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DOI
10.1016/j.scitotenv.2022.155530

Publication date
2022

Document Version
Final published version

Published in
Science of the Total Environment

License
CC BY-NC-ND

Citation for published version (APA):
Development of chemical emission scenarios using the Shared Socio-economic Pathways

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HIGHLIGHTS

• Emission scenarios for chemical emissions to water with SSPs
• Veterinary pharmaceuticals show the highest increase in emission of 30%.
• Ibuprofen shows 240% increase in SSP3.

GRAPHICAL ABSTRACT

ABSTRACT

The widespread use of chemicals has led to significant water quality concerns, and their use is still increasing. Hence, there is an urgent need to understand the possible future trends in chemical emissions to water systems. This paper proposes a general framework for developing emission scenarios for chemicals to water using the Shared Socio-economic Pathways (SSPs) based on an emission-factor approach. The proposed approach involves three steps: (i) identification of the main drivers of emissions, (ii) quantification of emission factors based on analysis of publicly available data, and (iii) projection of emissions based on projected changes in the drivers and emission factors. The approach was tested in Europe for five chemical groups and on a national scale for five specific chemicals representing pharmaceuticals, pesticides, and industrial chemicals. The resulting emission scenarios show widely diverging trends of increased emissions by 240% for ibuprofen in SSP3 (regional rivalry) to a 68% decrease for diclofenac in SSP1 (sustainable development) by 2050. While emissions typically decrease in SSP1, they follow the historical trend in SSP2 (middle-of-the-road scenario) and show an increase in the regional rivalry scenario SSP3 for most selected chemicals. Overall, the framework allows understanding of future chemical emissions trends as a function of the socio-economic trends as captured in the SSPs. Our scenarios for chemical emissions can thus be used to model future aqueous emissions to support risk assessment. While the framework can be easily extended to other pharmaceuticals and pesticides, it heavily leans on the availability and quality of historical emission data and a detailed understanding of emission sources for industrial chemicals.

Keywords:
Chemical pollution
Shared Socio-economic Pathways (SSPs)
Scenarios
Water emissions
Integrated assessment model

1. Introduction

Chemicals are an essential part of modern daily life and are used in various ways, such as plant protection products, industrial chemicals, pharmaceuticals, and personal care products. Over 350,000 chemicals and
mixtures of chemicals are registered worldwide for production and use (Wang et al., 2020). These chemicals and their transformation products can enter the environment and water systems during production, use and waste stages. About 220 million tons of the synthetic compounds used in industrial and consumer products are emitted into natural systems annually (Cribb, 2017). Agriculture accounts for 2 million tons of pesticide use each year (Sharma et al., 2019). As a result, chemical pollution poses a significant threat to freshwater quality and ecology (Lemm et al., 2021; Malaj et al., 2014).

The production and production of chemicals are rapidly increasing. Since 1970, the chemical industry (including the pharmaceutical and pesticide sectors) has raised its net worth more than twice as quickly compared to other global change variables such as CO₂ emissions and global population (Bernhardt et al., 2017). This increase is expected to continue, with projections showing a doubling of the global chemical industry’s net value from 2017 to 2030 (UNEP, 2019). A range of factors drives this upsurge. Increases in crop production, for instance, typically lead to more intensive use of pesticides (Mateo-Sagasta et al., 2018). The use of pharmaceuticals generally increases with the share of the elderly population (Schwabe and Pfaffrath, 2015). Furthermore, rising economic activity also leads to a general increase in the use of chemicals such as heavy metals, micropollutants, plasticisers, flame retardants, and biocides. Urbanisation can further lead to an increased load of untreated stormwater run-off containing different urban pollutants (Bunke et al., 2019). Considering the ongoing socio-economic developments driving increased consumption and production of chemicals combined with the sensitivity of aquatic ecosystems to chemical pollution (Posthuma et al., 2020), a better understanding of possible trends in emissions of chemicals to water systems is urgently needed to quantify the impacts of these emissions (Cousins et al., 2019).

Scenarios are often used in environmental assessments to understand possible future changes and long-term consequences of anthropogenic change and their mitigation options (Kriegler et al., 2012). Scenarios have been broadly applied to understand various environmental issues such as global environmental change (Raskin, 2004), climate change (IPCC, 2001, 2007), water supply and demand (Cosgrove and Rijsberman, 2000), greenhouse gas emissions (Nakicenovic et al., 2000), and air pollution (Rao et al., 2017). Especially for climate and air pollution, thousands of scenarios have been published, and they have successfully supported the environmental policy, such as in the case of the Paris Agreement for climate change (van Beek et al., 2020) and the European policies for air pollution. Remarkably, scenarios for emissions of chemicals to surface water are totally absent. A key reason is that developing scenarios for future chemical pollution is relatively complex, among others, due to many chemicals being used for various purposes and the complexity of the chemical supply and use chain. Although maintaining good water quality is formulated as a policy goal at the global scale (e.g. SDG 6.3) and at the European scale (Water Framework Directive), accurate and comprehensive monitoring of all chemical emissions is currently lacking. The resulting limited availability of reliable and publicly available data on historical emissions hampers the proper assessment of present emissions of chemicals to water as a basis for scenario development.

Despite the limitations in data availability, this paper explores how scenarios for future chemical emissions to water could be developed. This is quite urgent given the serious risks to water quality. The paper proposes a methodological framework for how such scenarios could be developed and makes an initial effort to understand the complex chemical emissions to surface waters by exploring possible outcomes for different groups of chemicals and selected chemicals in Europe. These applications are mostly illustrative in identifying further research needs. In the end, the framework should answer how chemical emissions change under the influence of socio-economic change in the future? The framework simplifies the chemical emissions based on the source, driver, and pathway system. The method is based directly on the methods used by the climate change and the air pollution community for their emission scenarios (US EPA, 1995; van der Poel and Bakker, 2002; Stehfest et al., 2014; Rao et al., 2017).

The emission scenarios for chemicals are based on the Shared Socio-economic Pathways (SSPs) (O’Neill et al., 2015), a set of scenarios describing the future societal development pathways over the 21st century on the global scale. The SSPs consist of qualitative storyline and associated quantification of key scenario drivers (O’Neill et al., 2014). The storylines describe five alternative development pathways that include sustainable development (SSP1), middle-of-the-road development (SSP2), regional rivalry (SSP3), inequality (SSP4) and fossil-fuelled development (SSP5). The narratives consist of qualitative descriptions of future demographics, human development, economy and lifestyle, policies and institutions, technology, and environment and natural resources. The quantitative element includes projections of key scenario drivers such as population growth, urbanisation, economic and technological development, and land-use change. These drivers are estimated with the help of spatially and temporally explicit Integrated Assessment Models (IAM), such as the Integrated Model to Assess the Global Environment (IMAGE) (Stehfest et al., 2014).

The chemical emission scenario framework proposed in the paper projects emissions based on SSP scenarios for the underlying activities leading to chemical use and emission. To understand the overall changes in emissions of chemicals, the emission scenarios framework was first tested on five groups of chemicals: pharmaceuticals, veterinary pharmaceuticals, plant protection products, industrial chemicals and biocides. Then we elaborated the framework in three steps to a set of five specific chemicals representing pharmaceuticals (ibuprofen and diclofenac), pesticides (terbuthylazine) and industrial chemicals (cadium and diethyhexyl phthalate) focusing on Europe. Firstly, we identified key drivers of emissions of chemicals in Europe. Secondly, historical data on emissions were used to understand the trends in activity levels and emission factors. Thirdly, the emission scenarios for chemicals for 2050 were developed using SSP drivers from the IMAGE model as an input to the empirical emission models. Finally, the merits and limitations of applying the emission scenario framework to other chemicals were discussed.

2. Methods and data

2.1. Emission scenario framework

The emission scenario framework (Fig. 1) is based on an emission-factor approach. Here, we describe time and spatially specified future chemical emissions as a (summed) product of the activity causing these emissions and an emission factor (EF) (US EPA, 1995; Stehfest et al., 2014).

Step 1: Identification of activity

The activity indicator reflects the (a series of) specific emissions sources of chemicals. The activity indicator is directly or indirectly related to socio-economic development indicators included in the SSP scenarios. The selection of activity indicators is based on the main emission sources of the chemical from literature review, data analysis or expert judgment. The number of underlying activity indicators differs per chemical/group and should sum up to total emissions.

Step 2: Historical analysis

Emission factors (EF) over a specified past period and space for a specified (summed) activity/ies can be derived from the ratio of the emission data and the (summed) activity indicator(s). These underlying activities need to be consistent in historical and future analysis (see Eq. (1)):

\[
EF_{\text{future}}(r, i) = \frac{\text{Emissions}_{\text{future}}(r, i) - \text{baseline}_{\text{future}}(r, i)}{\text{Activity}_{\text{baseline}}(r, i)}
\]

\[
(1)
\]

Emissions_{baseline}(r, i) indicates the emission of specific chemical in the historical period in region r, and for sector i. Activity_{baseline}(r, i) indicates activities influencing emissions such as population, agricultural area and production over the historical period. EF_{baseline}(r, i) are the emission factor specific for that activity for the same period.

Step 3: Future analysis

The EF_{future}(r, i) are extrapolated based on EF_{baseline}(r, i) derived from past emission data or comparisons across countries to upscale on a continental
scale. Changes in EF can be a function of other socio-economic variables used in scenario development.

$$\text{Emissions}_{\text{future}(r,i)} = \text{Activity}_{\text{future}(r,i)} \times \text{EF}_{\text{future}(r,i)}$$  \hspace{2cm} (2)

where, \(\text{Emissions}_{\text{future}(r,i)}\) represent the emission of a specific chemical in the future, \(\text{Activity}_{\text{future}(r,i)}\) – Activities influencing emissions over the future time period and are projections of \(\text{Activity}_{\text{base}(r,i)}\). The temporal and spatially specific development of both Activity and EF can be described using scenarios for the underlying drivers, such as demography, economic and technological developments and policies.

2.2. Application of the framework for chemicals and coupling to SSP key quantitative drivers

We first applied the emission scenario framework for groups of chemicals, i.e. pharmaceuticals, veterinary pharmaceuticals, plant protection products, industrial chemicals and biocides, as used in Europe for indications of possible trends and to test the framework. Each group of chemicals was coupled to relevant SSP activity indicators based on their use in specific sectors (Fig. 2). We did not include change climate drivers as they can primarily influence the fate of chemicals. For simplicity, we assumed that EFs are constant investigating the possible change in chemical emission without any change in technology or legislation and implying that changes in activity indicators solely drive emissions. This assumption is not necessarily correct, and further in the article, we explore the impact of changing EF factors for the SSPs. However, here the objective was to provide insights into potential emission trends where emission data are extremely sparse.

Pharmaceuticals are used mainly in household and healthcare facilities (Kümmerer, 2008; Woehler et al., 2020) and are coupled to the human population size based on their sales and use (Heberer and Feldmann, 2005; Boxall et al., 2012). Veterinary pharmaceuticals are used as anti-infectives and additives for livestock health, nutrition, reproduction, and productivity (Clement et al., 2019; Kaczala and Blum, 2016). Cattle production contributes to major veterinary pharmaceuticals, followed by poultry, pig, and sheep production (Kaczala and Blum, 2016; Kümmerer, 2008). Hence, veterinary pharmaceutical emissions are linked to total animal production (Doelman et al., 2018) as an activity indicator. We did not include veterinary pharmaceuticals used for pets and aquaculture due to a lack of data. Plant protection products (commonly known as pesticides) are used in agriculture and are coupled to cropland areas (Popp et al., 2017). Biocides are used as disinfectants, preservatives and pest control under different product types (ECHA, 2021), applied in healthcare, food production, consumer products, animal husbandry, and wastewater treatment (SCENIHR, 2009). By comparing the different biocidal product types and applied sectoral use, the assumption was made that all emissions would end up in the wastewater. Therefore, biocide emissions were coupled to the total population accounting for both treated and untreated wastewater (Fig. 2).

Industrial chemicals constitute a large chemical group and are analysed based on their sub-groups, including heavy metals, chlorinated organic substances, other organic substances and inorganic substances (European Environment Agency, 2019). The four sub-groups are selected as they are used in the standard classification of industrial chemicals according to mental Agency (EEA) and European Pollutant Release and Transfer Register (EPRTR). Municipal and Industrial Wastewater Treatment Plants (WWTP) contribute significantly to the emissions of heavy metals and organic chemical substances. We linked these to the change in total population (KC and Lutz, 2017). Pulp, paper and wood industries contributed to the highest emissions of chlorinated organic chemical substances and are connected to paper production (Roorda and Neelis, 2006) as a suitable indicator. The chemical industry contributes to emissions of inorganic chemicals, coupled with chemical demand as an SSP driver. Energy supply and iron and steel industries had a relatively lower heavy metal emission contribution; they are linked to energy production (van Vuuren et al., 2017) and steel production (Deetman et al., 2018), respectively, as activity indicators.

An overview of different chemical groups with their primary emission sources in Europe coupled with activity indicators is presented in Fig. 2, which can further differ per chemical, product type, country, and period. The assumptions of main emission sources per chemical group and their emission share were derived through a literature survey (Refer to SI-2) and are represented in emission source band size in Fig. 2. However, it is essential to note that the band size of chemical groups in Fig. 2 is not demonstrative of the group size. For historical and future analysis of chemical emissions per chemical group, emission share per sector was considered to be the EF. The EFs are assumed to be constant, implying that changes in activity indicators solely drive emission changes. So in the future analysis, the difference in activity indicators explained the variability among scenarios. We contemplated three scenarios, SSP1, SSP2 and SSP3, for estimating future chemical emissions as they explain low, medium and high challenges to mitigation and adaptation.

2.3. Application of the framework for individual chemicals and coupling to SSP key quantitative drivers

Given the differences between chemicals in terms of use, properties and fate during technological treatment, we further applied the framework for individual chemicals. The assumption of constant EFs is not necessarily correct – therefore, we also explored the impact of changing EFs for the SSPs for individual chemicals. Five chemicals representing the chemical groups were selected: i.e. pharmaceuticals (ibuprofen and diclofenac), pesticides
(terbutylazine) and industrial chemicals (cadmium and diethyl phthalate). This selection was further based on relevance for water quality in Europe (European Environment Agency, 2018), availability of data on historical emissions (Leclerc et al., 2019) and applicability to be coupled with SSP activity indicators. The EFs of chemicals are estimated for EU countries. The emission estimation varies depending on the availability of SSP activity indicator data which is either for EU countries or Western and Central European regions. When possible, the future chemical emissions are estimated for EU countries and then aggregated as EU regions. However, all the future chemical emissions are presented on Western and Central European regions for consistency.

2.3.1. Pharmaceuticals: diclofenac and ibuprofen

Diclofenac and ibuprofen are the most detected anti-inflammatory drugs in surface waters (Fekadu et al., 2019). The EF_base per country of diclofenac and ibuprofen was determined by coupling their emissions per country over the historical period (Leclerc et al., 2019) with the respective country population size (World Bank OpenData, 2020). \( EF_{\text{base}, \text{diclofenac}} \) and \( EF_{\text{base}, \text{ibuprofen}} \) are the emission factors for diclofenac and ibuprofen, respectively, in \( \text{g cap}^{-1} \text{y}^{-1} \), with \( r \) referring to different EU Member States (MS). Eqs. (3) & (4) to estimate \( EF_{\text{base}} \) were applied across EU-28 countries between 2000 and 2014.

\[
EF_{\text{base}, \text{diclofenac}} = \frac{\text{Emissions}_{\text{base}, \text{diclofenac}} (\text{kg y}^{-1})}{\text{Total Population}_{\text{base}} (r)} \quad (3)
\]

\[
EF_{\text{base}, \text{ibuprofen}} = \frac{\text{Emissions}_{\text{base}, \text{ibuprofen}} (\text{kg y}^{-1})}{\text{Total Population}_{\text{base}} (r)} \quad (4)
\]

Then \( EF_{\text{base}} \) were statistically analysed to understand their trends over time with per country GDP per capita (US$) and population density (cap km\(^{-2}\)) for the EU countries. Historic analysis was performed on the country level to derive \( EF_{\text{future}} \) by analysing emission profiles and linking them with the scenario storyline. SSP1 assumes a relatively fast demographic transition leading to a low population, which reduces the amount of pharmaceuticals used. The SSP1 scenarios emphasise relatively high investments in environmental technology, focusing on the better removal of pharmaceuticals. The SSP1 scenario also assumes that \( EF_{\text{future}} \) reaches the lowest mean \( EF_{\text{base}} \) of all EU countries by 2050. SSP2 represents trends in consumption and emissions that do not shift markedly from historical patterns due to

![Sankey diagram depicting chemical groups and their emission sources and activities](image-url)
2.3.2. Plant protection products (PPP): terbuthylazine

Terbuthylazine is a herbicide used in most European countries for agriculture. Following the framework, EF-base (r, terbuthylazine) (kg ha\(^{-1}\) yr\(^{-1}\)) was determined from historical terbuthylazine emissions and cropland area (ha) per country (FAOSTAT, 2020). Due to limitations in the availability of data, EF-base (r, terbuthylazine) was estimated across 18 European countries. The emission profiles of EF-base (r, terbuthylazine) from 2000 to 2014 were then statistically analysed with GDP per capita (US$) and crop production (Mt yr\(^{-1}\)) of countries to associate them with scenario storylines.

The SSP storylines describing SSP1, with the low population growth, leads to reduced pressure on agricultural production and the harvested area, thereby decreasing the amount of herbicides used. Investments in environmental technology encourage advanced herbicide application methods that consume lesser herbicide than the average. This is implemented by assuming that EF-future (r, terbuthylazine) converges to mean EF-base (r, terbuthylazine) – standard deviation (thus representing the lower 33rd percentile of the emissions). In SSP2, trends do not shift markedly from historical patterns due to moderate population growth and technological development. Hence, EF-future (r, terbuthylazine) converges to the mean EF-base (r, terbuthylazine). In SSP3, policies shifting towards national and regional security issues, particularly in agricultural markets, increase pressure on food, feed, and biofuel crops. This, in turn, increases the use of herbicides and harvested areas with added pressure from climate change. Hence the EF-future (r, terbuthylazine) is assumed to converge to the mean EF-base (r, terbuthylazine) + standard deviation. Though the EF-base (r, terbuthylazine) was estimated at the country level, the EF-future (r, terbuthylazine) were assessed for Western and Eastern European region owing to regional cropland area projections (Popp et al., 2017). Hence, the EF-future (r, terbuthylazine) assumptions are made considering the mean and standard deviation of the Western and Eastern European regions. The future terbuthylazine emissions were calculated from Eq. (5) for the IMAGE regions of Western Europe and Eastern Europe.

\[
\text{Emissions}_\text{future(r, terbuthylazine)} \text{ (kg y}^{-1}\text{)} = \text{EF}_\text{future(r, terbuthylazine)} \text{ (kg y}^{-1}\text{ ha}^{-1}\text{yr}^{-1}\text{)} \times \text{Cropland}_\text{future(r)} \text{ (ha)}
\]  

(5)

2.3.3. Industrial chemicals: DEHP and cadmium

Two industrial chemicals, DEHP and cadmium, were selected. DEHP (bis(2-ethylhexyl) phthalate) is mainly used as a plasticiser in polymers. DEHP emissions mostly occur via wastewater effluents (E-PRTR, 2019; Musgrave et al., 2011) originating from the release of various plastic products through slow diffusion from surfaces of products, washing/cleaning operations or in wet weather (from outdoor domestic uses and surface run-off intercepted by combined sewers) (ECHA, 2019). Such emissions can originate from populated areas with or without connection to wastewater treatment plants. Therefore, we couple DEHP emissions to the total population per country to determine EF-base NL (DEHP). The future DEHP emissions per European country were calculated based on Eq. (6) for 2050 across EU-28 countries.

\[
\text{Emissions}_\text{future(r, DEHP)} \text{ (kg y}^{-1}\text{)} = \text{EF}_\text{future(r, DEHP)} \text{ (kg y}^{-1}\text{ cap}^{-1}\text{)} \times \text{Total Population}_\text{future(r)}
\]  

(6)

In the SSP1 scenario, wastewater treatment could potentially reduce DEHP released by better removal efficiencies. This could be further based on economic growth shifting towards reduced use of plastics. Investment in environmental technology focusing on WWTP helps in better removal efficiencies, resulting in reduced DEHP emissions to surface waters. The lowest mean EF-base NL (DEHP) among all EU countries is considered the best case. Hence, the EF-future NL (DEHP) is converging to the lowest mean EF-base NL (DEHP) by 2050. SSP2 representing the business as usual scenario is described by keeping EF-future NL (DEHP) as the mean EF-base NL (DEHP) of all countries. In the SSP3 scenario, declining investments in technological development and environmental protection are expected to lead to higher material use and, subsequently, a higher mean EF-base NL (DEHP) of all countries.

Cadmium is a heavy metal mainly produced as a by-product of mining, smelting, and refining sulphide ores of zinc, lead, and copper (UNEP, 2010). The primary emission sources of cadmium into surface waters are agriculture, industry, wastewater treatment plants and atmospheric deposition (Pan et al., 2010). EF-base for cadmium was determined by taking publicly available Dutch cadmium emissions to surface water from 2005 (Vos and Janssen, 2008). EF-base for the different sources of cadmium emissions were modelled by connecting them with corresponding activity indicators. Cadmium emissions from WWTP were coupled to the total population (cap\(^{-1}\)), agricultural emissions were coupled to cropland area of food crops (ha), emissions from the paper industry were coupled to the paper production (t yr\(^{-1}\)), and finally, emissions from the metal industry were coupled to steel production (t yr\(^{-1}\)). These baseline data on emission sources were extracted from databases (Eurostat, 2020; FAOSTAT, 2020; OECD Statistics, 2020). Eqs. (7) to (10) were used to determine EF from the corresponding emission sources.

\[
\text{EF}_\text{base NL, WWTP} = \text{Emissions}_\text{base NL, WWTP (kg y}^{-1}\text{)} / \text{Population connected to WWTP} \times \text{base NL}
\]  

(7)

\[
\text{EF}_\text{base NL, ag} = \text{Emissions}_\text{base NL, ag (kg y}^{-1}\text{)} / \text{Harvested area food crops - base NL (ha)}
\]  

(8)

\[
\text{EF}_\text{base NL, P} = \text{Emissions}_\text{base NL, P (kg y}^{-1}\text{)} / \text{Paper production - base NL (t yr}^{-1}\text{)}
\]  

(9)

\[
\text{EF}_\text{base NL, MI} = \text{Emissions}_\text{base NL, MI (kg y}^{-1}\text{)} / \text{Steel production - base NL (t yr}^{-1}\text{)}
\]  

(10)

The EF-base NL (DEHP) and EF-base NL (Cd) values for the Netherlands for Cd are used to make EF-future NL (DEHP) and EF-future NL (Cd) for Western Europe for the case of cadmium. The emissions from 2015 to 2050 were estimated for Western Europe by keeping EF-future NL (DEHP) and EF-future NL (Cd) assumptions constant for the three SSPs due to the lack of yearly data available over countries. The scenario assumptions for cadmium emissions up to 2050 followed the source-specific future activity rate projections from the IMAGE model for SSP1, SSP2 and SSP3. The main scenario assumptions are presented in Table 1.
Table 1
Overview of five selected chemicals with their activity indicators and EF assumptions for 2050. The quantitative assumptions are presented for the Western and Eastern European regions for SSP1, SSP2, and SSP3.

<table>
<thead>
<tr>
<th>Chemical</th>
<th>Region</th>
<th>Activity indicators</th>
<th>Quantitative values (2050)</th>
<th>EF_future (2050)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Western Europe</td>
<td>Eastern Europe</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Low</td>
<td>455.9 (million)</td>
<td>0.06 gcap⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Medium</td>
<td>442.1 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Low</td>
<td>373 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Medium</td>
<td>100.4 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Medium</td>
<td>99 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Low</td>
<td>91.6 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Medium</td>
<td>142.1 (million)</td>
<td>0.44 gcap⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Low</td>
<td>373 (million)</td>
<td>(convergence to highest EF_mean, r)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Low</td>
<td>100.4 (million)</td>
<td>0.027 gcap⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Medium</td>
<td>99 (million)</td>
<td>(convergence to lowest EF_mean, r)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Low</td>
<td>91.6 (million)</td>
<td>(convergence to highest EF_mean, r)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Low</td>
<td>6.89E+05 km²</td>
<td>0.12 gha⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Medium</td>
<td>3.80E+05 km²</td>
<td>0.015 gha⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Low</td>
<td>4.35E+05 km²</td>
<td>0.14 gha⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Medium</td>
<td>5.3E+05 km²</td>
<td>0.02 gha⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Medium</td>
<td>455.9 (million)</td>
<td>0.05 gcap⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Medium</td>
<td>100.4 (million)</td>
<td>(convergence to lowest EF_mean, r)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Medium</td>
<td>99 (million)</td>
<td>(convergence to highest EF_mean, r)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Medium</td>
<td>373 (million)</td>
<td>0.34 gcap⁻¹ y⁻¹</td>
</tr>
<tr>
<td>Terbuthylazine</td>
<td>Western &amp; Eastern Europe</td>
<td>Cropland area (Popp et al., 2017)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Low</td>
<td>11.3 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Medium</td>
<td>14.6 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP1 Low</td>
<td>21.5 (million)</td>
<td>EF_mean, r</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Medium</td>
<td>5.07E+05 km²</td>
<td>0.67 gkton⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP2 Medium</td>
<td>1.35E+05 km²</td>
<td>0.67 gkton⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Low</td>
<td>8.07E+05 km²</td>
<td>0.67 gkton⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Low</td>
<td>4.35E+05 km²</td>
<td>0.67 gkton⁻¹ y⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>SSP3 Low</td>
<td>5.3E+05 km²</td>
<td>0.67 gkton⁻¹ y⁻¹</td>
</tr>
</tbody>
</table>

*: EF_mean, r = the mean EF from 2000 to 2014 of individual EU countries.

SSP3 (following the population trend). Similarly, following trends in animal production, veterinary pharmaceutical emissions could decline by 30% in SSP1 (given the less meat-intensive diets) and increase by 30% in SSP3 (for the opposite reason). Assuming that plant protection products follow trends in crop production, SSP2 shows a slight increase of 8%. Biocide emissions from wastewater treatment plants can be similarly coupled to trends in population size.

Industrial chemicals are used for different purposes, and therefore one needs to add up the trends in individual activities causing these emissions (Table 2). Again, each activity can be coupled to corresponding changes in the IMAGE SSP scenarios, showing trends reflecting the different storylines.

On aggregating all emissions from the contributing sectors for industrial chemical sub-groups, emissions show a rise for SSP1 and SSP2 (Table 2). Inorganic substances show the highest increase in emissions among all subgroups. In SSP1, emissions display an increase of 20%, followed by SSP2 with 19%. The emission projections for inorganic chemicals are in line with the coupled projections of chemical energy demand. Heavy metals present a similar rise in emissions of 3% in SSP1, 7% in SSP2 and a decline of 7% in SSP3. While the emissions increase in SSP1 and SSP2 for other organic substances, SSP3 show a decline in emissions of 10.3% by 2050. Chlorinated organic substances are projected to decrease for SSP1 and SSP3 due to the (declining) trends in pulp and paper production.

3.2. Analysing historical trends and determining EF_future

The EFs for ten larger Western and Eastern Europe MS are presented in Fig. 3 for ibuprofen and DEHP. The EF_diclofenac and EF_terbuthylazine remains relatively constant over the 15 years for all EU MS and are presented separately in the appendix: SI-A. Note that the EFs have different units. For diclofenac, terbuthylazine and DEHP, Western Europe MS have higher EFs compared to Eastern Europe. Ibuprofen, in contrast, shows higher EFs comparatively in Eastern Europe and variation in EFs over time. EF_dehp display slight temporal variation compared to ibuprofen, in which UK and Slovakia had higher mean EF (Fig. 3).

In the next step, the EFs per country per chemical are correlated to several drivers to check if there are additional trends for Western and Eastern Europe. In Fig. 4, correlations between GDP per capita and the yearly derived EFs per country per chemical are calculated. EF_tributylphosphate shows a decreasing trend with increasing GDP for most European MS, with Eastern EU MS displaying the most correlation of r² value 0.182. While correlating the EF_terbuthylazine with GDP per capita, we notice a slight increase in EF with GDP in Eastern Europe, influenced mainly by Slovenia’s higher EF_terbuthylazine variability (Fig. 4), which displays a 15% decrease in EF_terbuthylazine over the years. In contrast to other selected chemicals, EF_dehp increased from 2000 to 2014 for Eastern EU MS with an r² value of 0.365. So, by comparing the variations in EFs of selected chemicals between EU MS, we conclude that there are no clear positive or negative trends on average. In SI-B, individual correlation coefficients per country are reported. Although individual EFs give a high correlation for some countries, no general pattern is found against GDP per capita.

Additionally, the following correlations are checked: 1) EFs of diclofenac, ibuprofen, and DEHP with population density, 2) EF of terbuthylazine with crop production, and 3) EF of DEHP with percentage urban population connected to WWTP (Refer appendix: SI-A). Again at the scale of Western and Eastern Europe, no clear relations between chemical EFs with those drivers are found. In appendix: SI-B individual correlation coefficients per country are reported.

As no clear trends are found between GDP, population density, crop production and percentage of the urban population connected to WWTP, we...
assume that the derived EFs are independent of those drivers and can be set constant for future projections. Hence, we base future emissions on the product of the activity indicator and chemical EF (Table 1).

### 3.3. Scenario results for selected chemicals coupled to SSPs and introducing variable EFs

The assumptions on future trends in EF \(_{\text{future}}(r,i)\) of all five chemicals are based on the historical analysis of EFs and the scenario storyline of the SSPs. While the calculations are performed on a country level, the assumptions of EF \(_{\text{future}}(r,i)\) are made for Western Europe and Central Europe region to keep projections uniform across the chemical groups. Moreover, the EF assumptions are made at the regional level, and also most scenario data on the drivers are available at the regional level only. The main scenario assumptions are presented in Table 1.

The future emission trends of selected pharmaceuticals, ibuprofen and diclofenac, are presented in Fig. 5. For SSP3, in Western Europe, the emission trends for both ibuprofen and diclofenac show a significant increase in emissions up to 2050. While for Eastern Europe, ibuprofen follows in line with the historical path, diclofenac emissions slightly increase.

### Table 2

Summary of the future emission scenarios (2050) for the five chemical groups in SSP1, SSP2 and SSP3. The scenarios are presented with coupled activity rates (2050) for three SSPs and the associated emission sector. Note that activity and emission projections are rounded up to the nearest whole number.

<table>
<thead>
<tr>
<th>Chemical group</th>
<th>Emission sector</th>
<th>Emission share</th>
<th>Activity (2050)</th>
<th>Emissions (2050)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pharmaceuticals</td>
<td>Household</td>
<td>90%</td>
<td>4%</td>
<td>4%</td>
</tr>
<tr>
<td></td>
<td>Healthcare</td>
<td>10%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Veterinary</td>
<td>Poultry &amp; Pig</td>
<td>33.3%</td>
<td>30%</td>
<td>30%</td>
</tr>
<tr>
<td>pharmaceuticals</td>
<td>Sheep</td>
<td>13.7%</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cattle</td>
<td>53%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plant protection</td>
<td>Agriculture</td>
<td>90%</td>
<td>20%</td>
<td>20%</td>
</tr>
<tr>
<td>products</td>
<td>Other</td>
<td>10%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Biocides</td>
<td>WWTP</td>
<td>100%</td>
<td>4%</td>
<td>4%</td>
</tr>
<tr>
<td>Industrial chemicals</td>
<td>WWTP</td>
<td>40-70%</td>
<td>4%</td>
<td>4%</td>
</tr>
<tr>
<td></td>
<td>Energy</td>
<td>5-20%</td>
<td>23%</td>
<td>23%</td>
</tr>
<tr>
<td></td>
<td>Iron &amp; steel</td>
<td>5-20%</td>
<td>0%</td>
<td>27%</td>
</tr>
<tr>
<td></td>
<td>Chemicals</td>
<td>5-20%</td>
<td>32%</td>
<td>32%</td>
</tr>
<tr>
<td></td>
<td>Pulp paper &amp; wood</td>
<td>5-20%</td>
<td>9%</td>
<td>21%</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>3%</td>
<td>6%</td>
</tr>
<tr>
<td>Chlorinated organic chemicals</td>
<td>WWTP</td>
<td>20-40%</td>
<td>4%</td>
<td>4%</td>
</tr>
<tr>
<td></td>
<td>Chemicals</td>
<td>5-20%</td>
<td>32%</td>
<td>32%</td>
</tr>
<tr>
<td></td>
<td>Pulp paper &amp; wood</td>
<td>40-70%</td>
<td>9%</td>
<td>21%</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>3%</td>
<td>0%</td>
</tr>
<tr>
<td>Other organic chemicals</td>
<td>WWTP</td>
<td>40-70%</td>
<td>4%</td>
<td>4%</td>
</tr>
<tr>
<td></td>
<td>Chemicals</td>
<td>5-20%</td>
<td>32%</td>
<td>32%</td>
</tr>
<tr>
<td></td>
<td>Pulp paper &amp; wood</td>
<td>20-40%</td>
<td>9%</td>
<td>21%</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>5%</td>
<td>3%</td>
</tr>
<tr>
<td>Inorganic chemicals</td>
<td>WWTP</td>
<td>20-40%</td>
<td>4%</td>
<td>4%</td>
</tr>
<tr>
<td></td>
<td>Chemicals</td>
<td>40-70%</td>
<td>32%</td>
<td>32%</td>
</tr>
<tr>
<td></td>
<td>Pulp paper &amp; wood</td>
<td>20-40%</td>
<td>3%</td>
<td>8%</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>20%</td>
<td>19%</td>
</tr>
</tbody>
</table>

* The total emissions per chemical group are presented in bold and colored cells.
emissions for SSP2 remain relatively constant for both regions and pharmaceuticals. In contrast, we notice a significant decrease in emissions for SSP1, mainly due to a decreasing population and decreasing emission factors.

A decrease in terbuthylazine emissions in SSP1 is noticed for Western and Eastern European regions (Fig. 5). While SSP3 and SSP2 show a significant increase in future emissions. For Eastern Europe, the emissions were slightly higher than for Western Europe (Fig. 5). DEHP’s future emission trends are displayed in Fig. 5, where it is noticeable that emissions almost linearly decrease in Western Europe for SSP1. Still, in Eastern Europe, emissions increase at the same pace as SSP2. For SSP3, it shows an increase in both regions. In contrast, we notice SSP2 displays a constant emission trend over the years.

Cadmium emissions are presented in Fig. 6 for different sectors. For emissions from WWTP, SSP1 and SSP2 shows a slight increase in emissions with the numbers following population projections for Europe. On the contrary, emissions decreases in SSP3 as Cadmium projections are solely dependent on future change activity and can occur if there are no changes in the EP implemented. The cadmium emissions for SSP2 increased from agriculture and decreased for SSP1. Their emissions from the paper industry are declining, with SSP2 having the lowest emissions, followed by SSP3 and SSP1. The steel industry follows a similar emission trend, with SSP2 decreasing to lowermost emissions. Whereas emissions in SSP1 and SSP3 slightly increase before starting to decrease.

4. Discussion

4.1. Data limitations and uncertainties

The presented method of developing emission scenarios has significant uncertainties despite the previously mentioned advantages. Data limitations are one of the crucial causes of uncertainties. Although databases (E-PRTR, 2019; Umweltbundesamt, 2019) exist with spatial and temporal environmental concentrations for pharmaceuticals and pesticides, they are not consistent over the years across locations to conduct a historical analysis. Leclerc et al. (2019) developed a harmonised inventory for 468 toxic pollutant emissions to air, soil and water for the EU MS. Though the inventory can be used to apply the emission scenario framework for chemicals with simple use, such as pharmaceuticals, pesticides and selected industrial chemicals. Existing inventory can be limited for chemicals like cadmium with multiple sectoral uses as it provides total emissions per chemical and not per sector share.

Additionally, there is not enough evidence of their actual use or emissions for industrial chemicals. While the REACH database has information on industrial chemicals registered for use, representative information is on tonnage bands which does not give sufficient information on the actual usage at specific locations or in specific EU MS. Alternatively, even with E-PRTR arguing about representing 60% of total emissions in a year across 65 economic activities, it is limited to only a few chemicals (E-PRTR, 2020). The level of emissions from specific sources is often not complete, certainly for time trends, and data gaps typically exist for some countries. It is also challenging to get a complete picture despite being the largest category of chemicals being in use. The lack of information on different uses of chemicals, their emission volumes, and spatial differences hampers the chemical risk assessment (Dijk et al., 2021; van Gils et al., 2019) and adds to the uncertainty.

Alternatively, Life Cycle Assessment (LCA) based studies have shown the immense contribution of historical water and soil-borne emissions and their impact on ecosystems and human health (Leclerc et al., 2019). Though lifecycle assessments are beneficial to analyse emissions on a city or country level, aggregating multiple LCAs for various uses across different spatial and temporal scales is challenging. Substance Flow Analysis (SFA) can also be used to understand the life cycle of a chemical at the country level. However, adopting SFAs here was not possible as the available information was often too old and available for a limited number of EU countries. Altogether there is a lack of up-to-date and complete data on emissions. Though the uncertainties related to data limitations cannot be fully addressed, it was adjusted by compiling relevant data from other publications.

Furthermore, uncertainties originate at different stages of emission scenario framework application. First, the choice of activity indicator reflected a certainty uncertainty (certainly when the indicators represent a more heterogeneous activity). Second, there can be heterogeneity in the selected activity indicator derived for 2000–2014 from public databases (e.g. FAOSTAT, Worldbank and Eurostat) and IMAGE future projections for 2015–2050. The uncertainty is addressed by keeping the activity indicators consistent across historical and future periods. For example, EFterbuthylazine was estimated by a consistent selection of cropland area only for food and feed crops from both FAOSTAT and IMAGE in historical and future periods. The IMAGE future projections for cropland area, pulp & paper production
and steel production are only available for Western and Central Europe regions, which limits the future emission estimation explicitly at the country level for Terbutylazine and Cadmium. In comparison, the future emissions of diclofenac, ibuprofen and DEHP are estimated for EU countries with the availability of population growth projections at the country level. Third, uncertainty can arise from other relevant drivers, which can influence emissions. Hence, the correlation of EF-base is checked with other drivers GDP, population density, connection, crop production and wastewater treatment systems; however, there was no correlation between EF-base and drivers; they are not used in future emission estimation (Refer appendix: SI-A).

Uncertainties in the framework also arise with the future drivers used for the projections, such as population, cropland area, and pulp and paper production. To some degree, such projections can be validated by multimodel comparisons (Riahi et al., 2017). Maybe, more importantly, there are uncertainties in the EF-future assumptions themselves. The EF-future assumptions are based on emission data from 18 to 28 EU countries for 15 years, which addresses a broader uncertainty range and are presented in different scenarios. However, other information (e.g. on technology or legislation) would be more interesting to use that comparison across countries.

Overall, there are considerable uncertainties in the future emissions estimations. However, the emission scenario framework and the application demonstrate the challenges posed by chemical emissions in future.

4.2. Discussion of emission scenarios

The emission scenario projections can be influenced by changing activity levels or EF levels. In the first experiment, we assumed that EF would remain constant in time, meaning that future emissions are only a function of varying activity levels. In the subsequent work, however, we show that historically EFs change over time and are different across countries. The differences may occur from different use patterns and technological and policy measures over time. Therefore, the model with dynamic EFs is better, and historical data could create best and worst-case scenarios.

Further on comparing the emission scenario projections with results of synthetic chemical change from Bernhardt et al. (2017), we noticed a similar pattern in emissions for chemical groups by 2050, even though the proportional change of synthetic chemicals in the paper was presented for 1970 to 2015. The pharmaceutical emissions are projected to increase by 3–4% in SSP1 and SSP2 for Europe, which is identical to the steep increase...

![Fig. 4. Correlation between GDP per capita (US$) and emission factors for the four chemicals over the historical period between 2000 and 2014. Correlation is presented for Western (left) and Eastern (right) European member states.](image-url)
in pharmaceutical consumption by four times. Industrial emissions as a whole increase for both SSP1 and SSP2 being proportional to the chemical industry output from developed countries from Bernhardt et al. (2017). Though SSP1 and SSP2 had a rise in emissions compared to Bernhardt et al. (2017), SSP3 had a conflicting trend owing to the activity indicators.

On comparing emission projections for selected chemicals—diclofenac, ibuprofen and DEHP in SSP3 for both Western and Eastern Europe were similar to trends of synthetic chemical change from Bernhardt et al. (2017), but for terbuthylazine, the emission results were only comparable for Western Europe.

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**Fig. 5.** Emissions up to 2050 of (a) diclofenac (b) ibuprofen (t y$^{-1}$), (c) terbuthylazine (t y$^{-1}$) and (d) DEHP (t y$^{-1}$).

**Fig. 6.** Cadmium emissions (kg y$^{-1}$) up to 2050 from (a) wastewater treatment plants, (b) agriculture, (c) paper industry and (d) steel production.
4.3 Emission scenario framework

The emission scenario framework developed in the current research helps understand the future chemical emission trends to freshwater. The framework adopts extensively used activity and emission factor based emission estimation (US EPA, 1995) and extends the existing SSP scenarios based on past trends. Similar to other efforts on understanding phosphorus and nitrogen pollution (Beusen et al., 2016; van Puijenbroek et al., 2019) and air pollution (Rao et al., 2017). The chemical emission projections up to 2050 are in line with SSP storylines, SSP3 having higher emission levels indicating high challenges to mitigation and adaptation and SSP1 being a sustainable scenario with lower emissions. This approach displayed significant potential in understanding the future risk of chemical emissions to water.

The framework can be applied similarly to other pharmaceuticals and pesticides. For example, other pharmaceuticals can be coupled to population growth, and pesticides can be related to the harvested area or crop production based on their use on different crops. However, the method can be improved by determining age and gender-based consumption for other pharmaceutical classes such as anti-diabetics and contraceptives and finding specific crop use for pesticides. The framework needs a thorough analysis of various emission sources for the case of industrial chemicals. Simultaneously, the basic approach follows the efforts already been made on a global level to estimate future air pollution (Rao et al., 2017). However, in contrast to emissions to air, extensive data sources are missing for emissions to water.

The emission scenario framework can be further extended to include specific policies such as the EU Green Deal’s ambitions to overcome the challenges of environmental degradation with a zero-pollution vision (European Commission, 2019). The goals of the Zero-pollution action plan to 2050 can be translated into quantitative assumptions as part of EF. Future. Dijk et al. (2021) discussed the importance of analysing changes in pollution and its effects over space and time in the risk assessments to achieve the EU policy goals. The scenario-based framework can help to improve risk assessments by quantifying future policy targets such as improved wastewater treatment and reduced use per activity, region and time.

However, to include the EU Green deal scenario in the current research, it was unclear to translate the zero-pollution action goals into quantitative assumptions.

Often chemicals can be substitutes, which could be the case for diclofenac and ibuprofen (as pain killers) – but this is clearly also the case for herbicides like terbuthylazine and industrial chemicals like DEHP. To deal with this, ideally, an alternative model formulation would be built around the function of the chemical (so pain killer use; herbicide use), and both the activity levels and emissions factors would be introduced at this function level. Subsequently, one could describe the specific chemicals used. However, this would require introducing “substitution equivalents”, and a complete description of all underlying chemicals and all possible chemical uses, going beyond the illustrative purposes of the present article.

5. Conclusion

The emission-factor approach presented in the paper can be used to create emission scenarios for chemicals to water systems. The emission-factor approach, which is extensively used in understanding atmospheric emissions, is used in this paper to relate and quantify chemical emissions with relevant socio-economic drivers. This approach displayed significant potential in understanding the future risk of chemical emissions. It provides a framework to include socio-economic changes that influence the use and emissions of chemicals in futuristic risk assessments. The estimated emissions of chemicals can be based on chemical emissions data available from public databases, which theoretically makes it convenient to reproduce this method for other chemicals.

The scenarios created in this paper show widely diverging trends for different SSPs. The chemical emission scenarios developed in the current research paper are the first efforts to extend SSPs to understand water pollution. The emission scenarios are developed for five chemical groups: pharmaceuticals, veterinary pharmaceuticals, plant protection products, biocides and industrial chemicals, representing agricultural, industrial, and urban systems. For all the chemical groups, emissions are projected to increase in SSP1 and SSP2 and decrease in SSP3. Veterinary pharmaceuticals show the highest increase in emission of 30%, followed by plant protection products and industrial chemicals, displaying a 20% increase. The emission scenario projections for selected chemicals widely vary per SSP and chemical.

The approach can be applied similarly to other pharmaceuticals and pesticides. While the framework can be extended to other pharmaceuticals and pesticides, it requires a detailed understanding of complex emission sources for industrial chemicals.

The most crucial obstacle in developing scenarios for future emissions to water is the lack of reliable and complete data of chemical emissions. In developing the scenarios, it was challenging to find reliable and comprehensive data on historical emissions. Creating databases of emissions to water for relevant chemicals is, therefore, a priority.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.155530.

CRediT authorship contribution statement

Poornima Nagesh, Conceptualization, Methodology, Formal analysis, Visualization, Writing – original draft, Drafting – review & editing.

Hugo J. de Boer, Supervision, Writing – review & editing.

Annamarie P. van Wezel, Supervision, Writing – review & editing.

Stefan C. Dekker, Supervision, Writing – review & editing.

Dietv P. van Vuuren, Methodology, Supervision, Writing – review & editing.

Declaration of competing interest

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

Acknowledgements

This work is part of the Innovative Training Network ECORISK2050 and was supported by the European Union’s Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No [813124].

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