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## Review

# Enabling forecasts of environmental exposure to chemicals in European agriculture under global change



John D. Hader<sup>a,1</sup>, Taylor Lane<sup>b,1</sup>, Alistair B.A. Boxall<sup>b</sup>, Matthew MacLeod<sup>a,\*</sup>, Antonio Di Guardo<sup>c</sup>

<sup>a</sup> Department of Environmental Science, Stockholm University, 10691 Stockholm, Sweden

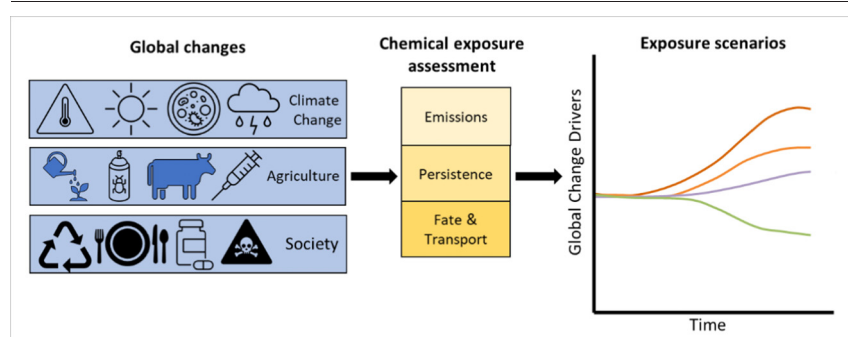
<sup>b</sup> Department of Environment and Geography, University of York, Heslington, York, North Yorkshire YO10 5NG, United Kingdom

<sup>c</sup> Department of Science and High Technology, University of Insubria, Via Valleggio 11, 22100 Como, CO, Italy

## HIGHLIGHTS

- Global change will impact agricultural chemical emissions, persistence, & transport.
- We identify knowledge & data gaps for future chemical exposure scenario assessment.
- We present Agricultural Chemical Exposure (ACE) scenarios as forecasting framework.
- Forecasts of agricultural chemical exposures will guide sustainable development.

## GRAPHICAL ABSTRACT



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## ABSTRACT

European agricultural development in the 21st century will be affected by a host of global changes, including climate change, changes in agricultural technologies and practices, and a shift towards a circular economy. The type and quantity of chemicals used, emitted, and cycled through agricultural systems in Europe will change, driven by shifts in the use patterns of pesticides, veterinary pharmaceuticals, reclaimed wastewater used for irrigation, and biosolids. Climate change will also impact the chemical persistence, fate, and transport processes that dictate environmental exposure. Here, we review the literature to identify research that will enable scenario-based forecasting of environmental exposures to organic chemicals in European agriculture under global change. Enabling exposure forecasts requires understanding current and possible future 1.) emissions, 2.) persistence and transformation, and 3.) fate and transport of agricultural chemicals. We discuss current knowledge in these three areas, the impact global change drivers may have on them, and we identify knowledge and data gaps that must be overcome to enable predictive scenario-based forecasts of environmental exposure under global change. Key research gaps identified are: improved understanding of relationships between global change and chemical emissions in agricultural settings; better understanding of environment-microbe interactions in the context of chemical degradation under future conditions; and better methods for downscaling climate change-driven intense precipitation events for chemical fate and transport modelling. We introduce a set of narrative Agricultural Chemical Exposure (ACE) scenarios — augmenting the IPCC's Shared Socio-economic Pathways (SSPs) — as a framework for forecasting chemical exposure in European agriculture. The proposed ACE scenarios cover a plausible range of optimistic to pessimistic 21st century development pathways. Filling the

\* Corresponding author.

E-mail address: [matthew.macleod@aces.su.se](mailto:matthew.macleod@aces.su.se) (M. MacLeod).

<sup>1</sup> Shared first authorship.

knowledge and data gaps identified within this study and using the ACE scenario approach for chemical exposure forecasting will support stakeholder planning and regulatory intervention strategies to ensure European agricultural practices develop in a sustainable manner.

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## 1. Introduction

The 20th century was marked by unprecedented changes in human population, demographics, land use, technology, and emissions of climate-changing greenhouse gases (e.g., [Steffen et al., 2007](#)). Agriculture, which “sits at the interface between the environment and society” ([Masseroni et al., 2017](#)), has both driven these changes, and been driven to respond to them. Currently, agriculture is estimated to account for 38 % of land use globally ([FAO, 2020](#)), 70 % of freshwater withdrawals ([FAO, 2017](#)), and requires the use of hundreds of millions of tonnes of chemical or mineral fertilizer nutrient additions annually ([FAO, 2021a](#)). Globally, since the mid-20th century, per-hectare land equipped for irrigation has doubled, gross agricultural output has more than doubled, and nutrient fertilizer use has increased by over 600 % ([Fig. 1](#)). Similar patterns are seen across Europe, except for nutrient fertilizer use which has remained relatively stable since 1990.

In addition to its large land, water, and nutrient footprint, a wide range of non-nutrient chemicals is emitted by, and cycled within, agricultural systems. Pesticides, veterinary pharmaceuticals, and feed additives enter the environment as a direct consequence of their use in agriculture (e.g., [Boxall et al., 2003](#); [Kuppusamy et al., 2018](#); [Silva et al., 2019](#); [de Souza et al., 2020](#)). Other chemicals, including pharmaceuticals and personal care products, are introduced unintentionally into agricultural systems through the application of reclaimed wastewater for irrigation and sludge biosolids applied as a soil amendment (see [Christou et al., 2017](#); [Topp et al., 2010](#)). In Europe, this diverse group of chemicals in agriculture is subject to exposure and risk assessment under different regulatory instruments, including [Regulation \(EU\) 2019/6](#) for veterinary medicines, [Regulation \(EU\) No 528/2012](#) for biocidal products, [Regulation \(EC\) No 1107/2009](#) for plant protection products, and the Registration, Evaluation, Authorisation, and Restriction of Chemicals (REACH) regulation for many chemicals emitted down the drain ([Regulation \(EC\) No 1907/2006](#)).

A number of global change drivers over the 21st century (notably, changes in climate, disease and pest pressures, human diet, technology, and policy) have the potential to change the types and amounts of non-nutrient chemicals that cycle within the agricultural system in Europe (see [Welch et al., 2021](#)). These changes will be superimposed on changing

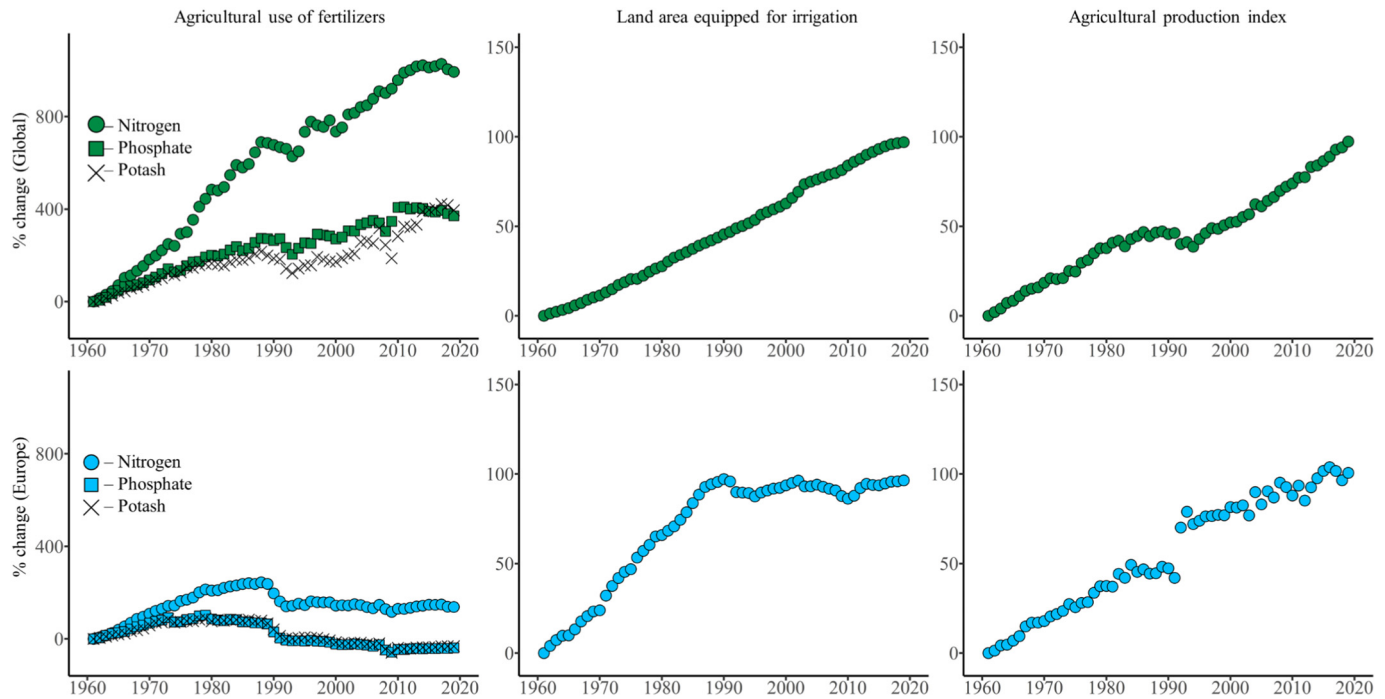
environmental conditions, such as increases in heat waves, floods, and droughts (e.g., [IPCC, 2021](#); [Fig. SI 1](#)) which themselves will impact chemical transformation, fate, and transport.

The goals of this paper are to 1.) review the scientific literature regarding how global change drivers may impact the introduction of organic chemicals into agriculture and how chemical fate, transport, and exposure processes might change in the future, and the associated implications for fate testing and modelling; and 2.) based on this literature review, develop a set of preliminary, narrative scenarios describing a plausible range of alternative pathways of human development and environmental change that, when developed into quantitative scenarios, could drive exposure modelling simulations of European agricultural systems to organic chemicals. The development and analysis of such alternative pathways is a common practice in climate science (e.g., [IPCC, 2021](#); [Riahi et al., 2017](#)) and has also been applied specifically to European agricultural development ([Mitter et al., 2020](#)). Our proposed Agricultural Chemical Exposure (ACE) scenarios augment previous scenario frameworks with consideration of global change's possible impacts on agricultural chemical exposures. While exposure to inorganic chemicals and microplastics will likely also be impacted by the global change drivers we discuss, our main focus is on organic chemicals, and we focus on agricultural practices in Europe.

In [Section 2](#) we present the literature review focused on impacts of global change on chemical emissions ([Section 2.1](#)), chemical persistence and transformation ([Section 2.2](#)), and chemical fate and transport ([Section 2.3](#)) in agricultural systems. Research and data needs that must be addressed to develop our narrative scenarios into quantitative scenarios that can drive chemical exposure models are summarized in each of these three subsections. In [Section 3](#), we present our narrative Agricultural Chemical Exposure (ACE) scenarios, and in [Section 4](#), we discuss the ACE scenarios from the perspective of modelling and how the outputs of scenario analysis could be used.

## 2. Review of the impact of global change on agricultural chemical exposure

[Bloomfield et al. \(2006\)](#) adopted a “source-pathway-receptor” framework for their review of how climate change may impact pesticides in surface and groundwater in the United Kingdom. For our literature review, we



**Fig. 1.** Percent change (relative to 1961 values) in per-hectare agricultural use of fertilizers (left column), land area equipped for irrigation (middle column), and agricultural production index (right column) globally (top row) and across Europe (bottom row) from 1961 to 2019. Note the y-axis scale difference between the figures for fertilizer use and the other two categories. Geographic and temporal comparisons should be made with caution, due to variability and inconsistencies in methods and data coverage. Data from FAOSTAT (FAO, 2021a). Data analysis and plotting conducted using R with ggplot2 (R Core Team, 2019; Wickham, 2016).

focus on sources of organic chemicals and exposure of the agricultural system by considering chemical emissions, persistence & transformation, and fate & transport (see Fig. 2). The focus of our literature review is on non-nutrient, organic chemicals introduced into agricultural environments for crop protection (pesticides), animal rearing (veterinary pharmaceuticals), and as a result of soil irrigation/amendment, especially with wastewater and biosolids.

## 2.1. Emissions of chemicals into agriculture and drivers of future change

### 2.1.1. Overview

**2.1.1.1. Pesticides.** Data from the Food and Agricultural Organization (FAO, 2021a) show that per-hectare European pesticide usage has been stable since 1990, while global usage nearly doubled between 1990 and 2010 (see Fig. 3), then plateaued during 2010–2019 at a level of tonnes of pesticides used per year.

Future changes in temperature and precipitation patterns due to climate change (see Fig. SI 1) are forecasted to shift agricultural activity and associated pest pressures, potentially requiring more widespread, more frequent, and higher intensity pesticide use (e.g., Bloomfield et al., 2006; Delcours et al., 2015). For example, the area suitable for crops in Europe increases by 27 % between 2020 and 2100 under the Intergovernmental Panel on Climate Change's (IPCC's) Representative Concentration Pathway 4.5 scenario (Grünig et al., 2020). Increased temperatures may increase pest generation and growth rates, lengthen plant growing seasons, and shift crop growth regions northward (Noyes et al., 2009; Delcours et al., 2015). Grünig et al. (2020) modelled how the climatic suitability for 89 insect pests in Europe may change under different climate change scenarios, and found on average a 9 % increase in suitable area for pests across Europe under the RCP 4.5 scenario. A notable example was that the suitable area for *S. frugiperda*, an insect capable of destroying crops and which has a documented history of invasive behaviour (see EPPO, 2022), increased by 51 %.

Other factors that could drive increased pesticide use include increasing resistance of target organisms to pesticides (see Bloomfield et al., 2006;

Delcours et al., 2015), and increased proliferation of plant diseases driven by increased precipitation and/or frequency of extreme events (Delcours et al., 2015). Furthermore, changes in the timing of pest activity due to climate change (e.g., warmer temperatures leading to earlier pest activity during the year; Bloomfield et al., 2006; see also Delcours et al., 2015) may require changes in the timing of pesticide application. New pesticide active ingredients may also be required to adapt to changes in plant tolerance and pest resistance (e.g., see Bloomfield et al., 2006; Delcours et al., 2015), and previous shifts in pesticide formulations have been associated with increased toxicity for some ecological endpoints (Schulz et al., 2021).

While increasing suitable agricultural land areas and increased pressure and ranges of pests may drive increases in the use of pesticides in the coming decades, new technology and changes in agricultural practice may act to reduce the amount of pesticides that are used and emitted. For example, the development of modern agronomic techniques alongside novel agrochemical delivery methods may enable the use of less traditional plant protection products (e.g., Bonanomi et al., 2008; Kookana et al., 2014; Kah and Hofmann, 2014). One such technique is the use of 'nanopesticides', which can include pesticides contained within nanoemulsions or polymeric capsules. These can be designed such that the active ingredient is protected from degradation or that delivery is enhanced via increased solubility (Kah and Hofmann, 2014; Kookana et al., 2014). While the use of such technologies could result in an overall reduction in the quantity of pesticide being used, they will result in novel materials (such as nanomaterials) being introduced to agricultural systems, and exposure and risk assessment methods for these materials need to be further explored (Kookana et al., 2014).

Precision agriculture (e.g., using satellite data and drones to optimize inputs to and yields from agriculture) can also reduce pesticide emissions (e.g., EEA, 2019). Furthermore, as part of their Zero Pollution Action Plan, the EU has a target of reducing the use of and risk from chemical pesticides by 50 % by 2030, in pursuit of their ultimate goal by 2050 of reducing air, water, and soil pollution to levels that are not harmful to ecosystems (i.e., a "toxic-free environment"; EC, 2021a).

Another driver of reduced pesticide emissions is the trend towards organic crop production. The per-hectare amount of agricultural area devoted



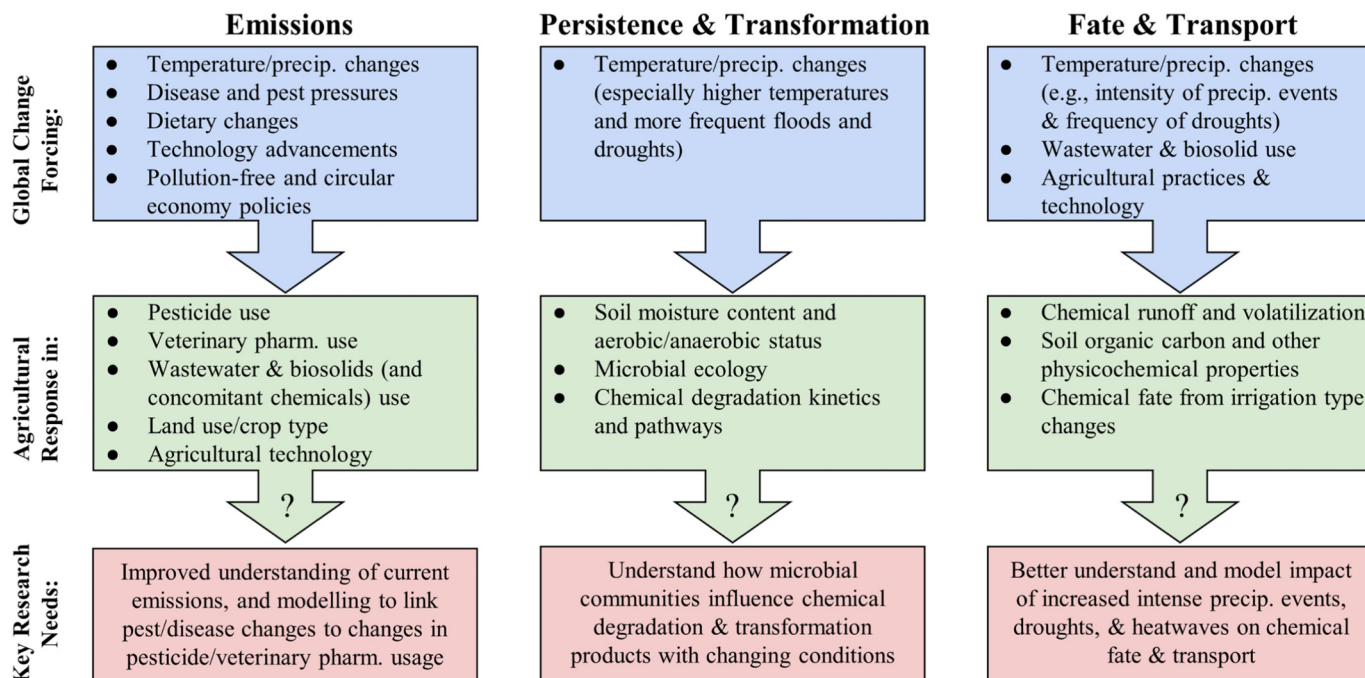


Fig. 2. Flow diagram enumerating—for the three themes of emissions, persistence & transformation, and fate & transport of agricultural chemicals in Europe—the expected global change forcings, the responses in agriculture, and the associated key research needs for evaluating scenario-based forecasts of environmental exposures. Abbreviations: precip. = precipitation; agri. = agriculture; pharm. = pharmaceuticals.

to organic agriculture has roughly doubled in Europe since 2004, with the increase in organic cropland being substantially higher (see Fig. SI 2). As part of the EU Green Deal's "Farm to Fork" strategy (EC, 2020a), the EU has set a goal of having at least 25 % of agricultural land being organic by 2030, an increase over the 8.5 % reported in 2019 (Eurostat, 2021). It should be noted, however, that certain pesticides are still permitted for use in organic farming in Europe (such as various copper- or sulphur-based pesticides, see Annex I of EC, 2021b), and that the environmental impact of organic pesticides can be more uncertain compared to those used in conventional farming due to a lack of life cycle assessment characterization factors (see Meier et al., 2015).

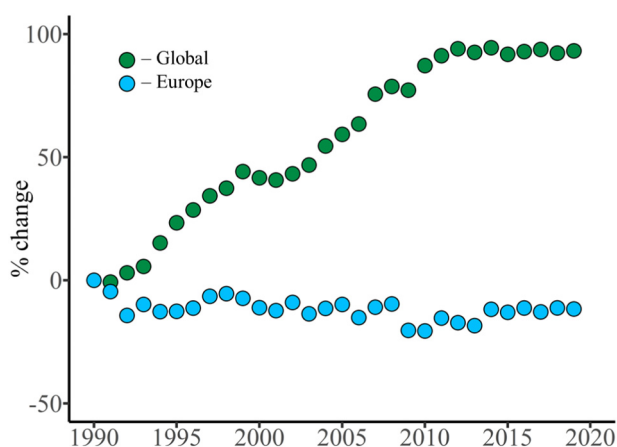


Fig. 3. Percent change (relative to 1990) of per-hectare pesticide usage globally and in Europe from 1990 to 2019. Pesticide usage data are defined as the "quantities (in tonnes of active ingredients) of pesticides used in or sold to the agricultural sector for crops and seeds" (see source for more details on data collection). Geographic and temporal comparisons should be made with caution, due to variability and inconsistencies in methods and data coverage. Data from FAOSTAT (FAO, 2021a). Data analysis and plotting conducted using R with ggplot2 (R Core Team, 2019; Wickham, 2016).

In summary, some global change drivers are likely to increase pesticide usage, while others portend potential decreases. Bounding scenarios for future emissions of pesticides under global change should reflect both increases and reductions, and represent potential changes in pesticide active ingredients and formulations (see Section 3).

**2.1.1.2. Veterinary pharmaceuticals.** The use of antibiotics, steroidal growth-promoting substances, *endo*- and *ecto*-parasiticides, and other veterinary pharmaceuticals in livestock rearing is known to result in residues and metabolites of these compounds in agricultural environments, generally after treatment of livestock and the spread of manure onto fields (e.g., Boxall et al., 2003; Sarmah et al., 2006; Challis et al., 2021). In 2018, there were over 6430 tonnes of active ingredients of veterinary pharmaceuticals purchased across European countries (EMA, 2020). Looking forward, climate change and a societal movement towards plant-based foods could drive changes in the use and emissions of, and therefore environmental exposure to, veterinary pharmaceuticals.

Changing climate may increase disease pressures for animals in agriculture (Gale et al., 2007). For example, more frequent flooding may increase the spread of diseases through spreading of pathogen spores (e.g., anthrax, cryptosporidium), and by creating conditions in which vectors for parasites and viruses (e.g., snails or mosquitos) thrive. Future environmental conditions may also be more suitable for non-native pathogens (Gale et al., 2007). Adaptation to climate change may also require different animal housing practices and thus increase the spread of diseases through animal-animal contacts (Gale et al., 2007). All of these factors could result in an increase in the use, and therefore environmental emissions, of veterinary pharmaceuticals (e.g., Boxall et al., 2009).

On the other hand, over recent years there has been an increase in consumer demand for non-animal (or 'alternative') sources of protein (e.g., plant-based meat or dairy alternatives; Aschemann-Witzel et al., 2021). An analysis of retail market data conducted by the Smart Protein EU Horizon 2020 project showed that sales within the plant-based meat/dairy sector increased by 49 % between 2018 and 2020 across 11 European countries (Smart Protein Project, 2021). In addition to having a smaller environmental footprint in terms of land and water usage and greenhouse gas emissions compared to animal sources of protein

(e.g., Poore and Nemecek, 2018), plant-based proteins do not require the use and emissions of veterinary pharmaceuticals. Demand for alternative protein sources is expected to continue to grow. Barclays Investment Bank (2019) projects that between ~2019 and 2029, the global market share of alternative meats (e.g., plant-based and lab-grown meats) will increase 10-fold from 1 % to 10 %. Furthermore, the EU, as part of their Zero Pollution Action Plan, intends to decrease the sale of farmed animal/aquaculture antimicrobials by 50 % by 2030 (EC, 2021a). However, the projected increase in alternative protein demand will likely occur against a global backdrop of increasing demand for animal protein due to growing population and rising incomes (e.g., OECD/FAO, 2021; see also Bijl et al., 2017).

A range of scenarios that encompass different paths for veterinary pharmaceutical emissions are thus possible, depending on the magnitude of different competing global change forcings that may increase or decrease emissions, as described below in Section 3.

**2.1.1.3. Reclaimed wastewater and biosolids.** Reclaimed municipal wastewater is used in agriculture in water-stressed and arid regions such as the Middle East, North Africa, and southern Europe (Carter et al., 2019). In Europe (EU-28), 50 % of the sludge produced from wastewater treatment is applied to agricultural fields, with the fraction varying widely across member countries (see Collivignarelli et al., 2019). These waste resources contain contaminants such as heavy metals (Xu et al., 2010), persistent organic pollutants, chemicals of emerging concern like pharmaceuticals, endocrine disrupting chemicals, and personal care products (e.g., Tavazzi et al., 2012; Christou et al., 2017), and microplastics (e.g., Corradini et al., 2019).

The types and concentrations of organic chemicals present in reclaimed wastewater and biosolids are determined by down-the-drain disposal of chemicals from connected households and businesses, and the wastewater treatment processes employed (e.g., Struijs, 2014). Estimating down-the-drain emissions of chemicals and determining their fate in wastewater treatment plants is an area of active research, with recent examples for pharmaceuticals in Canada (Grill et al., 2016), estrogens in China (Grill et al., 2018), triclosan and linear alkylbenzene sulphonate in the UK (Kilgallon et al., 2017), and personal care products in the US, South Korea, the Netherlands, and France (Douziech et al., 2018).

The FAO's Global Information System on Water and Agriculture (AQUASTAT, see FAO, 2021b) provides data on the amount of treated or untreated municipal wastewater used directly for irrigation purposes (Fig. 4). The ratio of reported treated municipal wastewater used for irrigation to the amount of treated wastewater produced is the highest in water-stressed regions such as the Middle East, Chile, Australia, and North Africa. Within Europe, the only countries for which these data are available from AQUASTAT are Cyprus, Greece, and Italy (Fig. 4). However, other literature indicates that additional EU countries use treated wastewater for irrigation (e.g., Spain; see Paranychianakis et al., 2015), and several issues in reporting of reclaimed wastewater usage in Europe are discussed in a 2015 report on optimising EU water reuse prepared for the EU's Directorate-General for the Environment (BIO, 2015). The values in Fig. 4 should thus be viewed as incomplete, not directly comparable between countries, and are subject to unquantified uncertainties. Such data gaps and uncertainties prevent accurate analysis of trends in the use of reclaimed wastewater in agriculture in Europe and globally.

Climate change is expected to increase agricultural irrigation water demand by roughly 20 % or more in southern Europe in the 21st century, with some regions showing higher increases (see EEA, 2017). While some of the increased demand for irrigation water may be offset by increased irrigation efficiency (e.g., Nikolaou et al., 2020), one proposed solution to meet some of the projected demand is the increased use of reclaimed wastewater (Regulation (EU) 2020/741; Nikolaou et al., 2020). Only about 2.4 % of treated urban wastewater (roughly 1100 million m<sup>3</sup>/year) is estimated to be reused in Europe as of 2015, corresponding to roughly 0.4 % of freshwater withdrawals in the EU annually (BIO, 2015)—this includes use in agriculture irrigation as well as other uses including industrial use and non-

potable water use in urban areas. While projections are limited and highly uncertain, one estimate of potential increased wastewater reuse in Europe is for it to reach 6000 million m<sup>3</sup>/year by 2025 (BIO, 2015). Furthermore, increased wastewater reuse for irrigation is part of the European Commission's Circular Economy Action plan (Section 3.7 of EC, 2020b). This increased use of reclaimed wastewater could result in increased emissions of chemicals such as pharmaceuticals and other down-the-drain chemicals into agricultural environments (e.g., see Carter et al., 2019).

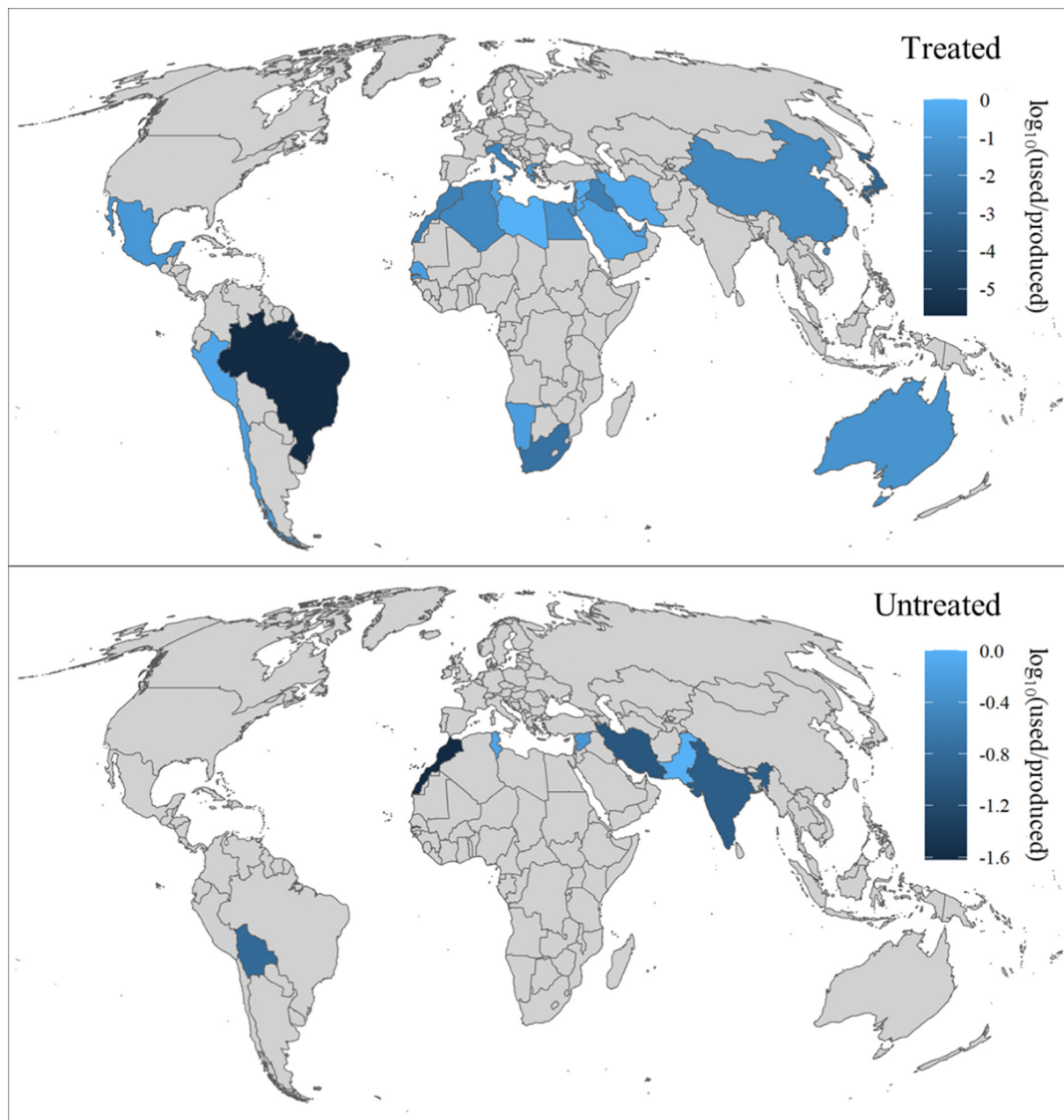
The magnitude and trend of biosolids application in agriculture varies widely across European countries, as exemplified by data from Spain, the Netherlands, and Norway (Fig. 5, based on data from Eurostat, 2022), however gaps in data reporting make comparisons between countries difficult. Looking ahead, as part of the European Commission's Circular Economy Action Plan, an "Integrated Nutrient Management Plan" will be developed to stimulate the recovered nutrient market and increase nutrient application sustainability, with a review of the current directive for sewage sludge to be considered (EC, 2020b). Such measures could create continent-wide incentives to increase the use of biosolids in agriculture. Given the wide variability of current biosolids usage across Europe (Fig. 5), there is therefore a wide variability of possible increased biosolids application patterns and associated flux of chemicals present in biosolids to agricultural environments. Furthermore, increases in the area suitable for crops (see Grünig et al., 2020) may drive shifts in where biosolids and/or reclaimed wastewater are applied, and thus where chemicals are being emitted.

However, with the implementation of the EU's Chemicals Strategy for a Toxic-Free environment, additional safety requirements for chemicals that are present in consumer products (e.g., applying to persistent/bioaccumulative chemicals or endocrine disruptors; see EC, 2020c) could result in lower down-the-drain emissions of potentially harmful chemicals from consumer products that reach wastewater treatment plants. Furthermore, as with the emissions of pesticides and veterinary antimicrobials, the implementation of the EU's Zero Pollution Action Plan may result in decreased amounts of pharmaceuticals and other micropollutants in wastewater and biosolids that are ultimately applied to agriculture (EC, 2021a). The implications of these pollution-reducing strategies, in tandem with possible changes in the usage of waste resources in agriculture, are considered within the bounds of possible future scenarios of agricultural chemical emissions presented in Section 3.

### 2.1.2. Research needs to parameterize quantitative scenarios for emissions of chemicals in agriculture under global change scenarios

An improved understanding of current locations and trends of agricultural chemical usage are needed to establish a baseline of emissions to agricultural settings of pesticides, veterinary pharmaceuticals, and chemicals in waste resources. As an example, improved spatial resolution (e.g., at the field or catchment scale) of pesticide and veterinary pharmaceutical usage, in contrast to using the country- or regionally-resolved application or sales data that is currently available for development of emissions estimates (e.g., see Maggi et al., 2019; Werner et al., 2018) would greatly aid in chemical fate and exposure modelling efforts. Lack of data on current chemical usage has been identified as a key knowledge gap in establishing forward-looking emissions scenarios in other studies (see Nagesh et al., 2022; Desrousseaux et al., in press).

Additionally, the types and quantities of chemicals present in reclaimed wastewater and biosolids using current and emerging treatment technologies should be studied further, and include suspect screening and non-target approaches (e.g., González-Gaya et al., 2021). More modelling investigations of urban down-the-drain chemical emissions and their fate within the wastewater treatment process (e.g., Grill et al., 2016) would also aid in understanding the chemical load entering waste resources, which is crucial to understand the flux of these chemicals into the environment upon application to agricultural areas. Furthermore, to address the large uncertainties and data gaps with the use of wastewater and biosolids in agriculture (see Section 2.1.1.3), European governments should coordinate to harmonise data reporting to AQUASTAT. Specifically, data on the source, amount, treatment quality,



**Fig. 4.** Log-transformed proportion of produced wastewater used for irrigation from treated (top) and untreated (bottom) municipal wastewater in 2017. Wastewater irrigation is defined by AQUASTAT as “wastewater applied artificially (irrigation) and directly (i.e., with no or little prior dilution with freshwater during most of the year) on land to assist the growth of crops and fruit trees. [Wastewater] applied artificially and directly for landscaping and forestry also falls under this category.” Units are:  $\log_{10}(\text{wastewater type used km}^3\text{year}^{-1} / \text{wastewater type produced km}^3\text{year}^{-1})$ . Gray countries indicate one or both of the wastewater used or wastewater produced variables were not available for that country. For Oman in the top panel and for Mexico in the bottom panel, ratios of used to produced wastewater were greater than one, potentially indicating an error in data reporting, and these values were set to missing. Data from AQUASTAT (FAO, 2021b). Data analysis and plotting conducted using R with ggplot2 (R Core Team, 2019; Wickham, 2016).

location, affected crop type, time, and method of application of waste resources in agriculture would be useful additions to the reported data for environmental exposure assessment.

Relationships between global changes and resultant chemical loading to agricultural settings should also be explored. Specifically, spatially-explicit modelling of how the use patterns of veterinary pharmaceuticals and pesticides may change in response to changing animal and plant pest and disease pressures under climate change is needed. Relationships between factors such as technological adoption or diet change and the patterns of chemical usage down to the field scale should be determined for extrapolation in future scenarios. Additionally, identification of new technologies that might be employed in agriculture in the future with a focus on the mixture of

chemicals that may be available on the European market in response to European “Toxic-free environment” policies is needed.

## 2.2. Understanding the effects of global change on chemical persistence and transformation

### 2.2.1. Overview

Temperature is a key factor that influences rates of chemical degradation and therefore chemical persistence (Matthies and Beulke, 2017). Regulatory tests that assess chemical persistence are conducted at one static temperature, often 12 °C or 20 °C, and chemical half-lives can then be extrapolated to other temperatures using the Arrhenius equation approach



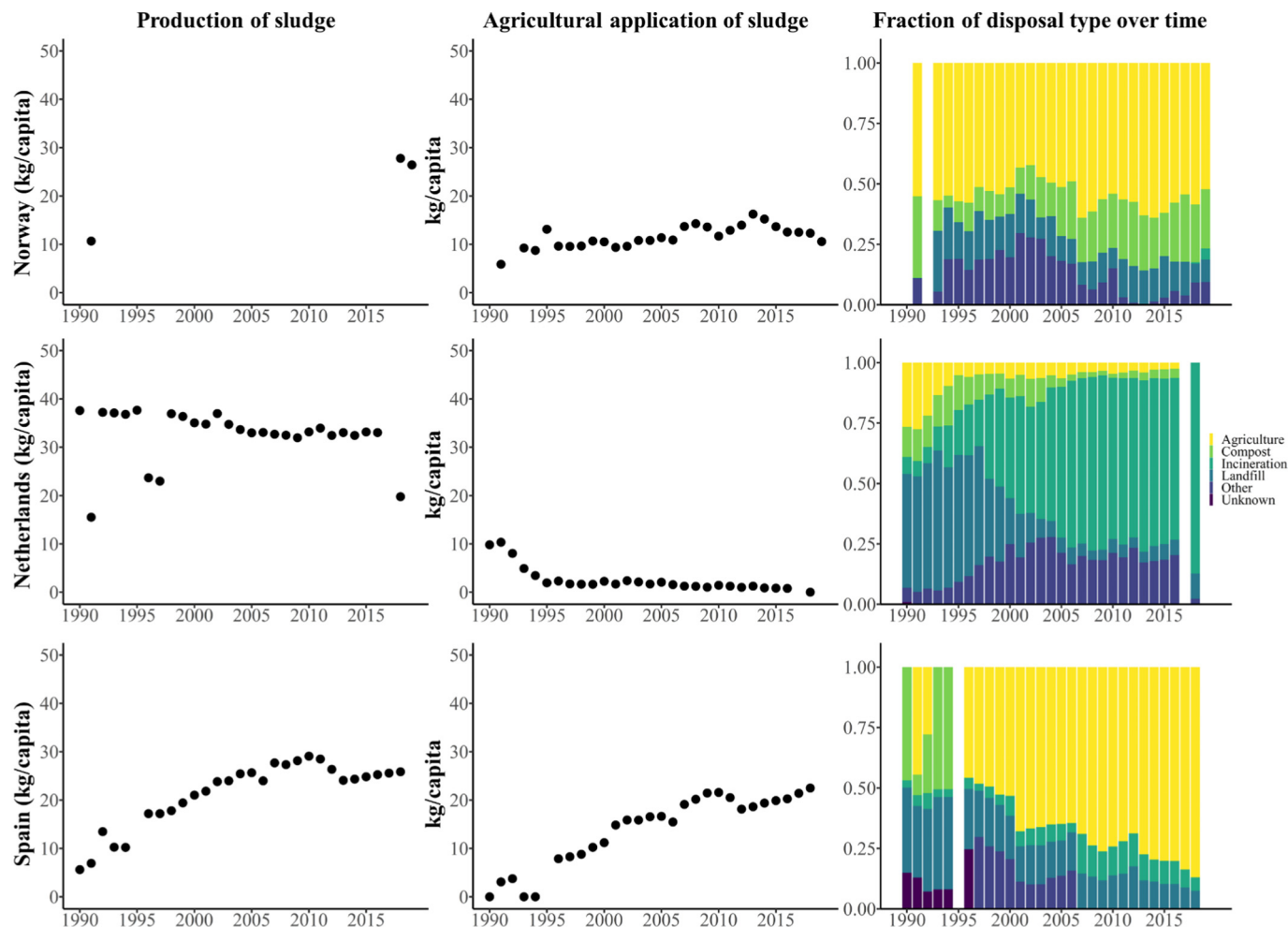


Fig. 5. Comparison of time series of dry weight sludge production and disposal patterns in Norway (top row), the Netherlands (middle row), and Spain (bottom row). Left column: per-capita sludge generation. Middle column: per-capita sludge disposal on agricultural fields. Right column: time series of fraction of different disposal methods of sludge. For the panels in the right column, the “Unknown” category makes up for discrepancies in the data between the total reported sludge (not shown) and the summed amount in the other provided categories (i.e., Agriculture, Compost, Incineration, Landfill, and Other). For any instances where the provided total reported sludge was less than the sum of the other provided categories, the provided categories are shown as-is. Geographic and temporal comparisons should be made with caution due to possible data gaps, breaks in reporting, and regional differences. Data from Eurostat, 2022. Data analysis and plotting conducted using R with ggplot2 (R Core Team, 2019; Wickham, 2016).

as suggested by the Forum for the Coordination of Pesticide Fate Models and their Use (FOCUS, 2006). Other environmental factors, such as intense precipitation, heatwaves, droughts, and how these conditions may impact soil conditions and the microbial communities responsible for chemical biodegradation are omitted from current persistence assessment methodologies. The FOCUS approach is recognized as being specific to pesticides and should be applied cautiously to other substances emitted to agricultural systems (Culleres et al., 2007).

The application of the Arrhenius approach across a temperature range of 4–40 °C was studied by Meynet et al. (2020) who determined the temperature dependence of degradation rates for 42 micropollutants in sewage. The authors reported exponential decreases in half-lives with inverse temperature as temperatures increased up to 20 °C, which would be well-described by the Arrhenius equation. However, the Arrhenius equation did not accurately describe the data as temperatures increased through 30 °C and 40 °C. At these higher temperatures, the Arrhenius equation tended to overestimate degradation rates, except for sulfonamides (Meynet et al., 2020). At the extreme lowest (4 °C) and highest (40 °C) temperatures there were also chemicals that did not demonstrate any notable dissipation over the course of the 120-day study (Meynet et al., 2020).

Modified versions of the Arrhenius equation, such as the quadratic Arrhenius or deformed Arrhenius, might provide better fits to degradation or biotransformation kinetics data when deviations from the classical

Arrhenius equation occur (Aquilanti et al., 2010). However, if temperature influences the nature of the microbiological community and therefore the capacity for degradation to take place, using modified forms of the Arrhenius equation might not allow for robust prediction, since the biological changes might not be reproducible as a function of temperature in different systems.

Aside from temperature, the biological impact of microbial community on chemical degradation is ambiguous. Fenner et al. (2020) have demonstrated that common routes of biodegradation can exist despite differences in microbial community structure. Coll et al. (2020) have reported that “fast” and “slow” chemical dissipation can be related to differences in microbial community composition, and that normalizing dissipation times according to microbial biomass can reduce variability in the kinetic measurements. When considering how seasonal variations in temperature could drive changes in microbial community composition in sediment and water and therefore in degradation rates of the fungicide isopyrazam, Southwell et al. (2020) were unable to find any linkages between microbial taxa and degradation rates among differences existing in community composition. Even with established differences in microbial community composition, a species was always present to metabolise isopyrazam (Southwell et al., 2020), suggesting that functional redundancy among microbial consortia might cause similar rates of chemical degradation despite there being differences in the microbial community composition (Fenner et al., 2020; Southwell et al., 2020).



Climate change-driven heat waves and droughts also introduce uncertainties to estimations of chemical persistence, but according to most reports they favour increased persistence. A report in the early 1990s indicated that pesticide application before or during a drought could lead to “carry-over” of pesticides via suppressed degradation (Leavitt et al., 1991). Subsequently, chemical degradation during a drought was demonstrated to be less efficient, as lack of water can disrupt soil conditions and substrates available for soil microbes to use (Franco-Andreu et al., 2016; Schimel et al., 2007). The combination of chemical exposure by the pesticides chlorpyrifos or oxyfluorfen and drought conditions resulted in reduced enzymatic activities and changes in the microbial community composition that reduced rates of degradation compared to watered soils (Franco-Andreu et al., 2016).

Chemical persistence under extensive precipitation leading to water-logged soils or flood-like conditions is another scenario that must be considered under climate change. Under water-logged conditions, soil can become anaerobic, which potentially leads to longer chemical half-lives (Biel-Maeso et al., 2019). Under anaerobic conditions, dissipation of carbamazepine, ibuprofen, diclofenac, hydrochlorothiazide, and gemfibrozil in agricultural soils was reportedly non-existent over 30 days (Biel-Maeso et al., 2019). In another study, Pan and Chu (2016) observed increased dissipation times for tetracycline, sulfamethazine, norfloxacin, and erythromycin under anaerobic soil conditions compared to aerobic soil conditions, both under sterile or non-sterile conditions.

### 2.2.2. Research needs to improve quantitative understanding of persistence and degradation pathways for organic chemicals in agriculture

Monitoring how soil composition and conditions such as temperature, moisture content, aerobic or anaerobic status, and microbial community makeup shift in response to global change and affect chemical persistence is needed. Degradation rates and routes of degradation for chemicals representative of varying physicochemical properties determined across the range of environmental factors identified above, including extreme temperatures, water-logged soils, and drought conditions, would provide insight regarding the importance of each environmental variable. These data can be assessed alongside current persistence testing and modelling approaches to provide insight on the implications of these findings for exposure assessment. Limitations in the Arrhenius equation approach when extrapolating chemical half-lives between temperatures and under varying soil conditions should also be investigated to determine if alternative approaches are more suitable. If degradation rates and routes are not predictable under dynamic environmental conditions, studies will be needed to determine if this is due to microbial community structure and functionality, or if there are other underlying abiotic factors influencing persistence under these conditions which could be used to inform the development of new predictive persistence models. Investigation of the persistence of chemicals found in reclaimed wastewater and biosolids applied to agricultural soils under environmental conditions where temperatures are beyond 30 °C, and in soils enduring drought conditions, is also needed (e.g. Zhu et al., 2021).

### 2.3. Capturing the impact of changes in extreme weather events and agricultural practices in chemical fate and transport modelling

#### 2.3.1. Overview

**2.3.1.1. Extreme events.** Intense precipitation events have been shown to have a substantial impact on the fate and transport of chemicals in agriculture in modelling and measurement studies of agriculturally-intensive watersheds. For pesticides, for example, Camenzuli et al. (2012) found that a quarter of the diuron applied annually within an Australian watershed was transferred to the ocean from rainwater/flooding. Morselli et al. (2018) similarly found that intense rainfall events in a mountainous Italian watershed resulted in increases of peak riverine pesticide concentrations up to several orders of magnitude. Yang et al. (2012) simulated episodic rainfall events on field plots containing land-applied biosolids, and found that the amount of hormone chemicals mobilized from biosolids correlated

with the simulated precipitation amount, indicating that heavy rainfall can result in a pulse of chemical runoff.

Given the documented sensitivity of agrochemical fate and transport to floods and other intense precipitation events, it is important to understand how increases in the intensity and frequency of such events with global change (see Fig. SI 1; EEA, 2017) may impact chemical fate and transport. Bloomfield et al. (2006) found that changes in the timing (i.e., seasonality) and intensity of precipitation patterns and increased temperatures would be the primary climate-related drivers of change in pesticide fate and transport. Since that study, many investigations have found a small effect of climate change-induced shifts in precipitation on pesticide fate, however a recurring theme in these studies is an acknowledged weakness in the forecasts of changes in intense precipitation events derived from downscaled global climate models (e.g., Gagnon et al., 2016; Chiu et al., 2017). Climate models used to project changes in short-term precipitation events often do not explicitly resolve localised, heavy, convective precipitation events at sub-daily timescales (EEA, 2017; see also Ban et al., 2015). For example, Gagnon et al. (2016) used downscaled climate modelling output to investigate pesticide mobilization. While they found climate change would not impact the mean pesticide runoff from a range of Canadian agricultural fields between 1981 and 2040, they indicated the climate model downscaling employed did not consider all possible causes of precipitation intensity increases (namely, intensity changes from more frequent convective precipitation). Similarly, Chiu et al. (2017) employed downscaled daily climate model output and the Soil and Water Assessment Tool (SWAT) model to investigate pesticide application, fate, and transport in a California watershed. “Few changes” from the direct impacts of climate change on pesticide transport were expected, however Chiu et al. indicate their method of downscaling the climate model output may not have captured future changes in extreme event frequency. Climate models that do explicitly resolve convective precipitation suggest an increase in intense precipitation events in many regions (e.g., Westra et al., 2014; see also Kendon et al., 2014). Our literature review identified no studies that investigated the impact of climate change on the fate and transport of organic chemicals present in reclaimed wastewater or biosolids applied to agricultural fields.

Boxall et al. (2009) identified the need to generate flood emersion models in the context of chemical exposure pathways for agricultural systems. Since their review, a handful of studies have developed modelling frameworks that investigate the impact of flood dynamics on chemical fate and transport (e.g., Mendez et al., 2016; Camenzuli et al., 2012). Camenzuli et al. (2012), for example, developed a model to simulate flooding within an Australian river basin and the impact of this on the mobilization of diuron. Information for parameterizing the water runoff occurring during saturated soil conditions was not available, so they employed a range of possibilities and they indicate the need to constrain “dominant mass transfer processes during flood events” more in the future.

The increased frequency of droughts due to climate change in Europe (Fig. SI 1; see also EEA, 2017) could also impact chemical fate and transport processes. For example, Ademollo et al. (2011) indicate that “temporary rivers”, which will increase in prevalence in the Mediterranean region from climate change, result in pulsed emissions of compounds upon return of precipitation. They indicate the chemical fate and transport processes in such transient rivers are not well understood, particularly regarding chemical-sediment dynamics during dry periods (see also Ribarova et al. (2008) regarding the impact of “first floods” on chemical fate). Topaz et al. (2018) also found that antecedent conditions (i.e., the time between rain events) may impact pesticide application methodology and thus the number and types of pesticides mobilized once rainfall occurs. Additionally, changes in drought dynamics as a result of climate change may increase the formation of desiccation cracks in clayey soils (e.g., Bordoloi et al., 2020), which can increase the infiltration rate of water into soils (e.g., Cheng et al., 2021) and impact the transport of chemicals in agriculture to groundwater.

Furthermore, Bloomfield et al. (2006) identified that increased temperatures from climate change may increase the volatilization and atmospheric transport of pesticides (however whether increased degradation would

outweigh the impacts of enhanced volatilization would vary as a function of chemical properties; e.g., see Wöhrnschimmel et al., 2013). Volatilization and atmospheric transport have been identified as a cause of pesticide contamination of remote areas, including regional glaciers (Rizzi et al., 2019; see also review by Noyes et al., 2009). Furthermore, the melting of contaminant-laden snow and ice in the spring can cause pulsed chemical emissions, and Morselli et al. (2014) indicate the impact climate change may have on snow and ice melting dynamics needs to be investigated in the context of contaminant transport and exposure (e.g., an earlier spring melt causing earlier increased runoff and exposure; see Falloon and Betts, 2006).

**2.3.1.2. Agricultural practices.** Amendment of soils with reclaimed wastewater and/or biosolids has been shown to impact the properties of soil (e.g., Jueschke et al., 2008; Rana et al., 2010; Skowrońska et al., 2020), including possibly decreasing pH and increasing organic carbon content. pH and organic carbon content have been found to be important factors influencing chemical fate in soils (e.g., Müller et al., 2007; Terzaghi et al., 2020), so changes in the agricultural application of waste resources in response to global change may impact the fate and transport of chemicals in agriculture. Dissolved organic carbon (DOC) supports microorganism activity (von Lütow and Kögel-Knabner, 2009) and plays an important role in chemical transport and regulation of bioavailability and degradability (Terzaghi et al., 2020, 2021). Likewise, the addition of microplastics through biosolid application could impact the fate and transport of associated chemicals through sorption mechanisms dependent on microplastic composition and physical properties (e.g., Gao et al., 2020).

No or minimum tillage has been proposed as an agricultural practice that can be used to adapt to some of the stresses of climate change, as it can increase soil moisture, soil organic carbon content, and soil resilience to erosion (EEA, 2019). Such practices can increase pesticide persistence due to higher retention in surface soil and lower accessibility for biodegradation, decrease runoff losses of pesticides due to enhanced soil stability, and increase leaching amounts due to enhanced macropore connectivity (Alletto et al., 2010). However, Alletto et al. also indicate that the highly variable and complex interactions of different factors surrounding pesticide fate and transport in the context of different tillage systems requires additional studies to fully understand the associated spatiotemporal dynamics.

Changes in agricultural wastewater irrigation techniques to increase water use efficiency could also impact the route by which contaminants enter into the environment and their subsequent fate and transport processes. For example, Bhalsod et al. (2018) found that overhead versus surface irrigation of lettuce plants using contaminant-laden irrigation water resulted in an increased uptake of several pharmaceuticals. Narain-Ford et al. (2020) reviewed the fate processes associated with contaminants of emerging concern for a range of different wastewater irrigation types. They found that runoff and volatilization are key processes associated with sprinkler irrigation, drip irrigation involves an insignificant amount of runoff, and that sub-surface irrigation involves an insignificant amount of runoff and volatilization but an increased importance of chemical sorption and potentially enhanced transport to groundwater. Narain-Ford et al. indicate, however, that there are still many unknowns surrounding the chemical fate processes associated with sub-surface irrigation.

### 2.3.2. Research needs to improve quantitative understanding of the impact of global change on the fate and transport of organic chemicals in agriculture

Modelling studies that can be validated against field observations are needed to investigate the impact of extreme weather events on the fate and transport of chemicals in agriculture, and to build confidence that the models can be applied for scenario analysis in the context of global change. Particularly, the use of more enhanced methods for downscaling climate model output than those used in many previous studies is needed to better capture how intense precipitation events mobilize chemicals in agriculture. Validated models that incorporate chemical fate and transport during river flooding of agricultural areas are also needed (Boxall et al., 2009), and could build off the progress of e.g., Camenzuli et al. (2012). Further

investigations are also needed into how increases in the time between precipitation events due to climate change will impact chemical emissions, fate, and transport (e.g., Topaz et al., 2018; Ademollo et al., 2011). Additional investigation is also needed into whether more frequent and intense heat waves will result in episodic increased volatilization and atmospheric transport of chemicals in agriculture (Bloomfield et al., 2006), with a focus on regional/medium-range transport (e.g., Rizzi et al., 2019). Lab and field studies are also needed to further understand the impact that different tillage practices (Alletto et al., 2010) and irrigation techniques employed with reclaimed wastewater (Narain-Ford et al., 2020) have on the fate and transport of organic chemicals.

## 3. Narrative scenarios for environmental exposure to chemicals in agriculture under global change

Scenario analysis for potential future pathways of development is well established as a decision-support framework. The IPCC's Shared Socio-Economic Pathway (SSP) scenarios (IPCC, 2021) for human development and greenhouse gas emissions (see also e.g., O'Neill et al., 2016; Riahi et al., 2017) is a notable example. Furthermore, Mitter et al. (2020) established a set of European agriculture SSPs (Eur-Agri-SSPs) reflecting plausible development scenarios in agricultural systems in response to climate change mitigation/adaptation. While their scenarios capture changes in urbanization, economic/technology development, and policies, they do not cover the impacts of the storylines on chemicals in agriculture that are needed to forecast possible changes in environmental exposures. Here, we augment the existing SSPs and Eur-Agri-SSPs to propose narrative Agricultural Chemical Exposure (ACE) scenarios that represent bounding and more realistic assumptions of combinations of the impact of global change drivers on agricultural chemical emissions, transformation, and fate and transport (see Table 1).

### 3.1. ACE-1 “sustainable development”

In the highly-optimistic “Sustainable Development” scenario ACE-1 (see Table 1), the combined effects of climate, technology, policy, and other global change drivers exert limited forcings on pathways for environmental exposure to chemicals in agriculture, either increasing exposures relatively little or decreasing exposures. From a climate perspective, this scenario would assume a rapid decrease in greenhouse gas emissions that leads to limiting 21st century global warming to approximately 1.5 °C relative to 1850–1900 (corresponding to SSP1-1.9; see IPCC, 2021) and a corresponding relatively low increase in pest pressures and extreme weather events. Furthermore, in response to EU policies like the Zero Pollution Action Plan (EC, 2021a), increased amounts of organic farming as part of the EU's “Farm to Fork” strategy (EC, 2020a), and additional development and implementation of pesticide-reducing technologies like precision agriculture (e.g., EEA, 2019) and novel agrochemical delivery methods (e.g., Kah and Hofmann, 2014), pesticide usage decreases in both absolute tonnage and tonnage applied per hectare of crops in the ACE-1 scenario. More chemicals employed in agriculture as well as in consumer products that are emitted down the drain will be developed on the “safe and sustainable by design” principle (e.g., EC, 2020c), decreasing chemical persistence, mobility, and toxicity. Wastewater treatment facilities would also have a greater adoption of technologies for the removal of organic chemicals (e.g., Sabri et al., 2020; Wolf et al., 2022), resulting in chemical emissions to agriculture through wastewater and biosolids decreasing. Pressures on water resources will also have a relatively low increase in response to climate change in this scenario, resulting in little increased need to reuse reclaimed wastewater for irrigation. In response to environmental concerns of animal agriculture (e.g., Willett et al., 2019), animal rearing for food decreases, with a corresponding decrease in the usage of veterinary pharmaceuticals (aided further by the EU's push to reduce the sale of animal and aquaculture antimicrobials; see EC, 2021a). There would also be an increase in the adoption of agricultural practices that mitigate chemical transport, like soil incorporation of pesticides and practices

**Table 1**  
Proposed Agricultural Chemical Exposure (ACE) scenarios and corresponding magnitudes<sup>a</sup> of global change drivers that would influence the emissions, persistence/transformation, and fate/transport of chemicals in agriculture.

ACE scenario	Corresponding SSP <sup>b</sup>	Climate change	Pesticide usage	Down-the-drain chemicals	Agri. WW & biosolids application	Vet. pharm. usage	Microbe-driven chemical degradation	Agricultural practices
1	SSP1- 1.9 “Sustainable Development”	- Rapid decrease in GHG emissions	- Decreases	- Decrease in down-the-drain emissions	Constant	- Animal agri. decreases	Increased chemical biodegradability	- Increased adoption of agri. mitigation techniques
		- 1.4 °C global temperature increase <sup>c</sup>	- Less persistent chemicals employed (e.g., ‘green’ chemistry)	- Less persistent and mobile chemicals used		- Use of vet. pharm. decreases		- Increased irrigation efficiency
2	SSP2-4.5 “Middle-of-the-road Development”	- Minimal increase in extreme weather events	- Widespread tertiary treatment					- Agri. land use decreases
		- Current GHG emissions, decreasing around mid-21 <sup>st</sup> century	- Increases	- Down-the-drain emissions constant	Increases	- Animal agri. stays constant	Current chemical biodegradability	- Current agri. mitigation techniques
3	SSP3-7.0 “Regional Rivalry”	- 2.7 °C global temperature increase <sup>c</sup>	- Current chemicals remain employed	- Current chemical composition		- Use of vet. pharm. stays constant		- Current irrigation efficiency
		- Increase in pest pressures		- WW treatment remains constant				- Current agri. land use
4	SSP5-8.5 “Fossil-fuelled Development”	- Increase in extreme weather events						- Some shifts in crops grown and locations
		- Increased GHG emissions	- Large increase	- Increase in down-the-drain emissions	Large increase	- Animal agri. increases	Decreased chemical biodegradability	Large shifts in crops grown and locations
5	SSP5-8.5 “Fossil-fuelled Development”	- 3.6 °C global temperature increase <sup>c</sup>	- Current chemicals remain employed	- Current chemical composition		- Use of vet. pharm. increases		
		- Large increase in pest pressures		- WW treatment remains constant				
6	SSP5-8.5 “Fossil-fuelled Development”	- Large increase in extreme weather events						
		- Substantially increased GHG emissions	- Increases substantially	- Urban down-the-drain emissions increase substantially	Increases substantially	- Animal agri. increases substantially	Substantially decreased chemical biodegradability	Substantial shifts in crops grown and locations
7	SSP5-8.5 “Fossil-fuelled Development”	- 4.4 °C global temperature increase <sup>c</sup>	- More persistent/harmful chemicals employed	- More persistent/mobile chemicals employed		- Use of vet. pharm. increases		
		- Substantial increase in pest pressures		- WW treatment remains constant		- Use of vet. pharm. increases substantially		
8	SSP5-8.5 “Fossil-fuelled Development”	- Substantial increase in extreme weather events						

<sup>a</sup>Increases or decreases in magnitude of variables are relative to current levels. Discussion of reasoning behind magnitude changes for bounding scenarios provided in [Section 3](#). <sup>b</sup>Numbering and naming conventions are those from [IPCC, 2021](#) and [Riahi et al. \(2017\)](#). <sup>c</sup>Global surface temperature increase by end of 21st century relative to 1850–1900 ([IPCC, 2021](#)). Abbreviations: ACE = Agricultural Chemical Exposure; Agri. = Agricultural; SSP = Shared Socio-economic Pathway; Vet. Pharm. = Veterinary pharmaceuticals; WW = Wastewater.

that reduce agricultural runoff (see Gagnon et al., 2016), along with an increase in irrigation efficiency (e.g., Nikolaou et al., 2020) and an overall decrease in agricultural land use (see Riahi et al., 2017).

### 3.2. ACE-4 “fossil-fuelled development”

At the other end of the spectrum in the highly-pessimistic “Fossil-fuelled Development” ACE-4 scenario (see Table 1), global change drivers combine to exert forcings on pathways that substantially increase environmental exposure to chemicals in agriculture. In this scenario, an increase in greenhouse gas emissions continues through 2050 and beyond, leading to 21st century global warming of approximately 4.4 °C relative to 1850–1900 (corresponding to SSP5-8.5; see IPCC, 2021). A substantial increase in average temperatures, shifts in precipitation, and extreme weather events would occur (see IPCC, 2021), leading to substantially increased pest and disease pressures and thus increased use of pesticides and veterinary pharmaceuticals (i.e., see Gale et al., 2007; Boxall et al., 2009; Grünig et al., 2020). Furthermore, wastewater treatment technologies in this scenario would be assumed to remain constant, so with increased urbanization (e.g., EEA, 2017) and an increased usage of pharmaceuticals (see van der Aa et al., 2011), down-the-drain emissions of chemicals will increase substantially. Within this scenario, EU policies to reduce the production and usage of harmful chemicals (e.g., EC, 2020c; EC, 2021a) are not adequately implemented, resulting in more persistent, mobile, and toxic chemical emissions to agricultural settings through the application of biosolids and wastewater for irrigation contaminated with down-the-drain emissions. Animal agriculture would also increase in response to increasing demand for meat globally (e.g., OECD/FAO, 2021; Bijl et al., 2017), resulting in veterinary pharmaceutical usage and emissions increasing substantially. Large shifts in the types of crops grown and an expansion of areas where crops are grown due to climate change would also occur (e.g., see Grünig et al., 2020), along with an increased usage of wastewater for irrigation in response to climate change-induced stresses on water resources (e.g., EEA, 2017). The “Regional Rivalry” ACE-3 scenario reflects slightly less intense global warming of 3.6 °C compared to ACE-4 (and therefore slightly lower associated impacts on increased chemical emissions), however ACE-3 would incorporate a larger increase in cropland (corresponding to SSP3-7.0; see IPCC, 2021 & Riahi et al., 2017).

The ACE-1 and ACE-4 scenarios provide bounds on the forecasts for agricultural chemical exposures, while the “middle-of-the-road” scenario ACE-2 (corresponding to SSP2-4.5) reflects roughly constant CO<sub>2</sub> emissions until the mid-21st century, economic and technical developments that generally stay with historical patterns (IPCC, 2021 & Riahi et al., 2017), and agricultural practices that generally reflect those of today (see Table 1 for more details).

## 4. Discussion and summary

The four narrative ACE scenarios we propose in Section 3 and the research priorities we identified in Section 2 provide a basis for developing quantitative scenarios to drive model simulations. Additional stakeholder consultation could be warranted to further develop and refine the four narrative ACE scenarios we have proposed here, for example as was done for the Eur-Agri-SSPs in Mitter et al., 2020. These ACE scenarios can then provide a basis for constructing quantitative estimates of emission rates for each chemical category of interest, which could be guided by the framework for establishing future emissions outlined in Desrousseaux et al. (in press). Generally, interactions between economic development, climate, agriculture, and land use could be modelled using the IMAGE integrated assessment model (e.g., Doelman et al., 2018), which could be used to elucidate future chemical emission rates. Nagesh et al., 2022, for example, have conducted a quantitative assessment of scenarios of chemical emissions to surface waters in Europe for five select pharmaceuticals, pesticides, and industrial chemicals. They identified key drivers of emissions (namely population, cropland area, and industrial production of steel/paper), related these drivers to chemical emissions, and employed the IMAGE modelling

framework to project quantitative changes in the drivers and thus chemical emissions under future socio-economic scenarios. Their approach could be extended to explicitly consider emissions of chemicals to agricultural environments, rather than surface water. Additionally, incorporation of direct and indirect effects of climate change (e.g., increased disease and pest pressures, increased stress on fresh water resources; see Section 2.1) were not included in the projections of Nagesh et al. but would need to be incorporated in the ACE scenarios. Fulfilment of the research needs associated with chemical emissions (Section 2.1.2) is needed, however, to inform such modelling to ensure the use of robust baseline emissions data and robust relationships between global change drivers and emissions (e.g., changes in pest and disease pressures and usage of pesticides and veterinary pharmaceuticals).

The IMAGE model can be a basis for projections of chemical usage and emissions based on socio-economic development scenarios. Chemical fate and transport models would then be needed to extend emissions of chemicals to exposure. Modelling of down-the-drain emissions of chemicals to the waste stream (e.g., Douziech et al., 2018; Grill et al., 2016) would be needed to construct scenarios of chemical flux to agricultural fields through the application of reclaimed wastewater and biosolids, which could incorporate use of the SimpleTreat wastewater treatment plant chemical fate model (Struijs, 2014). Simulations of chemical fate and transport using models like the SWAT model (e.g., Chiu et al., 2017) could then be employed with projected environmental and field conditions to obtain expected environmental chemical concentrations. Modelling at the agricultural field-scale and at sub-daily time scales would be required to fully capture the effects of short-duration extreme precipitation events and other direct impacts of climate change. Generic modelling at the watershed level, however, could be conducted with much lower computational cost for a range of watersheds in Europe to capture the variability in expected climate change impacts and the wide diversity of soil properties, land use types, slopes, and crops grown across Europe. Work associated with filling the research needs for how chemical persistence and degradation will be impacted by changing environmental conditions (see Section 2.2.2) would be a key input to these forward-looking chemical fate and transport simulations. Similarly, improved usage of climate model downscaling, models that more fully capture the impact of flooding on chemical fate and transport, and improved understanding of how changes in river flow dynamics impact chemical fate and transport will be needed (see Section 2.3.2) to comprehensively drive these modelling exercises.

The proposed ACE scenarios would result in forecasts of plausible future ranges of chemical exposure concentrations, which could be used by ecotoxicologists and policymakers to advance environmental protection goals under global change. As the ACE scenarios would result in exposure concentrations for single chemicals and chemical mixtures in aquatic and terrestrial environments, they could be used in studies of the impact of multiple stressors on ecological endpoints (e.g., Polazzo et al., 2021). The ACE scenarios could further help inform and support policy efforts that look to minimize the exposure of environmental endpoints to chemicals in agriculture by identifying the specific global change drivers that have the largest influence on environmental exposures to identify pre-emptive exposure mitigation strategies. Furthermore, the scenario analyses of wastewater and biosolids use that we advocate for would help expand the understanding of environmental exposures to the associated organic chemicals under a wide range of environmental conditions and use scenarios. This could provide additional insight into the possible safe and responsible use of these waste resources, and support part of the UN's Sustainable Development Goal of improving the safe reuse of wastewater (see Target 6.3; UN, 2018).

Over the coming decades, drivers of global change have the potential to shift the way chemicals in agriculture are released into and move around the environment. The potential ramifications of these global changes on environmental exposures to chemicals in agriculture are, however, poorly understood. We have reviewed existing literature related to how global change drivers may impact the emissions of organic chemicals in agriculture, their persistence and transformation, and their fate and transport.



We have identified knowledge gaps related to each of these three key themes, and charted a path forward for filling these gaps using both experimental and numerical modelling techniques. Filling these knowledge gaps will support the analysis of the proposed Agricultural Chemical Exposure (ACE) scenarios. Optimistic, status-quo, severe, and extreme scenarios are proposed, which will provide bounding and more realistic forecasts of how global change drivers may impact environmental exposure to agricultural chemicals. While this review focused on European agriculture, many of the identified knowledge gaps and the overall framework for addressing them apply at the global scale as well. This exposure forecasting framework aims to further our understanding of the impact global change may have on environmental exposure to chemicals in agriculture and elucidate measures that can be taken to avoid unacceptable risks, while also providing additional insight into the safe and responsible reuse of waste resources. Achieving this will help with the transition to a sustainable and circular economy while at the same time a toxic-free environment.

### CRediT authorship contribution statement

**John D. Hader:** Conceptualization, Software, Investigation, Visualization, Writing - original draft, Writing - review & editing. **Taylor Lane:** Conceptualization, Investigation, Visualization, Writing - original draft, Writing - review & editing. **Alistair B.A. Boxall:** Conceptualization, Writing - review & editing, Funding acquisition. **Matthew MacLeod:** Conceptualization, Writing - review & editing, Funding acquisition. **Antonio Di Guardo:** Conceptualization, Writing - review & editing.

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### Declaration of competing interest

The authors declare no conflicts of interest.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.156478>.

### References

- Ademollo, N., Capri, S., Patrolecco, L., Puddu, A., Polesello, S., Rusconi, M., Valsecchi, S., Froebrich, J., 2011. Fate and monitoring of hazardous substances in temporary rivers. *TrAC Trends Anal. Chem.* 30, 1222–1232. <https://doi.org/10.1016/j.trac.2011.05.002>.
- Alletto, A., Coquet, Y., Benoit, P., Heddadi, D., Barriuso, E., 2010. Tillage management effects on pesticide fate in soils. A review. *Agron. Sustain. Dev.* 30, 367–400. <https://doi.org/10.1051/agro/2009018>.
- Aquilanti, V., Mundim, K.C., Elango, M., Kleijn, S., Kasai, T., 2010. Temperature dependence of chemical and biophysical rate processes: phenomenological approach to deviations from Arrhenius law. *Chem. Phys. Lett.* 498, 209–213. <https://doi.org/10.1016/j.cplett.2010.08.035>.
- Aschemann-Witzel, J., Gantriis, R.F., Fraga, P., Perez-Cueto, F.J.A., 2021. Plant-based food and protein trend from a business perspective: markets, consumers, and the challenges and opportunities in the future. *Crit. Rev. Food Sci. Nutr.* 61, 1–10. <https://doi.org/10.1080/10408398.2020.1793730>.
- Ban, N., Schmidli, J., Schär, C., 2015. Heavy precipitation in a changing climate: does short-term summer precipitation increase faster? *Geophys. Res. Lett.* 42, 1165–1172. <https://doi.org/10.1002/2014GL062588>.
- Barclays Investment Bank, 2019. Carving up the alternative meat market [WWW Document]. <https://www.investmentbank.barclays.com/our-insights/carving-up-the-alternative-meat-market.html> accessed 6.24.21.
- Bhalsod, G.D., Chuang, Y.H., Jeon, S., Gui, W., Li, H., Ryser, E.T., Guber, A.K., Zhang, W., 2018. Uptake and accumulation of pharmaceuticals in overhead- and surface-irrigated greenhouse lettuce. *J. Agric. Food Chem.* 66, 822–830. <https://doi.org/10.1021/acs.jafc.7b04355>.
- Biel-Maeso, M., González-González, C., Lara-Martín, P.A., Corada-Fernández, C., 2019. Sorption and degradation of contaminants of emerging concern in soils under aerobic and anaerobic conditions. *Sci. Total Environ.* 666, 662–671. <https://doi.org/10.1016/j.scitotenv.2019.02.279>.
- Bijl, D.L., Bogaart, P.W., Dekker, S.C., Stehfest, E., de Vries, B.J.M., van Vuuren, D.P., 2017. A physically-based model of long-term food demand. *Glob. Environ. Chang.* 45, 47–62. <https://doi.org/10.1016/j.gloenvcha.2017.04.003>.
- BIO by Deloitte, 2015. Optimising Water Reuse in the EU – Final Report Prepared for the European Commission (DG ENV), Part I. In collaboration with ICF and Cranfield University <https://doi.org/10.2779/603205>.
- Bloomfield, J.P., Williams, R.J., Goody, D.C., Cape, J.N., Guha, P., 2006. Impacts of climate change on the fate and behaviour of pesticides in surface and groundwater—a UK perspective. *Sci. Total Environ.* 369, 163–177. <https://doi.org/10.1016/j.scitotenv.2006.05.019>.
- Bonanomi, G., Chirurazzi, M., Caporaso, S., Del Sorbo, G., Moschetti, G., Felice, S., 2008. Soil solarization with biodegradable materials and its impact on soil microbial communities. *Soil Biol. Biochem.* 40, 1989–1998. <https://doi.org/10.1016/j.soilbio.2008.02.009>.
- Bordoloi, S., Ni, J., Ng, C.W.W., 2020. Soil desiccation cracking and its characterization in vegetated soil: a perspective review. *Sci. Total Environ.* 729, 138760. <https://doi.org/10.1016/j.scitotenv.2020.138760>.
- Boxall, A.B.A., Kolpin, D.W., Halling-Sørensen, B., Tolls, J., 2003. Are veterinary medicines causing environmental risks? *Environ. Sci. Technol.* 37, 286A–294A. <https://doi.org/10.1021/es032519b>.
- Boxall, A.B.A., Hardy, A., Beulke, S., Boucard, T., Burgin, L., Falloon, P.D., Haygarth, P.M., Hutchinson, T., Kovats, R.S., Leonardi, G., Levy, L.S., Nichols, G., Parsons, S.A., Potts, L., Stone, D., Topp, E., Turley, D.B., Walsh, K., Wellington, E.M.H., Williams, R.J., 2009. Impacts of climate change on indirect human exposure to pathogens and chemicals from agriculture. *Environ. Health Perspect.* 117, 508–514. <https://doi.org/10.1289/ehp.0800084>.
- Camenzuli, L., Scheringer, M., Gaus, C., Ng, C.A., Hungerbühler, K., 2012. Describing the environmental fate of diuron in a tropical river catchment. *Sci. Total Environ.* 440, 178–185. <https://doi.org/10.1016/j.scitotenv.2012.07.037>.
- Carter, L.J., Chefetz, B., Abdeen, Z., Boxall, A.B.A., 2019. Emerging investigator series: towards a framework for establishing the impacts of pharmaceuticals in wastewater irrigation systems on agro-ecosystems and human health. *Environ. Sci. Process. Impacts* 21, 605–622. <https://doi.org/10.1039/c9em00020h>.
- Challis, J.K., Sura, S., Cantin, J., Curtis, A.W., Shade, K.M., McAllister, T.A., Jones, P.D., Giesy, J.P., Larney, F.J., 2021. Ractopamine and other growth-promoting compounds in beef cattle operations: fate and transport in feedlot pens and adjacent environments. *Environ. Sci. Technol.* 55, 1730–1739. <https://doi.org/10.1021/acs.est.0c06450>.
- Cheng, Q., Tang, C.S., Xu, D., Zeng, H., Shi, B., 2021. Water infiltration in a cracked soil considering effect of drying-wetting cycles. *J. Hydrol.* 593, 125640. <https://doi.org/10.1016/j.jhydrol.2020.125640>.
- Chiu, M.C., Hunt, L., Resh, V.H., 2017. Climate-change influences on the response of macro-invertebrate communities to pesticide contamination in the Sacramento River, California watershed. *Sci. Total Environ.* 581–582, 741–749. <https://doi.org/10.1016/j.scitotenv.2017.01.002>.
- Christou, A., Agüera, A., Bayona, J.M., Cytryn, E., Fotopoulos, V., Lambropoulou, D., Manaia, C.M., Michael, C., Revitt, M., Schröder, P., Fatta-Kassinos, D., 2017. The potential implications of reclaimed wastewater reuse for irrigation on the agricultural environment: the knowns and unknowns of the fate of antibiotics and antibiotic resistant bacteria and resistance genes – a review. *Water Res.* 123, 448–467. <https://doi.org/10.1016/j.watres.2017.07.004>.
- Coll, C., Bier, R., Li, Z., Langenheder, S., Gorokhova, E., Sobek, A., 2020. Association between aquatic micropollutant dissipation and river sediment bacterial communities. *Environ. Sci. Technol.* 54, 14380–14392. <https://doi.org/10.1021/acs.est.0c04393>.
- Collivignarelli, M.C., Abbà, A., Frattarola, A., Miino, M.C., Padovani, S., Katsoyiannis, I., Torretta, V., 2019. Legislation for the reuse of biosolids on agricultural land in Europe: overview. *Sustainability* 11, 1–22. <https://doi.org/10.3390/su11216015>.
- Corradini, F., Meza, P., Eguiluz, R., Casado, F., Huerta-Lwanga, E., Geissen, V., 2019. Evidence of microplastic accumulation in agricultural soils from sewage sludge disposal. *Sci. Total Environ.* 671, 411–420. <https://doi.org/10.1016/j.scitotenv.2019.03.368>.
- Culleres, D.B., Boesten, J., Bolognesi, C., Boobis, A., Buchert, A., Capri, E., Coggon, D., Hardy, A., Hart, A., Koepp, H., Liess, M., Luttik, R., Meyer, O., Michaelidou-Canna, S., Montforts, M., Moretto, A., Mueller, M.D., Ossendorp, B., Steurbaut, W., Tasheva, M., Vlemminckx, C., 2007. Scientific Opinion of the Panel on Plant Protection Products and their Residues on a request from EFSA related to the default Q10 value used to describe the temperature effect on transformation rates of pesticides in soil. *EFSA J.* 622, 1–32. <https://doi.org/10.2903/j.efsa.2008.622>.
- de Souza, R.M., Seibert, D., Quesada, H.B., de Jesus Bassetti, F., Fagundes-Klen, M.R., Bergamasco, R., 2020. Occurrence, impacts and general aspects of pesticides in surface water: a review. *Process Saf. Environ. Prot.* 135, 22–37. <https://doi.org/10.1016/j.psep.2019.12.035>.
- Delcour, I., Spanoghe, P., Uyttendaele, M., 2015. Literature review: impact of climate change on pesticide use. *Food Res. Int.* 68, 7–15. <https://doi.org/10.1016/j.foodres.2014.09.030>.
- Desrousseaux, A.O.S., et al., 2022. A shared socio-economic pathway based framework for characterising future emissions of chemicals to the natural environment. *Futures*. In press.

- Doelman, J.C., Stehfest, E., Tabeau, A., van Meijl, H., Lassaletta, L., Gernaat, D.E.H.J., Neumann-Hermans, K., Harmsen, M., Daigoulou, V., Biemans, H., van der Sluis, S., van Vuuren, D.P., 2018. Exploring SSP land-use dynamics using the IMAGE model: regional and gridded scenarios of land-use change and land-based climate change mitigation. *Glob. Environ. Chang.* 48, 119–135. <https://doi.org/10.1016/j.gloenvcha.2017.11.014>.
- Douziech, M., van Zelm, R., Oldenkamp, R., Franco, A., Hendriks, A.J., King, H., Huijbregts, M.A.J., 2018. Estimation of chemical emissions from down-the-drain consumer products using consumer survey data at a country and wastewater treatment plant level. *Chemosphere* 193, 32–41. <https://doi.org/10.1016/j.chemosphere.2017.11.009>.
- EC (European Commission), 2020a. Farm to Fork Strategy for a fair, healthy and environmentally-friendly food system. [https://ec.europa.eu/food/system/files/2020-05/f2f\\_action-plan\\_2020\\_strategy-info\\_en.pdf](https://ec.europa.eu/food/system/files/2020-05/f2f_action-plan_2020_strategy-info_en.pdf) <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32009R1107&from=EN>.
- EC (European Commission), 2020b. New Circular Economy Action Plan for a cleaner and more competitive Europe. COM(2020) 98 final. <https://eur-lex.europa.eu/legal-content/EN/TXT/?qid=1583933814386&uri=COM:2020:98:FIN>.
- EC (European Commission), 2020c. Chemicals strategy for sustainability towards a toxic-free environment. COM(2020) 667 final. <https://ec.europa.eu/environment/pdf/chemicals/2020/10/Strategy.pdf>.
- EC (European Commission), 2021a. Pathway to a healthy planet for all. EU action plan: "Towards zero pollution for air, water and soil." COM(2021) 400 final. [https://eur-lex.europa.eu/resource.html?uri=cellar:a1c34a56-b314-11eb-8aca-01aa75ed71a1.0001.02/DOC\\_1&format=PDF](https://eur-lex.europa.eu/resource.html?uri=cellar:a1c34a56-b314-11eb-8aca-01aa75ed71a1.0001.02/DOC_1&format=PDF).
- EC (European Commission), 2021b. Authorising certain products and substances for use in organic production and establishing their lists. C(2021) 5149 final. [https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/12377-Organic-farming-list-of-products-&-substances-authorised-in-organic-production-update\\_en](https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/12377-Organic-farming-list-of-products-&-substances-authorised-in-organic-production-update_en).
- EEA (European Environment Agency), 2017. Climate Change, Impacts And Vulnerability in Europe 2016. <https://doi.org/10.2800/534806> Luxembourg.
- EEA (European Environment Agency), 2019. Climate Change Adaptation in the Agriculture Sector in Europe. EEA Rep. 112.
- EMA (European Medicines Agency), 2020. European Surveillance of Veterinary Antimicrobial Consumption. 'Sales of Veterinary Antimicrobial Agents in 31 European Countries in 2018'. (EMA/24309/2020).
- EPPO (European and Mediterranean Plant Protection Organization), 2022. Spodoptera frugiperda. EPPO datasheets on pests recommended for regulation. Available online <https://gd.eppo.int>.
- Eurostat, 2021. Organic farming statistics. [https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Organic\\_farming\\_statistics](https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Organic_farming_statistics).
- Eurostat, 2022. Sewage sludge production and disposal (env\_ww\_spd). [Dataset]. URL <https://ec.europa.eu/eurostat/web/environment/water> (accessed 2.22.22).
- Falloon, P.D., Betts, R.A., 2006. The impact of climate change on global river flow in HadGEM1 simulations. *Atmos. Sci. Lett.* 7, 62–68. <https://doi.org/10.1002/asl.133>.
- FAO, 2020. Land use in agriculture by the numbers. <http://www.fao.org/sustainability/news/detail/en/c/1274219/>.
- FAO, 2021a. Data ("Fertilizers by Nutrient", "Land Use", "Production Indices", and "Pesticides Use"). URL <http://www.fao.org/faostat/en/#data> (accessed 8.2.22).
- FAO, 2021b. AQUASTAT Database. [Dataset]. URL <http://www.fao.org/aquastat/statistics/query/index.html?lang=en> (accessed 15.9.21).
- FAO (Food and Agriculture Organization of the United Nations), 2017. Water for Sustainable Food And Agriculture Rome. ISBN 978-92-5-109977-3. <https://www.fao.org/3/i7959e/i7959e.pdf>.
- Fenner, K., Screpanti, C., Renold, P., Rouchdi, M., Vogler, B., Rich, S., 2020. Comparison of small molecule biotransformation half-lives between activated sludge and soil: opportunities for read-across? *Environ. Sci. Technol.* 54, 3148–3158. <https://doi.org/10.1021/acs.est.9b05104>.
- FOCUS (Forum for the coordination of pesticide fate models and their use), 2006. Guidance Document on Estimating Persistence and Degradation Kinetics from Environmental Fate Studies on Pesticides in EU Registration" Report of the FOCUS Work Group on Degradation Kinetics. URL (accessed 8.2.22).
- Franco-Andreu, L., Gómez, I., Parrado, J., García, C., Hernández, T., Tejada, M., 2016. Behavior of two pesticides in a soil subjected to severe drought. Effects on soil biology. *Appl. Soil Ecol.* 105, 17–24. <https://doi.org/10.1016/j.apsoil.2016.04.001>.
- Gagnon, P., Sheedy, C., Rousseau, A.N., Bourgeois, G., Chouinard, G., 2016. Integrated assessment of climate change impact on surface runoff contamination by pesticides. *Integr. Environ. Assess. Manag.* 12, 559–571. <https://doi.org/10.1002/ieam.1706>.
- Gale, P., Adkin, A., Snary, E., Cheung, S., Wooldridge, M., 2007. Assessing And Managing the Impacts of Climate Change on the Environmental Risks of Agricultural Pathogens And Contaminants (SD0441).
- Gao, D., Li, X., Yu, L., H. tao, 2020. Source, occurrence, migration and potential environmental risk of microplastics in sewage sludge and during sludge amendment to soil. *Sci. Total Environ.* 742, 140355. <https://doi.org/10.1016/j.scitotenv.2020.140355>.
- González-Gaya, B., Lopez-Herguedas, N., Bilbao, D., Mijangos, L., Iker, A.M., Etxebarria, N., Irazola, M., Prieto, A., Olivares, M., Zuloaga, O., 2021. Suspect and non-target screening: the last frontier in environmental analysis. *Anal. Methods* 13, 1876–1904. <https://doi.org/10.1039/d1ay00111f>.
- Grill, G., Khan, U., Lehner, B., Nicell, J., Ariwi, J., 2016. Risk assessment of down-the-drain chemicals at large spatial scales: model development and application to contaminants originating from urban areas in the Saint Lawrence River Basin. *Sci. Total Environ.* 541, 825–838. <https://doi.org/10.1016/j.scitotenv.2015.09.100>.
- Grill, G., Li, J., Khan, U., Zhong, Y., Lehner, B., Nicell, J., Ariwi, J., 2018. Estimating the ecotoxicological risk of estrogens in China's rivers using a high-resolution contaminant fate model. *Water Res.* 145, 707–720. <https://doi.org/10.1016/j.watres.2018.08.053>.
- Grünig, M., Mazzi, D., Calanca, P., Karger, D.N., Pellissier, L., 2020. Crop and forest pest metawebs shift towards increased linkage and suitability overlap under climate change. *Commun. Biol.* 3, 1–10. <https://doi.org/10.1038/s42003-020-0962-9>.
- IPCC, 2021. Summary for policymakers. In: Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S.L., Péan, C., Berger, S., Caud, N., Chen, Y., Goldfarb, L., Gomis, M.J., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J.B.R., Maycock, T.K., Waterfield, T., Yelekçi, O., Yu, R., Zhou, B. (Eds.), *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press. In Press.
- Jueschke, E., Marschner, B., Tarchitzky, J., Chen, Y., 2008. Effects of treated wastewater irrigation on the dissolved and soil organic carbon in Israeli soils. *Water Sci. Technol.* 57, 727–733. <https://doi.org/10.2166/wst.2008.173>.
- Kah, M., Hofmann, T., 2014. Nanopesticide research: current trends and future priorities. *Environ. Int.* 63, 224–235. <https://doi.org/10.1016/j.envint.2013.11.015>.
- Kendon, E.J., Roberts, N.M., Fowler, H.J., Roberts, M.J., Chan, S.C., Senior, C.A., 2014. Heavier summer downpours with climate change revealed by weather forecast resolution model. *Nat. Clim. Chang.* 4, 570–576. <https://doi.org/10.1038/nclimate2258>.
- Kilgallon, J., Franco, A., Price, O.R., Hodges, J.E.N., 2017. Application of a spatially resolved model to contextualise monitoring data for risk assessment of down-the-drain chemicals over large scales. *Environ. Pollut.* 230, 954–962. <https://doi.org/10.1016/j.envpol.2017.07.041>.
- Kookana, R.S., Boxall, A.B.A., Reeves, P.T., Ashauer, R., Beulke, S., Chaudhry, Q., Cornelis, G., Fernandes, T.F., Gan, J., Kah, M., Lynch, I., Ranville, J., Sinclair, C., Spurgeon, D., Tiede, K., Van Den Brink, P.J., 2014. Nanopesticides: guiding principles for regulatory evaluation of environmental risks. *J. Agric. Food Chem.* 62, 4227–4240. <https://doi.org/10.1021/jf500232f>.
- Kuppusamy, S., Kakarla, D., Venkateswarlu, K., Megharaj, M., Yoon, Y.E., Lee, Y.B., 2018. Veterinary antibiotics (VAs) contamination as a global agro-ecological issue: a critical view. *Agric. Ecosyst. Environ.* 257, 47–59. <https://doi.org/10.1016/j.agee.2018.01.026>.
- Leavitt, R.A., Kells, J.J., Bunkelmann, J.R., Hollingworth, R.M., 1991. Assessing atrazine persistence in soil following a severe drought. *Bull. Environ. Contam. Toxicol.* 46, 22–29. <https://doi.org/10.1007/BF01688250>.
- Maggi, F., Tang, F.H.M., la Cecilia, D., McBratney, A., 2019. PEST-CHEMGRIDS, global gridded maps of the top 20 crop-specific pesticide application rates from 2015 to 2025. *Sci. Data* 6, 1–20. <https://doi.org/10.1038/s41597-019-0169-4>.
- Masseroni, D., Ricart, S., de Cartagena, F.R., Monserrat, J., Gonçalves, J.M., de Lima, I., Facchi, A., Sali, G., Gandolfi, C., 2017. Prospects for improving gravity-fed surface irrigation systems in Mediterranean European contexts. *Water (Switzerland)* 9. <https://doi.org/10.3390/w9010020>.
- Matthies, M., Beulke, S., 2017. Considerations of temperature in the context of the persistence classification in the EU. *Environ. Sci. Eur.* 29, 1–11. <https://doi.org/10.1186/s12302-017-0113-1>.
- Meier, M.S., Stoessel, F., Jungbluth, N., Juraske, R., Schader, C., Stolze, M., 2015. Environmental impacts of organic and conventional agricultural products - are the differences captured by life cycle assessment? *J. Environ. Manag.* 149, 193–208. <https://doi.org/10.1016/j.jenvman.2014.10.006>.
- Mendez, A., Ng, C.A., Torres, J.P.M., Bastos, W., Bogdal, C., dos Reis, G.A., Hungerbuehler, K., 2016. Modeling the dynamics of DDT in a remote tropical floodplain: indications of post-ban use? *Environ. Sci. Pollut. Res.* 23, 10317–10334. <https://doi.org/10.1007/s11356-015-5641-x>.
- Meynet, P., Davenport, R.J., Fenner, K., 2020. Understanding the dependence of micropollutant biotransformation rates on short-term temperature shifts. *Environ. Sci. Technol.* 54, 12214–12225. <https://doi.org/10.1021/acs.est.0c04017>.
- Mitter, H., Techen, A., Sinabell, F., Helming, K., Schmid, E., Bodirsky, B.L., Holman, I., Kok, K., Lehtonen, H., Leip, A., Le, C., Mathijs, E., Mehdi, B., Mittenzwei, K., Mora, O., Østada, K., Øygarden, L., Priess, J.A., Reidsma, P., Schaldach, R., Schönhart, M., 2020. Shared socio-economic pathways for European agriculture and food systems: the Eur-Agri-SSPs. *Glob. Environ. Chang.* 65, 102159. <https://doi.org/10.1016/j.gloenvcha.2020.102159>.
- Morselli, M., Semplice, M., Villa, S., Di Guardo, A., 2014. Evaluating the temporal variability of concentrations of POPs in a glacier-fed stream food chain using a combined modeling approach. *Sci. Total Environ.* 493, 571–579. <https://doi.org/10.1016/j.scitotenv.2014.05.150>.
- Morselli, M., Vitale, C.M., Ippolito, A., Villa, S., Giacchini, R., Vighi, M., Di Guardo, A., 2018. Predicting pesticide fate in small cultivated mountain watersheds using the DynAPlus model: toward improved assessment of peak exposure. *Sci. Total Environ.* 615, 307–318. <https://doi.org/10.1016/j.scitotenv.2017.09.287>.
- Müller, K., Magesan, G.N., Bolan, N.S., 2007. A critical review of the influence of effluent irrigation on the fate of pesticides in soil. *Agric. Ecosyst. Environ.* 120, 93–116. <https://doi.org/10.1016/j.agee.2006.08.016>.
- Nagesh, P., Boer, H.J., De Wezel, A.P., Van Dekker, S.C., Vuuren, D.P., 2022. Development of chemical emission scenarios using the Shared Socio-economic Pathways. *Sci. Total Environ.* 155530. <https://doi.org/10.1016/j.scitotenv.2022.155530>.
- Narain-Ford, D.M., Bartholomew, R.P., Dekker, S.C., van Wezel, A.P., 2020. Natural purification through soils: risks and opportunities of sewage effluent reuse in sub-surface irrigation. [https://doi.org/10.1007/398\\_2020\\_49](https://doi.org/10.1007/398_2020_49).
- Nikolaou, G., Neocleous, D., Christou, A., Kitta, E., Katsoulas, N., 2020. Implementing sustainable irrigation in water-scarce regions under the impact of climate change. *Agronomy* 10, 1120. <https://doi.org/10.3390/agronomy10081120>.
- Noyes, P.D., McElwee, M.K., Miller, H.D., Clark, B.W., Van Tiem, L.A., Walcott, K.C., Erwin, K.N., Levin, E.D., 2009. The toxicology of climate change: environmental contaminants in a warming world. *Environ. Int.* 35, 971–986. <https://doi.org/10.1016/j.envint.2009.02.006>.
- O'Neill, B.C., Tebaldi, C., Van Vuuren, D.P., Eyring, V., Friedlingstein, P., Hurtt, G., Knutti, R., Kriegler, E., Lamarque, J.F., Lowe, J., Meehl, G.A., Moss, R., Riahi, K., Sanderson, B.M., 2016. The Scenario Model Intercomparison Project (ScenarioMIP) for CMIP6. *Geosci. Model Dev.* 9, 3461–3482. <https://doi.org/10.5194/gmd-9-3461-2016>.
- OECD/FAO, 2021. Meat. OECD-FAO Agricultural Outlook 2021–2030. OECD Publishing, Paris. <https://doi.org/10.1787/cf68bf79-en>.



- Pan, M., Chu, L.M., 2016. Adsorption and degradation of five selected antibiotics in agricultural soil. *Sci. Total Environ.* 545–546, 48–56. <https://doi.org/10.1016/j.scitotenv.2015.12.040>.
- Paranychniak, N.V., Salgot, M., Snyder, S.A., Angelakis, A.N., 2015. Water reuse in EU states: necessity for uniform criteria to mitigate human and environmental risks. *Crit. Rev. Environ. Sci. Technol.* 45, 1409–1468. <https://doi.org/10.1080/10643389.2014.955629>.
- Polazzo, F., Roth, S.K., Hermann, M., Mangold-Döring, A., Rico, A., Sobek, A., Van den Brink, P.J., Jackson, M.C., 2021. Combined effects of heatwaves and micropollutants on freshwater ecosystems: towards an integrated assessment of extreme events in multiple stressors research. *Glob. Chang. Biol.* 1–20. <https://doi.org/10.1111/gcb.15971>.
- Poore, J., Nemecek, T., 2018. Reducing food's environmental impacts through producers and consumers. *Science* (80-) 360, 987–992. <https://doi.org/10.1126/science.aag0216>.
- R Core Team, 2019. R: A Language And Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. <https://www.R-project.org/>.
- Rana, L., Dhankhar, R., Chhikara, S., 2010. Soil characteristics affected by long term application of sewage wastewater. *Int. J. Environ. Res.* 4, 513–518. <https://doi.org/10.22059/ijer.2010.237>.
- Regulation (EC) No 1107/2009, 2009. of the European Parliament and of the Council of 21 October 2009 concerning the placing of plant protection products on the market and repealing Council Directives 79/117/EEC and 91/414/EEC. *Off. J. Eur. Union* 309, 1–50. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32009R1107&from=EN>.
- Regulation (EC) no 1907/2006, 2006. of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), establishing a European Chemicals Agency, amending Directive 1999/45/EC and repealing Council Regulation (EEC) No 793/93 and Commission Regulation (EC) No 1488/94 as well as Council Directive 76/769/EEC and Commission Directives 91/155/EEC, 93/67/EEC, 93/105/EC and 2000/21/EC. *Off. J. Eur. Union* 396, 1–849. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32006R1907&from=EN>.
- Regulation (EU) 2019/6, 2019. of the European Parliament and of the Council of 11 December 2018 on veterinary medicinal products and repealing Directive 2001/82/EC. *Off. J. Eur. Union* 4, 43–167. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019R0006&from=EN>.
- Regulation (EU) 2020/741, 2020. of the European Parliament and of the Council of 25 May 2020 on minimum requirements for water reuse. *Off. J. Eur. Union* 177, 32–55. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32020R0741&from=EN>.
- Regulation (EU) No 528/2012, 2012. of the European Parliament and of the Council of 22 May 2012 concerning the making available on the market and use of biocidal products. *Off. J. Eur. Union* 167, 1–123. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32012R0528&from=EN>.
- Riahi, K., van Vuuren, D.P., Kriegler, E., Edmonds, J., O'Neill, B.C., Fujimori, S., Bauer, N., Calvin, K., Dellink, R., Fricko, O., Lutz, W., Popp, A., Cuaresma, J.C., KC, S., Leimbach, M., Jiang, L., Kram, T., Rao, S., Emmerling, J., Ebi, K., Hasegawa, T., Havlik, P., Humpenöder, F., Da Silva, L.A., Smith, S., Stehfest, E., Bosetti, V., Eom, J., Gernaat, D., Masui, T., Rogelj, J., Streffer, J., Drouet, L., Krey, V., Luderer, G., Harmsen, M., Takahashi, K., Baumstark, L., Doelman, J.C., Kainuma, M., Klimont, Z., Marangoni, G., Lotze-Campen, H., Obersteiner, M., Tabeau, A., Tavoni, M., 2017. The Shared Socioeconomic Pathways and their energy, land use, and greenhouse gas emissions implications: an overview. *Glob. Environ. Chang.* 42, 153–168. <https://doi.org/10.1016/j.gloenvcha.2016.05.009>.
- Ribarova, I., Ninov, P., Cooper, D., 2008. Modeling nutrient pollution during a first flood event using HSPF software: Iskar River case study, Bulgaria. *Ecol. Modell.* 211, 241–246. <https://doi.org/10.1016/j.ecolmodel.2007.09.022>.
- Rizzi, C., Finizio, A., Maggi, V., Villa, S., 2019. Spatial-temporal analysis and risk characterisation of pesticides in alpine glacial streams. *Environ. Pollut.* 248, 659–666. <https://doi.org/10.1016/j.envpol.2019.02.067>.
- Sabri, N.A., van Holst, S., Schmitt, H., van der Zaan, B.M., Gerritsen, H.W., Rijnaarts, H.H.M., Langenhoff, A.A.M., 2020. Fate of antibiotics and antibiotic resistance genes during conventional and additional treatment technologies in wastewater treatment plants. *Sci. Total Environ.* 741, 140199. <https://doi.org/10.1016/j.scitotenv.2020.140199>.
- Sarmah, A.K., Meyer, M.T., Boxall, A.B.A., 2006. A global perspective on the use, sales, exposure pathways, occurrence, fate and effects of veterinary antibiotics (VAs) in the environment. *Chemosphere* 65, 725–759. <https://doi.org/10.1016/j.chemosphere.2006.03.026>.
- Schimel, J., Balser, T.C., Wallenstein, M., 2007. Microbial stress-response physiology and its implications for ecosystem function. *Ecology* 88, 1386–1394. <https://doi.org/10.1890/06-0219>.
- Schulz, R., Bub, S., Petschick, L.L., Stehle, S., Wolfram, J., 2021. Applied pesticide toxicity shifts toward plants and invertebrates, even in GM crops. *Science* (80-) 372, 81–84. <https://doi.org/10.1126/science.abe1148>.
- Silva, V., Mol, H.G.J., Zomer, P., Tienstra, M., Ritsema, C.J., Geissen, V., 2019. Pesticide residues in European agricultural soils – a hidden reality unfolded. *Sci. Total Environ.* 653, 1532–1545. <https://doi.org/10.1016/j.scitotenv.2018.10.441>.
- Skowrońska, M., Bielińska, E.J., Szymański, K., Futa, B., Antoniewicz, J., Kołodziej, B., 2020. An integrated assessment of the long-term impact of municipal sewage sludge on the chemical and biological properties of soil. *Catena* 189, 104484. <https://doi.org/10.1016/j.catena.2020.104484>.
- Smart Protein Project, 2021. Plant-based foods in Europe: How big is the market? Smart Protein Plant-based Food Sector Report. European Union's Horizon 2020 research and innovation programme (No 862957). <https://smartproteinproject.eu/plant-based-food-sector-report>.
- Southwell, R.V., Hilton, S.L., Pearson, J.M., Hand, L.H., Bending, G.D., 2020. Inclusion of seasonal variation in river system microbial communities and phototroph activity increases environmental relevance of laboratory chemical persistence tests. *Sci. Total Environ.* 733, 139070. <https://doi.org/10.1016/j.scitotenv.2020.139070>.
- Steffen, W., Crutzen, P.J., McNeill, J.R., 2007. The anthropocene: are humans now overwhelming the great forces of nature? *Ambio* 36, 614–621. [https://doi.org/10.1579/0044-7447\(2007\)36\[614:TAHNO\]2.0.CO;2](https://doi.org/10.1579/0044-7447(2007)36[614:TAHNO]2.0.CO;2).
- Struijs, J., 2014. SimpleTreat 4.0: A Model to Predict Fate And Emission of Chemicals in Wastewater Treatment Plants. In Press.
- Tavazzi, S., Locoro, G., Comerio, S., Sobiecka, E., Loos, R., Gans, O., Ghiani, M., Umlauf, G., Suurkuusk, G., Paracchini, B., Cristache, C., Fissiaux, I., Riuz, A.A., Gawlik, B.M., 2012. Occurrence and levels of selected compounds in European sewage sludge samples (FATE SEES). <https://doi.org/10.2788/67153>.
- Terzaghi, E., Vitale, C.M., Di Guardo, A., 2020. Modelling peak exposure of pesticides in terrestrial and aquatic ecosystems: importance of dissolved organic carbon and vertical particle movement in soil. *SAR QSAR Environ. Res.* 31, 19–32. <https://doi.org/10.1080/1062936X.2019.1686715>.
- Terzaghi, E., Alberti, E., Raspa, G., Zanardini, E., Morosini, C., Anelli, S., Armiraglio, S., Di Guardo, A., 2021. A new dataset of PCB half-lives in soil: effect of plant species and organic carbon addition on biodegradation rates in a weathered contaminated soil. *Sci. Total Environ.* 750, 141411. <https://doi.org/10.1016/j.scitotenv.2020.141411>.
- Topaz, T., Egozi, R., Eshel, G., Chefetz, B., 2018. Pesticide load dynamics during stormwater flow events in Mediterranean coastal streams: Alexander stream case study. *Sci. Total Environ.* 625, 168–177. <https://doi.org/10.1016/j.scitotenv.2017.12.213>.
- Topp, E., Metcalfe, C.D., Boxall, A.B., Lapen, D.R., 2010. Transport of PPCPs and veterinary medicines from agricultural fields following application of biosolids or manure. In: Halden, R. (Ed.), *Contaminants of Emerging Concern in the Environment: Ecological And Human Health Considerations*. American Chemical Society, pp. 227–240. <https://doi.org/10.1021/bk-2010-1048.ch010>.
- UN, 2018. SDG 6 Synthesis Report 2018 on Water And Sanitation, SDG 6 Synthesis Report 2018 on Water And Sanitation. <https://doi.org/10.18356/e8fc060b-en>.
- van der Aa, N.G.F.M., Kommer, G.J., Van Montfort, J.E., Versteegh, J.F.M., 2011. Demographic projections of future pharmaceutical consumption in the Netherlands. *Water Sci. Technol.* 63, 825–831. <https://doi.org/10.2166/wst.2011.120>.
- von Lützw, M., Kögel-Knabner, I., 2009. Temperature sensitivity of soil organic matter decomposition-what do we know? *Biol. Fertil. Soils* 46, 1–15. <https://doi.org/10.1007/s00374-009-0413-8>.
- Welch, S.A., Lane, T., Desrousseaux, A.O.S., et al., 2021. 2021 ECORISK2050: an Innovative Training Network for predicting the effects of global change on the emission, fate, effects, and risks of chemicals in aquatic ecosystems [version 1; peer review: 1 approved, 1 approved with reservations]. *Open Res. Eur.* 1, 154. <https://doi.org/10.12688/openreseurope.14283.1>.
- Werner, N., McEwen, S., Kreienbrock, L., 2018. Monitoring antimicrobial drug usage in animals: methods and applications. *Microbiol. Spectr.* 6. <https://doi.org/10.1128/microbiolspec.arba-0015-2017>.
- Westra, S., Fowler, H.J., Evans, J.P., Alexander, L.V., Berg, P., Johnson, F., Kendon, E.J., Lenderink, G., Roberts, N.M., 2014. Future changes to the intensity and frequency of short-duration extreme rainfall. *Rev. Geophys.* 52, 522–555. <https://doi.org/10.1002/2014RG000464>.
- Wickham, H., 2016. *ggplot2: Elegant Graphics for Data Analysis*. Springer-Verlag, New York.
- Willett, W., Rockström, J., Loken, B., Springmann, M., Lang, T., Vermeulen, S., Garnett, T., Tilman, D., DeClerck, F., Wood, A., Jonell, M., Clark, M., Gordon, L.J., Fanzo, J., Hawkes, C., Zuraik, R., Rivera, J.A., De Vries, W., Majele Sibanda, L., Afshin, A., Chaudhary, A., Herrero, M., Agustina, R., Branca, F., Lartey, A., Fan, S., Crona, B., Fox, E., Bignet, V., Troell, M., Lindahl, T., Singh, S., Cornell, S.E., Srinath Reddy, K., Narain, S., Nishtar, S., Murray, C.J.L., 2019. Food in the anthropocene: the EAT–Lancet commission on healthy diets from sustainable food systems. *Lancet* 393, 447–492. [https://doi.org/10.1016/S0140-6736\(18\)31788-4](https://doi.org/10.1016/S0140-6736(18)31788-4).
- Wöhrschimmel, H., MacLeod, M., Hungerbühler, K., 2013. Emissions, fate and transport of persistent organic pollutants to the arctic in a changing global climate. *Environ. Sci. Technol.* 47, 2323–2330. <https://doi.org/10.1021/es304646n>.
- Wolf, Y., Oster, S., Shuliakovich, A., Brückner, I., Dolny, R., Linnemann, V., Pinnekamp, J., Hollert, H., Schiwuy, S., 2022. Improvement of wastewater and water quality via a full-scale ozonation plant? – a comprehensive analysis of the endocrine potential using effect-based methods. *Sci. Total Environ.* 803, 149756. <https://doi.org/10.1016/j.scitotenv.2021.149756>.
- Xu, J., Wu, L., Chang, A.C., Zhang, Y., 2010. Impact of long-term reclaimed wastewater irrigation on agricultural soils: a preliminary assessment. *J. Hazard. Mater.* 183, 780–786. <https://doi.org/10.1016/j.jhazmat.2010.07.094>.
- Yang, Y.Y., Gray, J.L., Furlong, E.T., Davis, J.G., Revello, R.C., Borch, T., 2012. Steroid hormone runoff from agricultural test plots applied with municipal biosolids. *Environ. Sci. Technol.* 46, 2746–2754. <https://doi.org/10.1021/es203896t>.
- Zhu, L., Jiang, C., Panthi, S., Allard, S.M., Sapkota, A.R., Sapkota, A., 2021. Impact of high precipitation and temperature events on the distribution of emerging contaminants in surface water in the Mid-Atlantic, United States. *Sci. Total Environ.* 755, 142552. <https://doi.org/10.1016/j.scitotenv.2020.142552>.