Supporting Information

Toward Assessing Absolute Environmental Sustainability of Chemical Pollution

Marissa Kosnika, Michael Zwicky Hauschilda, Peter Fantkea*

aQuantitative Sustainability Assessment, Department of Environmental and Resource Engineering, Technical University of Denmark, 2800 Kgs. Lyngby, Denmark

Contents
S-1. Prioritization data sources and integration ................................................................. 2
S-2. Pesticide ranking ........................................................................................................... 2
S-3. Impact calculations ....................................................................................................... 4
S-4. Identifying relevant chemicals – prioritization analysis .............................................. 4
S-5. Identifying relevant chemicals – additional challenges ............................................... 5
S-6. Estonia national data .................................................................................................... 6
S-7. Estonia crop and active ingredient spatial analysis ..................................................... 6
S-8. Estonia species richness spatial analysis ...................................................................... 7
S-9. Estonia water features spatial analysis ........................................................................ 7
S-10. Estonia crop and active ingredient temporal analysis .............................................. 8
S-11. Estonia species richness temporal analysis .............................................................. 8
S-12. Estonia water features temporal analysis .................................................................. 9
S-13. Data analysis and visualization .................................................................................. 9
S-14. Spatiotemporal considerations – temporal challenges ............................................ 9
S-15. Aligning metrics and scales – Estonia aggregation challenges .................................. 10
S-16. Aligning metrics and scales – Safe Operating Space (dis-)aggregation challenges ..... 10
S-17. Data challenges ......................................................................................................... 11
References .......................................................................................................................... 12
S-1. Prioritization data sources and integration

Annual pesticide application masses for countries in Europe were collected from national statistics combined with product label information for 516 pesticide active ingredients.

Toxicity values for pesticides were determined from the species sensitivity distributions (SSDs) developed by Posthuma et al. 2019. Data were derived from the “10LogSSDMedianConcentration(ug/L)-MuChronic NOEC” column (Log(NOEC)) of the “AcuteEC50&ChronicNOEC SSD data” sheet of the supplemental data. Chemicals with NA values for the NOEC were removed, leaving median toxicity values for 7540 compounds. Values were exponentially transformed and inverted (1/(10 ^ Log(NOEC)), hereafter referred to as 1/NOEC) so larger toxicity values represent more toxic compounds.

Soil half-lives and Koc values were collected for 470 pesticides from the Pesticides Properties Database. Koc values were inverted (1/Koc) so larger Koc values represent more mobile pesticides.

Values across the three resources were linked by matching CAS registry numbers and chemical names. The final dataset had 255 pesticides with all four values available (applied mass, toxicity, soil half-life, and Koc).

S-2. Pesticide ranking

To rank pesticides by their potential to cause ecosystem damage, the set of 255 pesticides was split into four, roughly equal bins of pesticides based on the applied mass. Bins were set around the 35%, 65%, and 85% quantiles of the applied mass values, and values were rounded to the nearest 5 to create clean bins (quantiles = 55, 320, 1160 1000 kg applied pesticide). The final bins for applied mass were: (0, 55], (55-320], (320-1160], (1160-3.2e4]. From there, each of the four bins were further split into four bins based on the soil half-lives. These bins were also determined around the 35%, 65%, and 85% quantiles for soil half-lives within each of the four applied mass bins. This yielded four 35%, four 65%, and four 85% soil half-life values. The mean of these quantile values was used to determine the overall soil half-life bins, and the values were rounded to the nearest 5 to create clean bins (quantiles = 10, 30, 80 days). The final bins for soil half-lives were: (0, 10], (10-30], (30-80], (80-5500]. From there, pesticides were split into 16 bins based on the combined applied mass and soil half-life bins (e.g., bin 1 = applied mass (0, 55], soil half-life (0, 10], bin 2= applied mass (0, 55], soil half-life (10-30)). Within each bin, 1/Koc and 1/NOEC were assessed on a continuous scale. These bins are visualized in Figure 3a (main text).

To rank the pesticides with applied mass, soil half-life, 1/Koc, and 1/NOEC equally weighted as indicators of potential to cause ecosystem damage, the set of 16 bins were combined into four bins based on the combined applied mass and soil half-life quantiles. These four bins correspond to the top right four diagonals in Figure 3a (main text), and are: the top bin (applied mass = (1160-3.2e4], soil half-life = (80-5500]), the second-highest set of bins (applied mass = (1160-3.2e4], soil half-life = (30-80]; applied mass = (320-1160], soil half-life = (80-5500]), the third-highest set of bins (applied mass = (1160-3.2e4], soil half-life = (10-30]; applied mass = (320-
Within each of the four sets of bins, the combined 1/NOEC and 1/Koc values were used to rank pesticides (as determined by multiply 1/NOEC * 1/Koc). From there, the final list of prioritized pesticides was the 1/NOEC * 1/Koc ranked pesticides in the top bin, the second-highest set of bins, third-highest set of bins, and the middle set of bins. The list of ranked pesticides is in Table S1.

Table S1. Set of 255 prioritized pesticides. The top 111 pesticides are ranked

| Rank | CASRN | Rank | CASRN | Rank | CASRN | CASRN |
|------|-------|------|-------|------|-------|-------|
| 1    | 133860-33-8 | 56   | 41394-05-2 | 111  | 81591-81-3 | 82097-60-5 |
| 2    | 138261-41-3 | 57   | 999-81-5   |       | 143390-89-0 | 95617-09-7 |
| 3    | 74070-46-5 | 58   | 25606-41-1 |       | 57966-95-7 | 243973-20-8 |
| 4    | 40487-42-1 | 59   | 1071-83-6 |       | 131807-57-3 | 422556-08-9 |
| 5    | 23950-58-5 | 60   | 115-29-7  |       | 374726-62-2 | 64902-72-3 |
| 6    | 2764-72-9 | 61   | 34123-59-6 |       | 70630-17-0 | 13684-56-5 |
| 7    | 119446-68-3 | 62   | 21087-64-9 |       | 161326-34-7 | 126535-15-7 |
| 8    | 2164-17-2 | 63   | 149979-41-9 |       | 141517-21-7 | 52918-63-5 |
| 9    | 142459-58-3 | 64   | 330-55-2  |       | 88671-89-0 | 119988-49-9 |
| 10   | 94361-06-5 | 65   | 50594-66-6 |       | 149961-52-4 | 16752-77-5 |
| 11   | 131983-72-7 | 66   | 25057-89-0 |       | 76738-62-0 | 121-75-5 |
| 12   | 210880-92-5 | 67   | 272451-65-7 |       | 43121-43-3 | 135410-20-7 |
| 13   | 107534-96-3 | 68   | 19044-88-3 |       | 143390-89-0 | 119168-77-3 |
| 14   | 658066-35-4 | 69   | 116255-48-2 |       | 57966-95-7 | 203313-25-1 |
| 15   | 60207-90-1 | 70   | 1918-00-9  |       | 131807-57-3 | 78587-05-0 |
| 16   | 118134-30-8 | 71   | 106700-29-2 |       | 374726-62-2 | 3213-35-8 |
| 17   | 188425-85-6 | 72   | 67129-08-2 |       | 70630-17-0 | 134098-61-6 |
| 18   | 7287-19-6 | 73   | 2164-08-1  |       | 161326-34-7 | 96489-71-3 |
| 19   | 907204-31-3 | 74   | 79622-59-6 |       | 141517-21-7 | 74115-24-5 |
| 20   | 76674-21-0 | 75   | 60-51-5   |       | 88671-89-0 | 69327-76-0 |
| 21   | 87674-68-8 | 76   | 52888-80-9 |       | 149961-52-4 | 35367-38-5 |
| 22   | 2921-88-2 | 77   | 81777-89-1 |       | 76738-62-0 | 120928-09-8 |
| 23   | 1897-45-6 | 78   | 122-34-9  |       | 43121-43-3 | 122-14-5 |
| 24   | 500008-45-7 | 79   | 91465-08-6 |       | 126833-17-8 | 120068-37-3 |
| 25   | 330-54-1 | 80   | 8018-01-7  |       | 120116-88-3 | 35554-44-0 |
| 26   | 2032-65-7 | 81   | 1582-09-8  |       | 182675-82-3 | 5224-68-4 |
| 27   | 19666-30-9 | 82   | 874967-67-6 |       | 123-33-1 | 79538-32-2 |
| 28   | 542-75-6 | 83   | 101-21-3  |       | 127277-53-6 | 36734-19-7 |
| 29   | 51218-45-2 | 84   | 23564-05-8 |       | 1689-83-4 | 533-74-4 |
| 30   | 67306-00-7 | 85   | 41814-78-2 |       | 100646-51-3 | 57018-04-9 |
| 31   | 111479-05-1 | 86   | 163515-14-8 |       | 99129-21-2 | 156052-68-5 |
| 32   | 34256-82-1 | 87   | 95626-40-3 |       | 122931-48-0 | 114369-43-6 |
| 33   | 5915-41-3 | 88   | 125225-28-7 |       | 79277-27-3 | 110235-47-7 |
| 34   | 1698-60-8 | 89   | 53112-28-0 |       | 62476-59-9 | 10004-44-1 |
| 35   | 10605-21-7 | 90   | 61213-25-0 |       | 94125-34-5 | 81335-37-7 |
| 36   | 87392-12-9 | 91   | 220899-03-6 |       | 145701-23-1 | 128639-02-1 |
| 37   | 94-75-7 | 92   | 1861-40-1  |       | 1912-24-9 | 16118-49-3 |
| 38   | 239110-15-7 | 93   | 55219-65-3 |       | 101200-48-0 | 42576-02-3 |
| 39   | 137-26-8 | 94   | 82558-50-7 |       | 81334-34-1 | 94-81-5 |

S3
S-3. Impact calculations

Countries in Europe with applied pesticide mass provided were identified from countries listed in Europe by FAOSTAT (http://www.fao.org/faostat/en/#data). This resulted in a list of 31 countries: Austria, Belarus, Belgium, Bulgaria, Croatia, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Netherlands, North Macedonia, Norway, Poland, Portugal, Romania, Russia, Slovakia, Slovenia, Spain, Sweden, Switzerland, Ukraine, and United Kingdom.

For each of these countries, the applied mass per active ingredient for the 255 pesticides described in 0 was identified. PestLCI 2.0.8\(^\text{S}\) was used to estimate pesticide primary distribution fractions for air, field soil, and off-field emission fractions (yielding kg emitted/kg applied). To calculate impacts per country, these emission fractions were multiplied by the applied mass per country per active ingredient, the off-field area fraction (m²/m²) for agricultural soil, natural soil, and freshwater from USEtox\(^4\) based on recommendations from Gentil et al.\(^5\), and the freshwater ecotoxicity impact characterization factors, CF (PAF m³ d/kg emitted) from USEtox for these pesticides for emissions to air, agricultural soil, natural soil, and freshwater. This yielded separate impact values for emissions to air, agricultural soil, natural soil, and freshwater per country per pesticide active ingredient (e.g., Freshwater impact = Applied mass \* Emission fraction to off-field \* Off-field area fraction of freshwater \* CF for freshwater emissions). Finally, the total impact per pesticide per country was calculated by summing the emission compartment-specific impacts for each country-pesticide combination (PAF m³ d/country/year).

To determine the percentage total country impacts of prioritized pesticides (Figure 3b in the main text), the total impacts were summed for all pesticides per country. These were defined as 100% of pesticide impacts per country. From there, the percentage of these impacts for the top 50 prioritized pesticides or the bottom 144 pesticides were determined (Table S1).

S-4. Identifying relevant chemicals – prioritization analysis

We developed a list of 255 pesticides applied across countries in Europe (Table S1), and derived the total applied mass (representing chemical use), soil half-life (as a proxy for chemical
persistence), Koc (as an indicator for chemical mobility and exposure in the aquatic environment), and the median of species sensitivity distribution (SSD) no-observed effect concentrations, NOEC (as an indicator for chemical toxicity, referred to as HC50(NOEC)). By combining these attributes, the 255 pesticides were ranked based on their potential to cause adverse effects in aquatic ecosystems (Figure 3a, main text).

Through this prioritization, the most persistent and frequently used pesticides with both the highest exposure and toxicity potential were identified. In the final list, only pesticides in the ten bins closer to the top right of Figure 3a in the main text were considered. This resulted in a list of 111 prioritized pesticides. The top-priority pesticides are the fungicide azoxystrobin, insecticide imidacloprid, and the herbicides aclonifen, pendimethalin, and propyzamide. In contrast, 144 pesticides fell into the six bins closer to the bottom left of Figure 3a in the main text. These pesticides have shorter half-lives and are used in lower quantities than the 111 prioritized pesticides. Therefore, even if these pesticides have high toxicity and exposure potential, they are less likely to be in the environment long enough or in a large enough quantity to lead to substantial adverse effects.

To test the prioritized pesticide list’s ranking of pesticides by their potential for ecosystem damage, we determined the cumulative chemical impact for 31 countries in Europe based on where each of the 255 pesticides are used. Through this, we assessed if the top prioritized pesticides were representative of the chemical pressure from all pesticides in the priority list. First, we determined the cumulative impacts for all 255 pesticides considered. Then, we determined the percentage of the cumulative impacts covered by subsets of the pesticide list (Figure 3b, main text). From the list, the bottom 144 pesticides only covered 4% of the total cumulative impacts across Europe. In contrast, the top 50 pesticides covered 69% (range of 25% in Denmark – 92% in Bulgaria) of the total impacts across Europe. This demonstrates that the top prioritized pesticides explained the majority of the total impacts. This could help reduce data needs in chemical pressure quantification for instances where data on all chemicals are not available (i.e. only data for the prioritized chemicals may be necessary for analysis).

S-5. Identifying relevant chemicals – additional challenges

While we used a set of individually prioritized chemicals, an additional challenge in chemical selection is determining chemical interactions. This applies for both chemical pressure quantification, and carrying capacity calculations as both depend on quantifying the cumulative impact of compounds released in ecosystems. The effect of chemical mixtures is a common question when assessing toxicological effects on organisms, yet quantifying chemical impacts introduces an additional challenge: not only do the chemical interactions themselves need to be determined (i.e. do chemicals act additively, synergistically, or antagonistically on a target), but there needs to be a method to combine the species responses to these chemical mixtures as well. For example, within a geographic region, a set of chemicals may act additively to induce endocrine disruption in one species, but act synergistically to impede reproduction in another species, and both of these effects need to be considered in the combined chemical pressure. As of now, approaches to address chemical mixtures in impact assessment are limited. Methods like
the mixture toxic pressure calculation measure cumulative impacts, but this depends on existing knowledge of chemical mechanisms of action or the most sensitive species endpoints. For our cumulative assessment, individual chemical impacts were summed as a first proxy, in line with concentration addition-based analyses. In addition to analyzing individual chemicals and utilizing mechanism of action information, the number of chemicals under study could be reduced by selecting indicator chemicals to represent a chemical class (e.g., abamectin could be an indicator for avermectin pesticides), or chemicals can be assessed based on representative chemical structures.

An under-considered challenge in describing chemical effects on an ecosystem is the role of metabolism. Both biotic and abiotic transformation processes are crucial to determine how chemicals (especially pesticides) are removed from the environment or may result in more hazardous transformation products. Maggi et al. 2020 conducted a global analysis on contamination of glyphosate and its main metabolite AMPA and found that, while glyphosate is non-persistent, AMPA may persist up to 20 years in top soil. To identify pesticide metabolites, the Pesticide Properties Database provides estimated maximum metabolite occurrence fractions in soil for several pesticides to indicate whether a metabolite may be relevant. Other sources like BioTransformer predict environmental microbial metabolites from input chemical SMILES codes. As metabolic products become better characterized (e.g., more information on their physicochemical properties become available), methods can be developed to weight the relative contribution of the original chemical and transformation products (e.g., based on the maximum occurrence fractions) to better consider the role of chemical metabolism in chemical pressure and carrying capacity quantification.

S-6. Estonia national data

A shape file for Estonia with corresponding latitude/longitudes for country and county borders was collected from level 1 GADM data using R/raster (version 3.4-5). These county/country-level boundaries were used to set restrictions for all data (e.g., species county locations were determined from the GADM county borders).

S-7. Estonia crop and active ingredient spatial analysis

Tables KK2081: Quantity of pesticides used and the basic area treated in agricultural holdings by active substance and crop (last updated 29.12.2020) and KK2082: Use of pesticides in agricultural holdings by county and crop (last updated 08.12.2016) were downloaded for the year 2015 from Statistics Estonia. Table KK2081 provided the kg of active substance applied and basic hectares treated per specific crop by each active substance. Entries with the kg of active substance applied or the basic hectares treated per crop entered as “0” were removed. Table KK2082 provided the total kg (not per active ingredient) of insecticides, fungicides, herbicides, and other pesticides (“others”) applied per broad crop class (e.g., “Cereals”) per county in Estonia.

To develop a dataset with the amount of pesticide applied per crop per county in Estonia, the application rate (kg/ha) for each active ingredient-specific crop entry in Table KK2081 was
calculated. Then, because Table KK2082 is at the broad pesticide-crop class level, the maximum application rate determined for Table KK2081 for the same pesticide-crop class level was collected (e.g., the specific pesticide-crop combination with the greatest application rate among all insecticides applied to the broad cereal class was the application rate of 0.3333 kg/ha for alpha-cypermethrin on rye, so this was used as the application rate for insecticides to cereals).

Then, for each Table KK2082 entry of kg applied per crop per county, the maximum application rate for each pesticide class was used to determine the hectares of crop treated per county (e.g., Harju county had 1561.615 kg of insecticides applied to cereals, so based on the 0.3333 kg/ha application rate for insecticides on cereals, 4684.167 ha were treated). Finally, the hectares treated per county were normalized to the total county area to give the percent of hectares of crop treated by each pesticide class per county. These results are plotted in Figure 4a (main text).

**S-8. Estonia species richness spatial analysis**

Animalia\(^{12}\) and Plantae\(^{13}\) observation data for Estonia were downloaded from the Global Biodiversity Information Facility. Data were provided as observations with the date of the observation, the location of the observation (latitude, longitude), and the species observed. Entries were restricted to the year 2015, and observations with no latitude, longitude, month, or species name provided were removed, or with observations outside of the Estonia GADM country shape file latitude/longitude borders. Plants were set as all species in the kingdom “Plantae”, insects were all species in the class “Insecta”, and birds were set as all species in the class “Aves”. This resulted in 203,727 plant observation entries, 18,533 insect observation entries, and 125,728 bird entries.

To approximate species richness across Estonia, latitude and longitude decimal degrees entries were restricted to the hundredths decimal place (most entries were provided with 5 decimal places). Then, observations were aggregated across these truncated latitude/longitude combinations to determine the number of unique species observed in each area. These results are plotted in Figure 4b (main text).

**S-9. Estonia water features spatial analysis**

The location of lakes (key = ‘water’, value = ‘lake’) and rivers (key = ‘waterway’, value = ‘river’) in Estonia were identified using the R/osmdata (version 0.1.5).\(^{14}\) These features were restricted to those within the bounds of the GADM Estonia shape file.

Hydrological data for Estonia was collected from the Estonian Weather Service ([https://www.ilmateenistus.ee/ilmateenistus/vaatlusvork/#hydro](https://www.ilmateenistus.ee/ilmateenistus/vaatlusvork/#hydro)). The location (latitude and longitude) for each hydrometric station is reported on the respective webpage, along with the catchment area and height of the zero measurement (i.e., the height of the gauge where the water level is measured from) for water level gauge datums. These values were collected for each of the 56 hydrometric stations in Estonia. The location of Estonian water bodies and three
hydrometric stations are shown in the first column of Figure 4c (main text), and the catchment area for all hydrometric stations is in the middle column of Figure 4c (main text).

Lake and river water volumes were extracted from HydroATLAS Version 1.0. The level 10 BasinATLAS shape file was overlaid with the Estonia county shape file, and the intersecting attribute table was extracted using ArcGIS (Desktop version 10.8). This provided grids of lake volumes (lkv_mc_usu column) and river volumes (riv_tc_ssu column) within each county of Estonia. Then, the sum of the lake and river volumes within each county was taken to provide the total water volume per county. This was divided by the total county area in hectares to provide the average water volume/ha for each county. This is shown in the last column of Figure 4c (main text).

S-10. Estonia crop and active ingredient temporal analysis

Three insecticide-crop combinations were analysed for differences in when specific active ingredients are applied to each crop. Based on the insecticides applied to each crop as reported by Statistics Estonia (Estonia crop and active ingredient spatial analysis), the corresponding crop stage of application for each active ingredient was determined.

Because crop stages on product labels were separated between spring and winter cereals, these were separated to determine BBCH crop stages. 9 insecticide active ingredients were applied to spring cereals, 11 were applied to winter cereals, 8 were applied to legumes, and 7 were applied to potatoes. For each active ingredient, the Register of Plant Protection Products authorised in Estonia was searched (https://portaal.agri.ee/avalik/#/taimekaitse/taimekaitsevahendid-otsing/en) to determine the product used that contains each active ingredient. In some instances, the BBCH crop stage for application of each active ingredient to each crop was provided on product labels available from this resource. Where possible, this was used to assign application start and end times to each active ingredient-crop combination (e.g., insecticide tau-fluvinate in the product Evure is listed with application times from BBCH 10 to 79 for legumes). When this was not provided, BBCH’s were determined by first searching for product labels (e.g., the Karate Zeon 5 CS, active ingredient lambda-cyhalothrin, lists application from BBCH 9 to 60 for spring cereals), then specific product developer websites (e.g., BASF lists Fastac 50, active ingredient alpha-cypermethrin, application from BBCH 10 to 89 for potatoes), then FAO reports for specific active ingredients.

From here, the number of pesticides that could be applied for each BBCH for each crop were summed (merging spring and winter cereals). These data are plotted in Figure 4d (main text).

S-11. Estonia species richness temporal analysis

Each observation for plants, insects, and birds from GBIF (Estonia species richness spatial analysis) had the month of the observation reported. The number of unique species observed for each month was determined, and these data are shown in Figure 4e (main text).
S-12. Estonia water features temporal analysis

Hourly reports of the water level (in cm) and water temperature (in °C) are reported at each hydrologic station in Estonia (http://www.ilmateenistus.ee/siseveed/vaatlusandmed/kaart/?lang=en). These data reports were downloaded for 14:00 for the 15th of each month in the year 2015.

The water level for each hydrologic station was reported relative to the gauge datum (i.e. the water level was reported in cm above the 0 measurement point). These gauge datum values for each station were already collected (Estonia water features spatial analysis), and the water level measurement for each month was added to the gauge datum of the relevant station to provide the actual water level at that station. These monthly water levels and the monthly water temperature measured at three sample stations are provided in the first and last column of Figure 4f (main text).

Daily water flow rates are provided by the Estonian Weather Service for each station as .CSVs for a relevant reference period (https://www.ilmateenistus.ee/siseveed/ajaloolised-vaatlusandmed/vooluhulgad/). The data for three water stations (Luguse, Oore, and Nurme) were collected.

Flow rates reported for each station on the 15th of each month in the year 2015 were selected to match the same date as the reported water level and temperature. These data are shown in the middle column of Figure 4f (main text).

S-13. Data analysis and visualization

Analyses were done using R version 3.6.3.16 Figures in the main text were generated using R/ggplot2 (version 3.3.3).17

S-14. Spatiotemporal considerations – temporal challenges

Temporal differences are important in impact quantification and carrying capacity development. There are differences in when crops are treated throughout the year (Figure 4d, main text), which can impact how species in an ecosystem are affected because, depending on when pesticides are applied, this could overlap with times when species are more sensitive (e.g., when offspring are born). For Estonia, the density of species observed varied throughout the year, with plants and insects having few observations in the winter with peaks in the summer, and birds observed more consistently over the whole year (Figure 4e, main text). Water characteristics also varied over time, as shown in Figure 4f in the main text for three hydrologic stations in Estonia. These are important because—depending on the water level—the dilution capacity for a pesticide can change, while the flow rate can dictate how far a pesticide moves into a catchment. Additionally, changing water characteristics like temperature can impact pesticides’ environmental fate. Therefore, when assessing chemical fate for quantifying chemical pressure or carrying capacity determination, analyses should be conducted at a level that accounts for discriminating spatial and temporal variability.
S-15. Aligning metrics and scales – Estonia aggregation challenges

To implement an absolute environmental sustainability assessment (AESA) framework for chemical pollution, data need to be aligned so the spatiotemporal granularity matches between the quantified chemical pressure and carrying capacity. For some attributes, the calculated values can be averaged either spatially or temporally. For example, we found that the water level at each hydrologic station does not change noticeably throughout the year, with the standard deviation across the months averaging only 3% of the mean water level (Figure 4f, main text). Therefore, when aggregating these data, values can be averaged without losing discrimination power. In contrast, water volume varied spatially by a factor of 7200:1 between the greatest and smallest county water volumes in Estonia (Figure 4c, main text). Further, species richness varied substantially both spatially and temporally (Figure 4b,e, main text). Only 64 unique insect species were observed between January and March while 1836 unique species were observed between June and August. If the insect species across Estonia are just averaged for the whole year, the average number of unique species observed is 101, which leads to different impact and carrying capacity quantifications than would be determined for 1836 species. However, one challenge with these data is that insects may not be directly observed throughout the entire year, but could be present at other life stages where they are sensitive (e.g., as eggs or larvae). An alternative approach to averaging values upon aggregation would be to weight the calculated chemical impacts or carrying capacities according to the relative contributions of the underlying data. For example, to quantify the calculated mixture toxic pressures over environmental compartments, Zjip et al. 2014\textsuperscript{6} weighted the calculated chemical impacts by the available compartment volumes to develop volume-weighted msPAF. In the same way, species richness could be used to weight the PAF in each geographic region and over time. Therefore, by applying correction factors where needed to account for spatiotemporal variability in both the chemical impacts and the carrying capacity, regional and temporal differences can still be accounted for in the final chemical pollution assessment calculations.

S-16. Aligning metrics and scales – Safe Operating Space (dis-)aggregation challenges

A derived safe space for chemicals needs to be both disaggregated to relevant actors and, if necessary, re-aggregated to the relevant organizational level to match the chemical pressure assessment. Allocating the Safe Operating Space (SOS) to different levels of stakeholders requires value choices, both to define the total SOS and determine the appropriate actor(s) receiving a portion of the SOS. How the space is assigned greatly influences whether an activity is found to be absolutely sustainable. For example, Ryberg et al. 2018\textsuperscript{18} used LCIA methodology to conduct a case study on laundry washing in the EU against planetary boundaries (not including chemical pollution), and used four different allocation principles. They found uncertainty from the choice of allocation principle was more influential on the outcome than the other decision points (e.g., life cycle inventory uncertainty). Therefore, allocation principles and methods for downscaling need to be harmonized to create a systematic and informed approach for AESA applications. Hjalsted et al. 2020\textsuperscript{19} propose allocating the SOS among relevant stakeholders following a two-step approach, where the SOS is first downscaled to the individual
level and then upscaled to the relevant, higher organizational level (e.g., company, sector, country, etc.). Therefore, if the aggregation level for chemical pressure and carrying capacity quantification was at the country level, the equal per capita egalitarian approach could be used to determine the SOS per (average) individual, and then upscaled based on the population of the country. Through this joint downscaling and upscaling approach, it is also possible to allocate to a company or sector by using the final consumption expenditure approach with the population of customers, and upscaling based on the spending patterns of individuals.

S-17. Data challenges

When developing emissions inventories, data are often available for only a few chemicals in a few regions, emissions may only be reported in aggregate (e.g., reporting chemical class emissions rather than specific chemicals), or the reported chemicals may be missing corresponding physicochemical properties and/or toxicity data.\textsuperscript{20,21} We detail the challenges underlying the development of an inventory of pesticide application data for Europe using an illustration of two countries with different types of pesticide data available:

To quantify the chemical pressure resulting from pesticide use in Europe, the first step is to develop an inventory of pesticide application data. For this, the data required are the active ingredient applied, the crop treated, the mass of active ingredient applied per crop, the growth stage of the crop at treatment, the time of year at crop treatment, the hectares of farmland treated, and the location (preferably at a detailed, regional level) of the farmland. No single source contains all these data. A possible start to acquire these data is to analyze individual country's reports on pesticides (see Table 1, main text), but data reporting across countries differs greatly. For example, Statistics Estonia provides data on the kilograms of active ingredient applied per crop and hectares treated as a single downloadable XLS file (available in English). However, these data are not reported per region, requiring integration with an additional dataset of crop-county data to determine where in Estonia pesticides are applied, which introduces assumptions about pesticide use across crops in different regions. Further, this dataset does not describe the growth stage of crop treatment or the time of year of treatment. Therefore, utilizing this dataset requires predicting the pesticide products applied that contain each reported active ingredient (e.g., predict Fastac 50 was used in instances where cereals were reported to be treated with alpha-cypermethrin), determining the typical crop growth stage of pesticide application for this product from the product label (typically available as a PDF from the manufacturer), and consulting farmer’s handbooks to determine when in the year crops are planted in Estonia. In contrast, the Bulgarian Food Safety Agency doesn't provide explicit pesticide application data, instead providing hundreds of pages of PDFs (available in Bulgarian) describing the different products, active ingredient composition, and a range of doses to be applied to crops in Bulgaria. While this data does not report active ingredient use per region in Bulgaria nor the hectares treated, this dataset does provide detailed information of the crop stage of treatment for each product. Therefore, utilizing this dataset requires extraction of the available active ingredient-dose-crop combinations from the PDFs, and prediction of the actual amount of pesticide applied based on the reported doses and reports of harvested crops in different regions of Bulgaria for a
In both datasets, information about application method is entirely missing, requiring further assumptions as application methods drive differences in emission patterns.

On top of these challenges, determining relevant species’ effects following chemical exposure is a challenge due to the diversity of species across ecosystems. While there is some data describing chemical effects on organisms, these data are often generated in a lab-based setting on a few test species. Despite being found in the tropics, in vivo zebrafish (Danio rerio) toxicity values are used to characterize chemical threats in ecosystems for European risk assessment. Further, the chemical toxicity endpoints tested are often acute, or non-specific, such as general 50% effect concentrations. Therefore, assessing the broad range of endpoints (e.g., developmental toxicity, endocrine disruption, lethality) that can affect the different species in a given ecosystem is a challenge. Spatiotemporally differentiated global and regional data on other features of ecosystems (water characteristics, species distributions, land use, etc.) are also scarce, but crucial in both chemical impact quantification and carrying capacity determinations. Finally, the SOS development requires data to determine the background load on an ecosystem from other chemical sources (e.g., water-borne discharges from industry, atmospheric deposition), non-toxic pollutants, and other man-made stressors that can occupy the SOS. Additionally, depending on how the allocation is performed, human activity, economic, or population data, is needed to allocate the SOS between different activities that contribute to chemical pollution pressure on the exposed ecosystem.

References

(1) Posthuma, L.; van Gils, J.; Zijp, M. C.; van de Meent, D.; de Zwartd, D. Species Sensitivity Distributions for Use in Environmental Protection, Assessment, and Management of Aquatic Ecosystems for 12 386 Chemicals. Environ. Toxicol. Chem. 2019, 38 (4), 703–711. https://doi.org/10.1002/etc.4373.

(2) Lewis, K. A.; Tzilivakis, J.; Warner, D. J.; Green, A. An International Database for Pesticide Risk Assessments and Management. Hum. Ecol. Risk Assess. An Int. J. 2016, 22 (4), 1050–1064. https://doi.org/10.1080/10807039.2015.1133242.

(3) Dijkman, T. J.; Birkved, M.; Hauschild, M. Z. PestLCI 2.0: A Second Generation Model for Estimating Emissions of Pesticides from Arable Land in LCA. Int. J. Life Cycle Assess. 2012, 17 (8), 973–986. https://doi.org/10.1007/s11367-012-0439-2.

(4) Rosenbaum, R. K.; Bachmann, T. M.; Gold, L. S.; Huijbregts, M. A. J.; Jolliet, O.; Jurase, R.; Koehler, A.; Larsen, H. F.; MacLeod, M.; Margni, M.; McKone, T. E.; Payet, J.; Schuhmacher, M.; Van De Meent, D.; Hauschild, M. Z. USEtox - The UNEP-SETAC Toxicity Model: Recommended Characterisation Factors for Human Toxicity and Freshwater Ecotoxicity in Life Cycle Impact Assessment. Int. J. Life Cycle Assess. 2008, 13 (7), 532–546. https://doi.org/10.1007/s11367-008-0038-4.

(5) Gentil, C.; Basset-Mens, C.; Manteaux, S.; Mottes, C.; Maillard, E.; Biard, Y.; Fantke, P. Coupling Pesticide Emission and Toxicity Characterization Models for LCA: Application to Open-Field Tomato Production in Martinique. J. Clean. Prod. 2020, 277, 124099. https://doi.org/10.1016/j.jclepro.2020.124099.

(6) Zijp, M. C.; Posthuma, L.; Van De Meent, D. Definition and Applications of a Versatile Chemical Pollution Footprint Methodology. Environ. Sci. Technol. 2014, 48 (18), 10588–
(7) Posthuma, L.; Zijp, M. C.; De Zwart, D.; Van de Meent, D.; Globevnik, L.; Koprivsek, M.; Focks, A.; Van Gils, J.; Birk, S. Chemical Pollution Imposes Limitations to the Ecological Status of European Surface Waters. *Sci. Rep.* **2020**, *10* (1), 1–12. https://doi.org/10.1038/s41598-020-71537-2.

(8) Bjørn, A.; Diamond, M.; Birkved, M.; Hauschild, M. Z. Chemical Footprint Method for Improved Communication of Freshwater Ecotoxicity Impacts in the Context of Ecological Limits. *Environ. Sci. Technol.* **2014**, *48* (22), 13253–13262. https://doi.org/10.1021/es503797d.

(9) Maggi, F.; la Cecilia, D.; Tang, F. H. M.; McBratney, A. The Global Environmental Hazard of Glyphosate Use. *Sci. Total Environ.* **2020**, *717*, 137167. https://doi.org/10.1016/j.scitotenv.2020.137167.

(10) Djoumbou-Feunang, Y.; Fiamoncini, J.; Gil-de-la-Fuente, A.; Greiner, R.; Manach, C.; Wishart, D. S. BioTransformer: A Comprehensive Computational Tool for Small Molecule Metabolism Prediction and Metabolite Identification. *J. Cheminform.* **2019**, *11* (1), 1–25. https://doi.org/10.1186/s13321-018-0324-5.

(11) Hijmans, R. J. Raster: Geographic Data Analysis and Modeling. R Package Version 3.4-5. Https://CRAN.R-Project.Org/Package=raster. **2020**.

(12) GBIF.org. (13 April 2021) GBIF Occurrence Download. https://doi.org/10.15468/dl.e243g4.

(13) GBIF.org. (13 April 2021) GBIF Occurrence Download. https://doi.org/10.15468/dl.ysg9zk.

(14) Padgham, M.; Lovelace, R.; Salmon, M.; Rudis, B. Osmsdata. *J. Open Source Softw.* **2017**, *2* (14), 305. https://doi.org/10.21105/joss.00305.

(15) Linke, S.; Lehner, B.; Ouellet Dallaire, C.; Ariwi, J.; Grill, G.; Anand, M.; Beames, P.; Burchard-Levine, V.; Maxwell, S.; Moidu, H.; Tan, F.; Thieme, M. Global Hydro-Environmental Sub-Basin and River Reach Characteristics at High Spatial Resolution. *Sci. Data* **2019**, *6* (1), 283. https://doi.org/10.1038/s41597-019-0300-6.

(16) R Core Team. R: A Language and Environment for Statistical Computing. **2019**.

(17) Wickham, H. Ggplot2: Elegant Graphics for Data Analysis. Springer-Verlag New York. **2016**.

(18) Ryberg, M. W.; Owsianiak, M.; Clavreul, J.; Mueller, C.; Sim, S.; King, H.; Hauschild, M. Z. How to Bring Absolute Sustainability into Decision-Making: An Industry Case Study Using a Planetary Boundary-Based Methodology. *Sci. Total Environ.* **2018**, *634*, 1406–1416. https://doi.org/10.1016/j.scitotenv.2018.04.075.

(19) Hjalsted, A. W.; Laurent, A.; Andersen, M. M.; Olsen, K. H.; Ryberg, M.; Hauschild, M. Sharing the Safe Operating Space: Exploring Ethical Allocation Principles to Operationalize the Planetary Boundaries and Assess Absolute Sustainability at Individual and Industrial Sector Levels. *J. Ind. Ecol.* **2020**, *1–14*. https://doi.org/10.1111/jiec.13050.

(20) Cucurachi, S.; Sala, S.; Laurent, A.; Heijungs, R. Building and Characterizing Regional and Global Emission Inventories of Toxic Pollutants. *Environ. Sci. Technol.* **2014**, *48* (10), 5674–5682. https://doi.org/10.1021/es405798x.

(21) Leclerc, A.; Laurent, A. Framework for Estimating Toxic Releases from the Application of Manure on Agricultural Soil: National Release Inventories for Heavy Metals in 2000–2014. *Sci. Total Environ.* **2017**, *590–591*, 452–460. https://doi.org/10.1016/j.scitotenv.2017.01.117.
(22) European Chemicals Agency. Guidance on Information Requirements and Chemical Safety Assessment Chapter R.7b: Endpoint Specific Guidance. 2017, Version 4.

(23) Aurisano, N.; Albizzati, P. F.; Hauschild, M.; Fantke, P. Extrapolation Factors for Characterizing Freshwater Ecotoxicity Effects. *Environ. Toxicol. Chem.* **2019**, *38* (11), 2568–2582. https://doi.org/10.1002/etc.4564.