, **p < .010, ***p < .001.

The overall adequacy of the model, the regression coefficients (b) and their associated statistics (t, p), and the nature of the moderation were the interpreted as described in the previous section. To interpret the mediation analysis, we considered the total effect of risk perception on the disposal of pharmaceuticals, and the direct effect of risk perception on the disposal of pharmaceuticals when included the moderated mediation effect of intention and environmental identity. It is assumed that mediation occurs when the direct effect is smaller than the total effect, considering that the introduction of the moderated mediation variables explained the relation between risk perception and the disposal of pharmaceuticals. Our hypothesis was corroborated.

Results illustrate that the moderated mediation model explained a moderate amount of the variability of disposal behaviour (31%; F(4,458) = 51.99, p < .001). A test of linear moderated mediation (see Hayes, 2018) supported the occurrence of moderated mediation as a 95% bootstrap confidence interval for an index of moderated mediation did not cross zero, 95% CI [0.01, 0.06]. As illustrated in Fig. 3, environmental and health risk perception of pharmaceuticals positively influenced proper disposal behaviour, $b_{\text{total effect}} = 0.27$, t = 5.58, p < .001, 95% CI [0.17, 0.36]. Risk perception also influenced proper disposal of pharmaceuticals, $b_{\text{direct effect}} = 0.12$, t = 2.52, p = .012, 95% CI [0.02, 0.22], through its positive effect on intention to properly dispose of them, $b_{\text{risk percetion}\rightarrow \text{intention}} = 0.25$, t = 6.16, p < .001, 95% CI [0.18, 0.33]. Intention highly influenced disposal behaviour, $b_{\text{intention} \rightarrow \text{behaviour}} = 0.60$, t = 12.68, p < .001, 95% CI [0.50, 0.70], and this effect was strengthened by environmental identity, $b_{\text{interaction}} = 0.12$, t = 2.96, p = .003, 95% CI [0.02, 0.21]. Simple slope analyses showed that the effect was stronger when environmental identity was higher (one standard deviation above average), b = 0.75, t = 10.28, p < .001, 95% CI [0.60, 0.89], also occurring when it was lower (one standard deviation below), b = 0.45, t = 6.94, p < .001, 95% CI [0.32, 0.58]. The three regression lines representing the correlation between intention to dispose and proper disposal behaviour when the environmental identity was high, average, and low are illustrated in Fig. 4. Respondents with higher environmental identity acted more on their intention than participants with lower environmental identity.

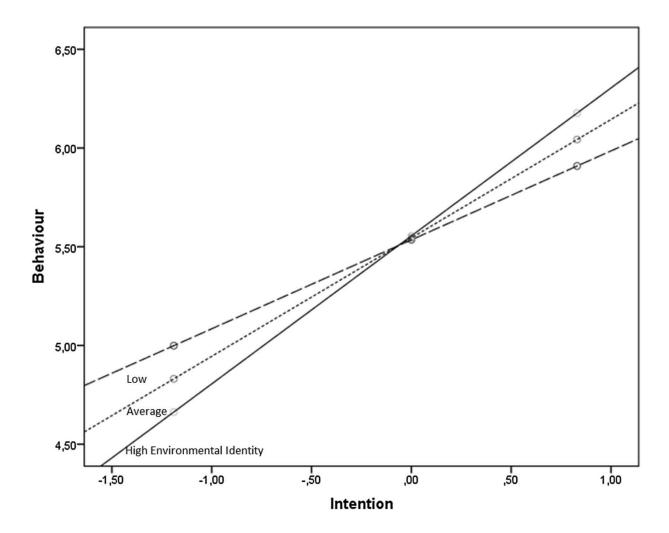


Fig. 4. Moderation effect of environmental identity on the relation between intention and proper disposal behaviour (mean-centered values).

To sum up, as expected, there was a direct effect of risk perception on behaviour, and an indirect effect through intention. This indirect effect was dependent on environmental identity being stronger when environmental identity is higher.

4. Discussion

The presence of pharmaceuticals in the environment is a transboundary environmental problem. We analysed data from 3 geographically close countries in Southwestern Europe (Portugal, Spain, and France). Although water and soil contamination by pharmaceutical residues does not recognize borders, these countries have different regulations and psychosocial contexts that should naturally be considered when managing this problem. The discussion will be organized according to the hypothesis stated.

4.1. Perceptions on pharmaceuticals in Portugal, Spain, and France

As expected, individual knowledge on pharmaceuticals in the environment, both factual and perceived, was low, and individuals reported needing more knowledge to deal effectively with the risks associated with this problem. Environmental and health risk perception was medium

/ high, and individuals moderately trusted that authorities were adequately managing this risk. Regarding their individual actions, the intention to properly dispose of pharmaceutical residues was slightly higher than the reported behaviour of proper disposal. Taken together, these results suggest the need to communicate about the risks associated with pharmaceuticals in the environment, what the authorities are doing to manage this risk, and what each individual can/should do. Differences between countries signal different needs, such as (1) a higher need for knowledge dissemination in Spain, where factual knowledge is lower, and in France, where individuals directly report the need for knowledge, and (2) the need to differentiate between environmental and human health risks in Portugal and Spain.

Data on the quantity of pharmaceuticals returned to pharmacies per capita led us to hypothesize that in France, where this amount was almost twice as large, individuals would have a higher risk perception and would report more proper discarding behaviours. However, this was not the case. French respondents only showed a higher need for knowledge. Regarding the remaining variables, respondents from France and Portugal tended to have similar results (factual and perceived knowledge, intention, behaviour). This illustrates that, although the objective quantity of pharmaceuticals recycled in France is much higher, it might be oversimplifying to assume that this should be reflected in differences in risk perception and self-reported behaviour. Other contextual variables that have not been identified are likely at stake.

4.2. On the relation between trust and risk perception

Research in risk perception has long evidenced that when individuals have low knowledge on a hazard, they tend to base their risk perception in trust assessments, typically leading to a strong negative relation between trust and risk perception. Our results were congruent with the heuristic nature of this process: the relation between trust and risk perception was stronger when people had a lower need for knowledge. However, this relation was not strong, and it was positive. As Viklund (2003) illustrates, the relations between trust and risk perception might be stronger in countries where trust is higher, risk perception is lower and risks are more dreadful. In our study, trust was medium, and risk perception was medium / high. It is also likely that the risk of pharmaceuticals in the environment is not highly dreadful, considering that its consequences are not severe and are delayed in time. Therefore, it is comprehensible that we did not find a strong relation and that only 10% of the variability of risk perception was explained. The risk perception of pharmaceuticals in the environment might also be explained by other types of variables, such as the psychometric paradigm dimensions.

The fact that we found a positive relation between trust in risk regulation and risk perception is intriguing. This result suggests that the more people trust that the risk of pharmaceuticals in the environment is being regulated, the higher will be their risk perception. Our interpretation is that, in case of this unknown risk, and of these countries, individuals might not have relied on trust as a heuristic to minimize risk but as a heuristic to amplify it. In other words, they might have considered that if their authorities were adequately regulating these new and unknown risks it would be because pharmaceuticals did in fact pose a considerable risk for the environment and health. Indeed, most research on the relation between trust and risk perception focus on known risks.

Causality could also be an issue. Therefore, we examined an alternative moderation model, entering risk perception as the independent variable and trust as the dependent variable. In this

model, the moderation effect was no longer significant, and the amount of variability explained was much smaller (4%). Nevertheless, this should not be discarded. Even a more complex bi-directional causation may apply, and research is needed to elucidate the cause and effect question regarding these variables. For instance, using an experimental design with repeated measures to inform about micropollutants in drinking water, Tobias (2016) illustrated that trust could have both positive and negative effects, and it could be both a determinant and a consequence of risk perception.

These questions highlight the need to understand the relation between trust in risk regulation and risk perception at a cross-national level, and how it applies to emergent and not highly dreadful environmental problems.

4.3. Explaining proper disposal of pharmaceuticals

To explore the proper disposal of pharmaceutical leftovers we built upon inputs from a) stages models of behaviour change, which highlighted the importance of risk perception to motivate intention and behaviour when awareness about the problem is low, and b) models to promote pro-environmental behaviour, which indicate that high environmental identity promotes pro-environmental behaviours. Based on these inputs, we designed a moderated mediation model that explained a considerable amount of self-reported proper disposal behaviour. This model suggested that this behaviour could be promoted by communicating about the risks of pharmaceuticals in the environment, not only focusing on health issues, but also by making environmental identity more salient. Research has indicated that one simple way to do this is by reminding people how often they behaved pro-environmentally in the past (Cornelissen et al., 2008, Fanghella et al., 2019, Van der Werff et al., 2014). An avenue for future research would be to experimentally explore the impact of nudging environmental identity on prompting proper disposal behaviour.

4.4. Limitations

When interpreting these findings, there are some limitations to consider. First, the cross-sectional nature of the evidence constrains interpretation of causes and effects, even if it seems reasonable to assume them based on the substantial body of research in stage models of behaviour change and risk perception. Secondly, the participants responded to the survey online, which means that digitally excluded individuals did not have the chance to participate and their responses might not be represented in the results. Thirdly, we did not consider the occupation of participants. This might have influenced the results if individuals with occupations in this area misrepresented the population.

5. Conclusion

Pharmaceuticals in the environment pose an emerging environmental problem, and, in recent years, numerous initiatives are being launched to decrease their presence. This study's main goal was to contribute towards an understanding of proper disposal procedures of unused pharmaceuticals, by exploring the role of risk perception, which is particularly relevant for emerging risks, in Southwestern European countries (Portugal, Spain, and France). Contrarily to our expectations, trust on risk regulation was not a strong predictor of risk perception, and higher trust was actually correlated with higher risk perception. As such, it might be important for risk regulators to communicate about what is being done to manage the risk of pharmaceuticals.

In addition, this work contributes to improve risk management processes by illustrating that risk perception is related to individual's proper disposal behaviour. This suggests that the amount of pharmaceutical leftovers returned to the pharmacies should increase with the enhancement of communication about pharmaceuticals and the risks they pose. Furthermore, it is illustrated that the effect of risk perception is explained by an increase in individuals' intention to properly dispose of pharmaceuticals, and that when they have higher environmental identity, intention is easily translated into proper behaviour. This suggests that making environmental identity salient could further contribute to increase the amount of pharmaceutical leftovers returned to the pharmacies.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

Adeel et al., 2017

M. Adeel, X. Song, Y. Wang, D. Francis, Y. Yang

Environmental impact of estrogens on human, animal and plant life: A critical review

Environ. Int., 99 (2017), pp. 107-119, 10.1016/j.envint.2016.12.010

• Amaral and Fop, 2013

M.J. Amaral, L. Fop. Unused Pharmaceuticals Where Do They End Up?, Health Care Without Harm, Brussels (2013)

• Bamberg, 2013

S. Bamberg. Changing environmentally harmful behaviors: A stage model of self-regulated behavioral change. J. Environ. Psychol., 34 (2013), pp. 151-159, 10.1016/j.jenvp.2013.01.002

• Brodin et al., 2014

T. Brodin, S. Piovano, J. Fick, J. Klaminder, M. Heynen, M. Jonsson. Ecological effects of pharmaceuticals in aquatic systems—impacts through behavioural alterations. Philos. Trans. R. Soc. Lond. B. Biol. Sci., 369 (2014), p. 20130580, <u>10.1098/rstb.2013.0580</u>

• Carfora et al., 2017

V. Carfora, D. Caso, P. Sparks, M. Conner. Moderating effects of pro-environmental self-identity on pro-environmental intentions and behaviour: A multi-behaviour study. J. Environ. Psychol., 53 (2017), pp. 92-99, 10.1016/j.jenvp.2017.07.001

• Carlson et al., 2009

J.P. Carlson, L.H. Vincent, D.M. Hardesty, W.O. Bearden. Objective and subjective knowledge relationships: a quantitative analysis of consumer research findings. J. Consum. Res., 35 (2009), pp. 864-876, 10.1086/593688

• Clayton, 2003

S. Clayton. Environmental identity: A conceptual and operational definition. S. Clayton, S. Opotow (Eds.), Identity and the Natural Environment. The Psychological Significance of Nature, MIT Press, Cambridge (2003), pp. 45-65

• Cornelissen et al., 2008

G. Cornelissen, M. Pandelaere, L. Warlop, S. Dewitte. Positive cueing: Promoting sustainable consumer behavior by cueing common environmental behaviors as environmental. Int. J. Res. Mark., 25 (2008), pp. 46-55, 10.1016/j.ijresmar.2007.06.002

• CYCLAMED, 2017

CYCLAMED, 2017. Chiffres de collecte des MNU - Médicaments périmés ou non = retour en pharmacie - Cyclamed [WWW Document]. URL https://www.cyclamed.org/association/chiffres (accessed 7.30.18).

• Daughton, 2016

C.G. Daughton. Pharmaceuticals and the Environment (PiE): Evolution and impact of the published literature revealed by bibliometric analysis. Total Environ. Sci. (2016), 10.1016/j.scitotenv.2016.03.109

• Dias-Ferreira et al., 2016

C. Dias-Ferreira, S. Valente, J. Vaz. Practices of pharmaceutical waste generation and discarding in households across Portugal. Waste Manag. Res., 34 (2016), pp. 1006-1013, 10.1177/0734242X16639388

• Earle, 2010

T.C. Earle. Trust in risk management: a model-based review of empirical research. Risk Anal., 30 (2010), pp. 541-574, 10.1111/j.1539-6924.2010.01398.x

• European Commission, 2019

European Commission, 2019. European Union Strategic Approach to Pharmaceuticals in the Environment. [WWW Document]. URL https://ec.europa.eu/environment/water/water-dangersub/pdf/strategic approach pharmaceuticals env.PDF (accessed 4.12.19).

• Eurostat, 2014a

Eurostat, 2014a. Self-reported use of non-prescribed medicines by sex, age and educational attainment level [WWW Document]. URL

http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=hlth_ehis_md2e&lang=en (accessed 7.30.18).

• Eurostat, 2014b

Eurostat, 2014b. Self-reported use of prescribed medicines by sex, age and educational attainment level [WWW Document]. URL

http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=hlth_ehis_md1e&lang=en_(accessed 7.30.18).

• Fanghella et al., 2019

V. Fanghella, G. d'Adda, M. Tavoni. On the use of nudges to affect spillovers in environmental behaviors

Front. Psychol., 10 (2019), <u>10.3389/fpsyg.2019.00061</u>

• Fatokun et al., 2011

O. Fatokun, A.W. Chang, W.N. Ng, T. Nair, V. Balakrishnan. Unused and expired medications disposal practices in the community: a cross-sectional survey in Cheras, Malaysia. Arch. Pharm. Pract., 2 (2011), pp. 82-83

• Faul et al., 2007

Franz Faul, Edgar Erdfelder, Albert-Georg Lang, Axel Buchner. G*Power 3: A flexible statistical power analysis program for the social, behavioral, and biomedical sciences. Behav. Res. Methods, 39 (2) (2007), pp. 175-191, 10.3758/BF03193146

• Götz et al., 2019

Konrad Götz, Audrey Courtier, Melina Stein, Linda Strelau, Georg Sunderer, Rodrigo Vidaurre, Martina Winker, Benoit Roig. Risk perception of pharmaceutical residues in the aquatic environment and precautionary measures. Management of Emerging Public Health Issues and Risks, Elsevier (2019), pp. 189-224, 10.1016/B978-0-12-813290-6.00008-1

• Green et al., 2015

C. Green, J. Brian, R. Kanda, M. Scholze, R. Williams, S. Jobling. Environmental concentrations of antiandrogenic pharmaceuticals do not impact sexual disruption in fish alone or in combination with steroid oestrogens. Aquat. Toxicol., 160 (2015), pp. 117-127, 10.1016/j.aquatox.2014.12.022

• Griffin et al., 1999

R.J. Griffin, S. Dunwoody, K. Neuwirth. Proposed model of the relationship of risk information seeking and processing to the development of preventive behaviors. Environ. Res., 80 (1999), pp. S230-S245, 10.1006/enrs.1998.3940

• Hayes, 2018

A.F. Hayes. Introduction to mediation, moderation, and conditional process analysis: a regression-based approach. Guilford Press, London (2018)

• Kümmerer, 2010

K. Kümmerer. Pharmaceuticals in the environment. Annu. Rev. Environ. Resour., 35 (2010), pp. 57-75, 10.1146/annurev-environ-052809-161223

• Lacorte et al., 2017

S. Lacorte, S. Luis, C. Gómez-Canela, T. Sala-Comorera, A. Courtier, B. Roig, A.M. Oliveira-Brett, C. Joannis-Cassan, J.I. Aragonés, L. Poggio, T. Noguer, L. Lima, C. Barata, C. Calas-Blanchard. Pharmaceuticals released from senior residences: occurrence and risk evaluation. Environ. Sci. Pollut. Res., 1–12 (2017), 10.1007/s11356-017-9755-1

• Lima and Branco, 2018

M.-L. Lima, C. Branco. Recycling for my neighbourhood? Using place identity and social norms to promote pro-environmental behaviour / ¿Reciclar para mi barrio? Empleando la identidad de lugar y las normas sociales para fomentar el comportamiento pro-ambiental. Psyecology, 9 (2018), pp. 1-32, 10.1080/21711976.2017.1412574

• Martinez, 2009

J.L. Martinez. The role of natural environments in the evolution of resistance traits in pathogenic bacteria. Proc. Biol. Sci., 276 (2009), pp. 2521-2530, 10.1098/rspb.2009.0320

• McEachran et al., 2017

A.D. McEachran, D. Shea, E.G. Nichols. Pharmaceuticals in a temperate forest-water reuse system Sci. Total Environ., 581–582 (2017), pp. 705-714, 10.1016/j.scitotenv.2016.12.185

• Mohanty and Misra, 2016

B. Mohanty, S. Misra. Statisitics for behavioural and social science. SAGE Texts (2016)

• Olivos and Aragonés, 2011

P. Olivos, J.I. Aragonés. Propiedades psicométricas de la Escala de Identidad Ambiental (EID)

Psyecology, 2 (2011), pp. 15-24, <u>10.1174/217119711794394671</u>

• Poortinga and Pidgeon, 2005

W. Poortinga, N.F. Pidgeon. Trust in risk regulation: cause or consequence of the acceptability of GM food?. Risk Anal., 25 (2005), pp. 199-209, 10.1111/j.0272-4332.2005.00579.x

• Siegrist and Cvetkovich, 2000

M. Siegrist, G. Cvetkovich. Perception of hazards: the role of social trust and knowledge. Risk Anal., 20 (2000), pp. 713-720, 10.1111/0272-4332.205064

• SIGRE, 2017

SIGRE, 2017. El sistema SIGRE en cifras.Laboratorios, farmacias, almacenes, recogida-Sigre [WWW Document]. URL https://www.sigre.es/sigre/cifras/ (accessed 8.10.18).

• Tobias, 2016

R. Tobias. Communication about micropollutants in drinking water: effects of the presentation and psychological processes. Risk Anal., 36 (2016), pp. 2011-2026, 10.1111/risa.12485

• VALORMED, 2017

VALORMED, 2017. Relatório de Actividades - Resumo. Algés. [WWW Document]. URL http://new.valormed.pt//assets/stores/1041/userfiles/Resumo_Relat%C3%B3rio%20de%20Actividades%202017l.pdf (accessed 7.30.18).

• Van der Werff et al., 2014

E. Van der Werff, L. Steg, K. Keizer. I Am What I Am, by looking past the present. Environ. Behav., 46 (2014), pp. 626-657, 10.1177/0013916512475209

• Viklund, 2003

M.J. Viklund. Trust and risk perception in Western Europe: a cross-national study. Risk Anal. (2003), p. 23, 10.1111/1539-6924.00351

• Visschers and Siegrist, 2008

V.H. Visschers, M. Siegrist. Exploring the triangular relationship between trust, affect, and risk perception: a review of the literature. Risk Manag., 10 (2008), pp. 156-167, 10.1057/rm.2008.1

• World Health Organization, 2012

World Health Organization, 2012. Pharmaceuticals in drinking-water. Geneva. [WWW Document]. URL

http://apps.who.int/iris/bitstream/handle/10665/44630/9789241502085_eng.pdf;jsessionid=192C36F1F8F2D6DC13A186B943CAB084?sequence=1 (accessed 7.30.18).

• Vollmer, 2010

G. Vollmer. Disposal of Pharmaceutical Waste in Households – A European Survey, in: Green and Sustainable Pharmacy, Springer Berlin Heidelberg, Berlin, Heidelberg (2010), pp. 165-178, 10.1007/978-3-642-05199-9 11

• Wu et al., 2015

X. Wu, L.K. Dodgen, J.L. Conkle, J. Gan. Plant uptake of pharmaceutical and personal care products from recycled water and biosolids: a review. Sci. Total Environ., 536 (2015), pp. 655-666, 10.1016/J.SCITOTENV.2015.07.129