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Article

Spatialized Life Cycle Assessment of Fluid Milk Production and Consumption in the United States

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1 Regions and water basins

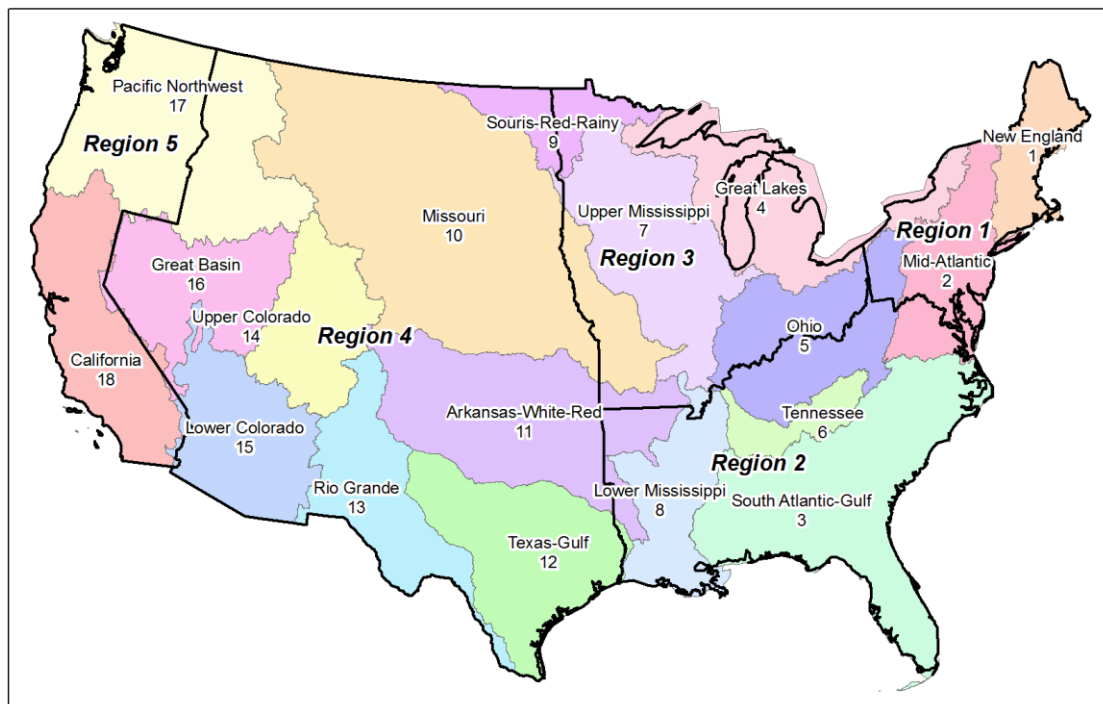


Figure S1. Location of milk producing regions (dark outlines) and USGS Hydrological units HUC-2 [29] watersheds.

Table S1. HUC-2 watersheds.

HUC Num	HUC2 Abbr	HUC2 Name
1	N.Eng.	New England
2	Mid-Atl.	Mid-Atlantic
3	S. Atl-Gulf	South Atlantic-Gulf
4	Gr.Lakes	Great Lakes
5	Ohio	Ohio
6	Tenn.	Tennessee
7	Up.Miss.	Upper Mississippi
8	Low.Miss	Lower Mississippi
9	Sour-R-R	Souris-Red-Rainy

HUC Num	HUC2 Abbr	HUC2 Name
10	Missour.	Missouri
11	Ark-W-R	Arkansas-White-Red
12	Tx-Gulf	Texas-Gulf
13	Rio Grnd.	Rio Grande
14	Up.Colo.	Upper Colorado
15	Low.Colo.	Lower Colorado
16	Gr.Basin	Great Basin
17	Pac-NW	Pacific Northwest
18	Calif.	California
Other	Other	Other

2 Inventory data

Table S2. Data description and sources for feed production data.

Data Type	Description	Matrix	Source	Data Available for Feeds	Geographic Coverage	Time Coverage
Production	total bushels produced	FT_{feed} (modeled)	Quickstats [61].	alfalfa hay / silage, corn grain / silage, & soy-bean	state	2007
Crop transport	supply states for crop consumption in each state	FT_{feed} (data)	Corn / soy studies [62,63] (see supporting information)	Corn, soy	state	1985
Area harvested	acres harvested	B_{feed}	Quickstats	alfalfa hay / silage, corn grain / silage, & soy-bean	state	2007
Yield	crop production / area	B_{feed}	calculated from production and area	alfalfa hay / silage, corn grain / silage, & soy-bean	state	2007
Irrigation rate	application rate (volume / area)	B_{feed}	Farm Ranch Irrigation Survey (FRIS) [64]	alfalfa hay / silage, corn grain / silage, & soy-bean	state	1998, 2003, and 2008
Irrigated area harvested	total irrigated acres	B_{feed} (water)		alfalfa hay / silage, corn grain / silage, & soy-bean	state	2007
Water source: surface vs. Groundwater	fraction water withdrawal from surface and groundwater	B_{feed} (water)	USGS National water use estimates [55]	all	state	2005
Nutrient application and loss	synthetic and manure-based nitrogen and phosphorus fertilizer (mass loss / area)	B_{feed}	National Nutrient Loss and Soil Carbon Database (NNLSC) [33].	corn grain / silage, legume hay, grass hay, soybean	field-level, aggregated to state	1997
Pesticide application	application rate (mass / area)	B_{feed}	Quickstats	alfalfa hay / silage, corn grain /	national	2009

Data type	Description	Matrix	Source	Data available for feeds	Geographic coverage	Time coverage
				silage, & soy-bean		

Table S3. Data available for on-farm dairy activities.

Data Type	Description	Matrix	Source	Geographic Coverage	Time Coverage
Animal rations	kg DM/kg FPCM	R_{feed}	Milk GHG [65].	regional	2009
Milk cow inventory	total cow population	B_{farm}	Quickstats [61].	state	2007
Milk production	total milk production	B_{feed}	Quickstats	state	2007
Milk production (lb/head)	milk production per head	$P_{milk}^{national}$	calculated from production and population	state	2007
Average herd demographics (including replacement animals)	herd demographic with replacement animals	B_{farm} R_{feed}	Milk LCA [23]	regional	2009
Manure management systems (MMS)	manure handling across 18 MMS or pasture	B_{farm}	Milk LCA [23]	regional	2009
Milk/beef allocation	fraction of flows or impacts attributed to dairy production	I_{milk}	Milk LCA [23]	regional	2009-2010
Washing water	washing, milk cooling, etc.	B_{farm}	Rotz, 2010, Personal communication	national	not applicable
Drinking water	drinking water for animals	B_{farm}	National Research Council [66].	national	not applicable
Land use	average land use / head	B_{farm} B_{feed}	Integrated Farm System Model (IFSM) [67]	national	not applicable
Nitrogen content of manure	kg N / head-day	B_{farm}	[68], Milk LCA [23]	not applicable	not applicable
Nitrogen losses from MMS	kg N / head-day	B_{farm}	[69,70]	[69]: not applicable; [70]: state	not applicable
Pesticide application	kg substance / head-day	B_{feed}	Quickstats	national	not applicable

3 Rations

Table S4. Main components of dairy feed in the U.S. (kg dry matter feed / kg unallocated FPCM). Regions defined according to [9] as shown in Figure S1.

Crop	Region				
	1	2	3	4	5
alfalfa hay	5.2E-2	2.4E-2	4.1E-2	2.1E-1	1.8E-1

Crop	Region				
	1	2	3	4	5
alfalfa silage	1.8E-1	3.3E-2	1.7E-1	1.1E-1	2.6E-2
corn grain	1.3E-1	1.4E-1	1.6E-1	1.4E-1	1.3E-1
corn silage	3.6E-1	1.2E-1	3.0E-1	1.9E-1	1.9E-1
DDGS dry	2.3E-2	8.4E-2	4.7E-2	6.0E-2	7.5E-2
DDGS wet	7.7E-3	0.0E+0	1.9E-2	1.8E-2	1.3E-2
grass hay	2.3E-2	1.1E-1	5.5E-2	5.9E-2	4.8E-2
grass pasture	2.7E-2	1.8E-1	1.5E-2	1.6E-2	1.0E-2
grass silage	2.6E-2	6.7E-2	1.2E-2	5.4E-2	6.5E-2
soybean	6.1E-3	3.0E-3	1.3E-2	1.1E-3	0.0E+0
soybean meal	9.3E-2	8.6E-2	8.8E-2	4.7E-2	6.2E-2
feed mix	1.5E-1	2.9E-1	9.7E-2	2.0E-1	2.4E-1

4 Feed production and transport

In order to spatially model impacts related to feed production, it is essential to identify the provenance of each given state's milk rations. Some crops (corn grain and soybean) are traded and shipped on a national scale. For these crops, thorough studies of corn and soybean movements were conducted and published at the end of the 1980s (Fruin et al. 1989; Larson et al. 1990), but have not been recently updated. These studies provide insight into the movement of these grains by train, barge, and truck, as they indicate where crops are shipped for processing. Although production of these crops has changed in the intervening years, a comparison of 1985 and 2007 corn grain and soybean production levels from USDA indicates that, with the exception of a few outliers, production has increased proportionally across states. Given the limited data availability, and given that the model required fractional, not absolute, supply information by state, it was assumed that past shipment information could be used to describe current inter-state transfer as presented in Figures S2 and S3. Processed ration components were also treated as national commodities: distillers dry and wet grains were modeled to be shipped as corn, soybean meal as soybean, and feed mix as a combination of corn and soybeans.

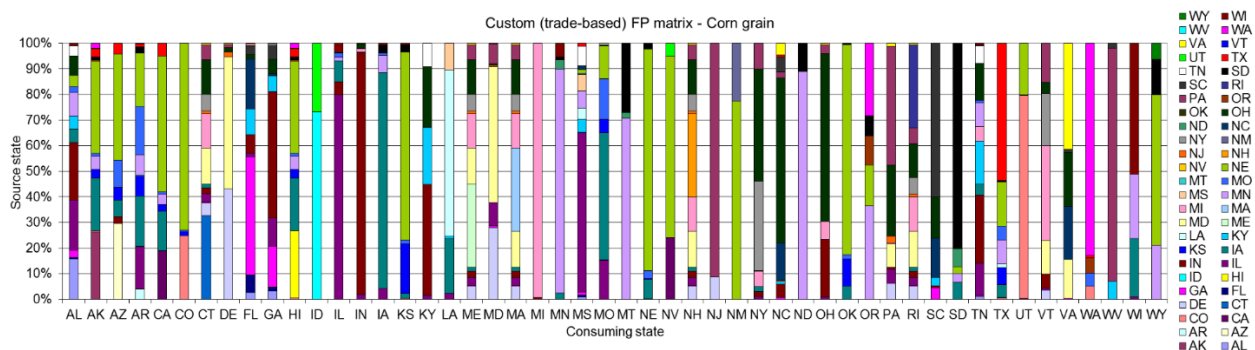


Figure S2. Trade-based FP matrix for corn grain, showing supplying states for each state.

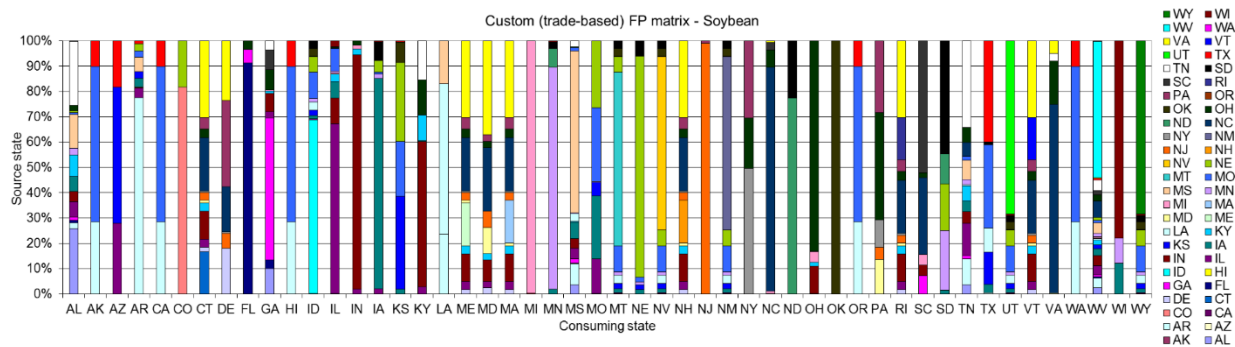


Figure S3. Trade-based FP matrix for soybean, showing supplying states for each state.

For alfalfa hay and grass hay, it is assumed that these feed components are produced within a given state's region, proportional to each state's production of that crop. For those components of the rations that are either high-moisture and heavy, and thus not easily transported (corn silage and alfalfa silage) or not transportable (pasture), it is assumed that the entire crop needed for a given state's milk production is produced in that state.

5 Uncertainty

The current state of practice in life cycle assessment includes a quantitative analysis of the uncertainty in inventory data. In this study, much of the background process data, which is part of the ecoinvent database, includes such uncertainty analyses. Possible underestimations of uncertainty associated with ecoinvent are known [71]; however, ecoinvent and agricultural *inventory* uncertainties are expected to be lower overall than *impact* uncertainty.

The base life cycle model constructed by Thoma et al. [65] and implemented in Simapro includes all stages of the milk life cycle: feed production, milk production, processing, packaging, transport, retail, and consumption. That model created new unit processes to reflect the regional nature of the study, and also includes estimates of data uncertainty that we used for upstream and post farm gate processes. For feed and dairy farm processes, raw data provided by the USDA does not consistently carry such estimates of uncertainty. Therefore we estimated the uncertainties of feed and dairy processes following the approach of ecoinvent [72] in order to ensure consistency with the rest of the life cycle model, which mostly relies on ecoinvent data.

Uncertainty is expressed as a GSD_{95^2} (a squared geometric standard deviation, assuming a lognormal distribution), which indicates that 95% of the data fall within the mean multiplied by the GSD_{95^2} and mean divided by the GSD_{95^2} . For brevity, we use GSD^2 to indicate the GSD_{95^2} . The basic uncertainty depends on the kind of inventory flows considered [72]; these were estimated based on ecoinvent guidelines and expert judgment (Basic GSD^2 column of Table S6.). For example, ecoinvent basic uncertainty for NO_x and N_2O emissions from agriculture is 1.40; this study has conservatively used 1.5 for manure management emissions. Secondly, the components of the data pedigree were estimated. The ecoinvent data pedigree is a qualitative score from 1-5, across six categories: reliability, completeness, temporal correlation, geographical correlation, further technological correlation, and sample size. Table S5 shows the relationship between pedigree and uncertainty. The selected pedigree is reported in Table S6. as a number sequence, separated by commas, matching the column order of Table S5.

Table S5. Relationship between data pedigree (left column) and GSD² [72]. Dashes indicate data not supplied byecoin-vent.

Pedi- gree	Reliabil- ity U_R	Complete- ness U_C	Temporal Correlation U_T	Geographical Correlation U_G	Technological Correlation U_L	Sample Size U.S.
1	1	1	1	1	1	1
2	1.05	1.02	1.03	1.01	-	1.02
3	1.1	1.05	1.1	1.02	1.2	1.05
4	1.2	1.1	1.2	-	1.5	1.1
5	1.5	1.2	1.5	1.1	2	1.2

Inventory components, their data sources, basic GSD², qualitative pedigree, an overall GSD² estimates are shown in Table S6. For most entries in the table below, the basic GSD² and pedigree were estimated separately. In some entries – those drawn from models or national level data – an overall GSD² was estimated directly, using expert judgment.

Table S6. Summary of uncertainty and pedigree for input data.

Inventory Data (or Modeling Assumption)	Source	Basic U_b	Pedigree	Overall GSD ²
Milk production	[61]	1.05	1,1,1,1,1,1	1.05
Cow population	[61]	1.02	1,1,1,1,1,1	1.02
Rations	GHG study [65]	1.05	2,3,1,1,1,1	1.09
Crop transport model (water)	this study	overall	overall	1.20
Crop transport model (eutroph.)	this study	overall	overall	1.10
Crop transport model (others)	this study	overall	overall	1.05
Crop allocation (e.g., soybean meal)	this study	overall	overall	1.05
Crop surface harvested	[61]	1.05	1,1,1,1,1,1	1.05
Crop production	[61]	1.05	1,1,1,1,1,1	1.05
Producer land use	IFSM [67]	1.5	2,2,2,2,3,3	1.57
Crop irrigation (m ³ /m ²)	FRIS [64]	1.01	2,2,2,1,1,1	1.06
Water source (surface vs. ground)	USGS [55]	overall	overall	1.10
Irrigation evaporation (assumed negligible)	this study	overall	overall	1.30
Producer wash water	NMSU [73]	overall	overall	1.50
Producer cow drinking water	Al Rotz, personal communication	1.2	2,2,2,2,3,3	1.31
Crop pesticide application	[61]	overall	overall	1.50
Milk producer pesticide application	[61]	overall	overall	1.50
MMS nutrient emissions	[69]	1.5	3,3,4,3,3,3	1.64
Crop nutrient emissions	[33,34]	1.3	2,1,4,1,1,1	1.38

Having estimated uncertainties for each component of the inventory, we then calculate an overall estimate for uncertainty associated with each spatially modeled category using equation S1.

The combined basic uncertainty and data pedigree were used to create an overall inventory uncertainty for each input set of data, using equation S1, which combines uncertainties for multiplicative factors.

$$GSD_O^2 = \exp^{\sqrt{(\ln U_b)^2 + (\ln U_R)^2 + (\ln U_C)^2 + (\ln U_T)^2 + (\ln U_G)^2 + (\ln U_L)^2 + (\ln U_S)^2}} \quad (S1)$$

Table S7 presents the combined GSD^2 combining all the feed production and milk producer components of each of the four spatially modeled categories. Applying a Monte-Carlo analysis to each of the inventory flows, figure S4 presents the resulting inventory uncertainties per kg FPCM associated with each midpoint impact category.

Table S7. Combined uncertainty for spatially modeled inventory components.

Modeled Inventory Flows	GSD ²
land (feed)	1.15
land (producer)	1.59
water (feed)	1.44
water (producer)	1.64
toxicity (feed)	1.54
toxicity (producer)	1.52
eutroph. (feed)	1.44
eutroph. (producer)	1.66

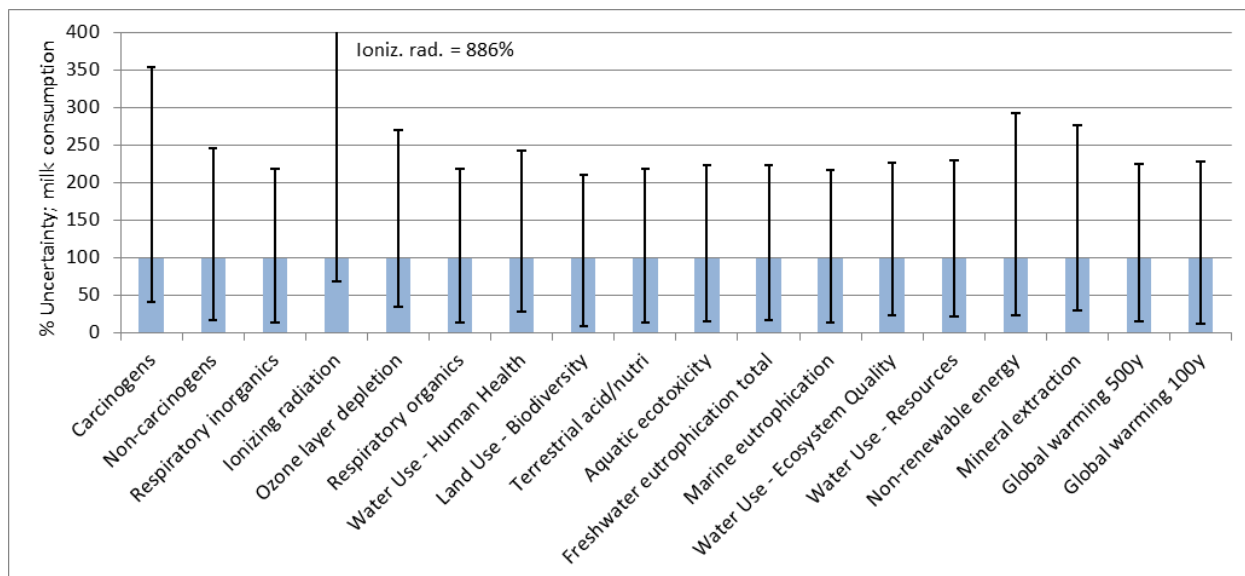


Figure S4. Overall characterization uncertainty analysis of consumption of 1 kg FPCM, Monte Carlo simulation, n=1000.

In keeping with the current state of practice in life cycle assessment, quantitative estimates of uncertainty for impact methods have not been determined. Therefore, uncertainty associated with each impact method will be qualitatively discussed. We use guidelines from Humbert et al. [74] for minimum significant differences for determining whether differences in LCIA results are meaningful. In the energy and global warming category, this minimum significant

difference is 10% (i.e., in comparing contributions to this category, a difference lower than 10% is not considered to be significant). For respiratory inorganics, acidification, and eutrophication, the minimum significant difference is 30%. Since this study has developed new eutrophication methods, this study uses a factor of 10 for eutrophication. For the toxicity categories, we use a minimum significant difference of a factor of 100, based on expert judgment. For the water scarcity impact category, Henderson et al. [21] have carried out a detailed Monte Carlo analysis, combining the above described inventory uncertainty with uncertainty on the water scarcity index from Pfister et al. [47], demonstrating for this impact category that spatial variability is larger than uncertainty, and that not systematically accounting for variability can lead to high uncertainties.

6 Eutrophication

6.1 Emissions and inventory

The data used to assess eutrophication-related emissions due to feed and dairy production are summarized in the main text. The application of manure to fields by the milk producer falls outside the system boundary of this national assessment; the inclusion of manure in crop production accounts for this manure from a life cycle perspective. The only sources of spatial phosphorus emissions are inorganic and manure fertilizer application to feed crops. Ammonia and nitrate emissions are also generated from crop production, and they may come from dairy cow housing and manure management. Inventory data for crop emissions are derived from the National Nutrient Loss and Soil Carbon Database (NNLSC) [33,75]. Data from Pinder et al. [70] were used to calculate spatially-varying ammonia emissions due to housing. IPCC estimates for ammonia losses during manure management and storage were used to calculate spatial inventories of ammonia losses [69].

6.2 Freshwater eutrophication impact assessment model

This study has created new, spatially-resolved characterization factors for freshwater eutrophication, building on recent developments in phosphorus fate modeling, coupling this to a concentration-dependent effect factor component. The fate mode is parameterized at a resolution of $0.5^\circ \times 0.5^\circ$ (at moderate latitudes, this is equivalent to approximately 50 x 50 km), making it possible to understand phosphorus fate and transport across the U.S. [48].

Table S8 summarizes the used effect factors for P-related freshwater eutrophication taken from Payet [51], reporting phosphorus concentration levels and corresponding effect factors for both river and lake systems. The largest variation is between lotic and lentic systems with a factor of 5 or 10 difference in EF, the differences between different trophic states being modest, lower than a factor of 2.4. In contrast, variations in fate factors span orders of magnitude. These effect factors report volume-based impact and were then normalized to account for freshwater depth to generate area-based effect factor. The fate model was then combined with the concentration-dependent effect factor component, enabling the creation of 0.5 by 0.5° characterization factors.

Table S8. Phosphorus levels and effect factors corresponding to river and lake systems at different trophic levels [51].

	Oligotrophic		Mesotrophic		Eutrophic (Assumed Equal to Mesotrophic)	
System	Total P ($\mu\text{g/L}$)	EF (PDF.m ³ .kg ⁻¹)	Total P ($\mu\text{g/L}$)	EF (PDF.m ³ .kg ⁻¹)	Total P ($\mu\text{g/L}$)	EF (PDF.m ³ .kg ⁻¹)
River (lotic)	0-100	680	100-250	280	250 +	280

Lake + Reservoir (lentic)	0-10	3000	10-35	2400	35-100	2400
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Figure S5 summarizes the resulting fate factors (a), effect factors (b), and characterization factors on the native scale of 0.5 by 0.5° (c) and aggregated by state (d).

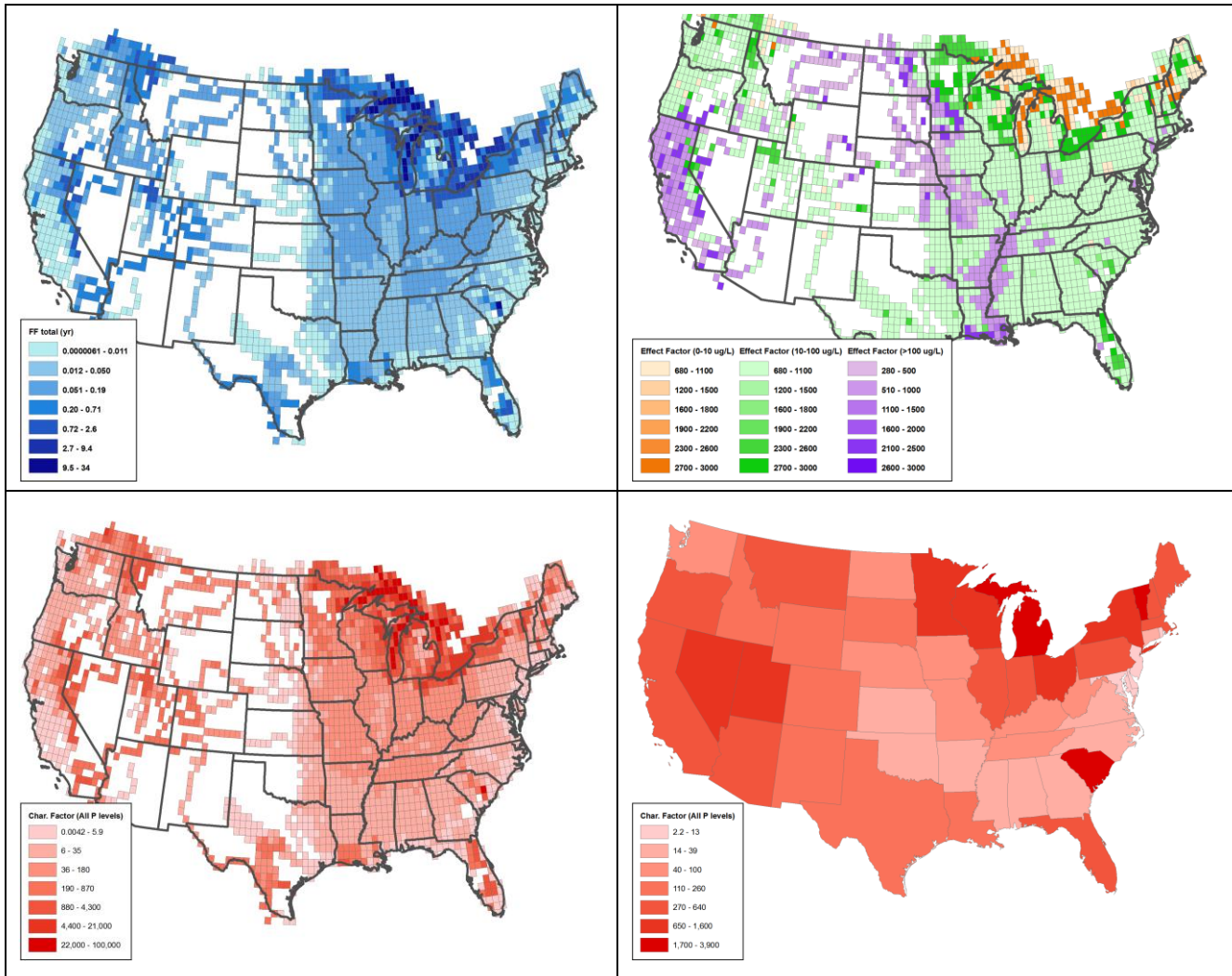


Figure S5. (a) Fate factors [yr], (b) Effect factor [PDF·m²·kg⁻¹], (c) native and (d) state average characterization factors [PDF·yr·m²·kg⁻¹] for phosphorous emissions in the United States.

The presence of large water bodies, such as the Great Lakes, has a significant impact on the fate factor and related range of variation in residence times, which spans several orders of magnitude (Figure S5a). Some cells in the Southwest have fate factor values of zero, shown as white, because evaporation exceeds precipitation in these arid cells. The effect factor is expressed in potentially disappeared fraction of species over one square meter per kilogram of phosphorus in water [PDF.m2.kg⁻¹]. Grid cells are color-coded according to their existing phosphorus level classification (Figure S5b). Cells with greater effect factors – especially in the Great Lakes region, correspond to cells with high fate factor. For characterization factor, Figure S5c,d) distinguish several regions of North America: An emission in the arid Southwest has a relatively low characterization factor, whereas an emission in the Great Lakes region has a high characterization factor, driven in large part by the high fate factor for that region and a relatively greater effect factor. Nevada, as its few non-arid cells are affected by dams, also has high CFs. Emissions near the coast have a lower

characterization factor than those further inland because the residence time of phosphorus tends to be greater further inland.

6.3 Marine eutrophication

For nitrogen species, this study used existing TRACI [41] fate factors for the transfer to the sea, but developed new estimates for marine eutrophication damages. Overall, nitrate and ammonia characterization factors vary between U.S. states by, at most, a factor of 50%.

To estimate marine eutrophication damage from the release of nitrogen to the sea, we combined data available for the export of nitrogen from U.S. watersheds to coastal systems to data for the eutrophication-induced oxygen depletion caused by these flows of nitrogen for selected areas, such as the Mississippi and Atchafalaya basins [76,77], and the Chesapeake basin [78]. We therefore calculated an area of coastal waters affected by marine eutrophication and relate this to the total nitrogen loading. For the purposes this simplified estimate, we assume that the fraction of species affected in an affected eutrophied area is unity, for the oxygen depletion represents a drastic change from normal, oxygenated conditions, causing a dramatic shift in species.

For the Gulf of Mexico, nitrogen exports between 1985 and 2005 averaged in the order of 1,400,000 metric tons of nitrogen per year. This corresponded to an annual hypoxic area of nearly 13,000 to 17,000 km². Thus, the eutrophication impacts of nitrogen in the Gulf of Mexico can be expressed as 9.7 to 12.6 PDF·m²·yr / kg N_{delivered}. For the Chesapeake Bay, total nitrogen flow into the bay was approximately 95,000 metric tons per year, corresponding to an average hypoxic area of approximately 1100 km², corresponding to the characterization factor of 11.7 to 13.0 PDF·m²·yr / kg N_{delivered}. As a first order approximation, these two characterization factors show very good agreement and also correspond to a similar ratio of 12.9 for the Baltic Sea. We retained as characterization factor the weighted average between these two characterization factors of 12.5 PDF·m²·yr / kg N_{delivered}.

Table S9. Determination of the effect factor for marine eutrophication based on total nitrogen delivered to three partly eutrophied water bodies and corresponding average observed hypoxia area.

Water Body	Analysis Years	Total Nitrogen/ Phosphorus (<i>t/ year</i>)	Average Hypoxia Area (<i>km²</i>)	Hypoxia Area/ kgN (PDF m ² year/ kg N)
Gulf of Mexico	1985-2010	1,419,760	13,810	9.7
Gulf of Mexico	2005-2010	1,368,200	17,300	12.6
Chesapeake Bay	1985-2011	91,330.45	1,183.20	13.0
Chesapeake Bay	1970-2011	94,823.95	1,105.40	11.7
Baltic Sea	1995-2009	3,729,000.00	48,000.00	12.9

7 Human toxicity and ecotoxicity

Crop production, dairy farm activities, and electricity use can all generate primary and secondary airborne particulate matter, which cause impacts on human health. In addition, application of pesticides, fungicides, and herbicides to feed, to dairy cows, and to dairy facilities can have toxicity impacts on ecosystems and humans. Application, fate and transport, and impact of pesticides, as well as ammonia emissions from the farm and related generation of particulate

matter, were modeled at a sub-national level. Other impacts, such as those corresponding to tractor and transport emissions, were assessed as upstream and downstream sources. Overall normalized results from Figure 3e and Figure 3f from the main text suggest that the contribution of pesticides to both human toxicity and aquatic ecotoxicity are restricted compared to the effect of respiratory organics for human health and to other ecosystem impacts. These primary and secondary particulate matter emissions stem from ammonia emissions at the dairy farm, tractor and other emissions during feed production, and transport of milk. The human toxicity impacts of pesticides and other substances were found to be an order of magnitude smaller than human health impacts due to respiratory inorganics. In the overall life cycle impact assessment for ecotoxicity, the pesticide impacts were found to be of equivalent importance as metal emissions from on farm activities (metals in phosphorus fertilizer) and upstream processes such as electricity generation (copper) and petroleum refining (barium). At the national milk level, a small group of substances account for the majority of the human toxicity and ecotoxicity impacts. For ecotoxicity, atrazine, metolachlor, cyfluthrin, and acetochlor account for 91% of total impact per kg milk. Atrazine was the dominant impact for both human toxicity and ecotoxicity. For human toxicity, atrazine, chlorpyrifos, dimethoate, and metolachlor account for 90% of total impact per kg milk.

Tables S10 and S11 disaggregate national level impacts according to main inventory flows contributing to Ecotoxicological and human toxicity impacts, and the processes of milk production that induce those flows. Feed mix (modeled as a mixture of corn grain and soybean), corn silage, and the related application of atrazine, chlorpyrifos, and metolachlor account for a large fraction of impacts. This analysis highlights the importance of working to minimize the use, as well as loss during use, of these substances.

Table S10. Top flow contributions for ecotoxicological impacts (PDF·m²·yr/kg FPCM from field and dairy farm), arranged in decreasing order along columns (sources of inventory flow) and rows (substances emitted).

Flow	Feed Mix	Corn Silage	Corn Grain	Facility Pest.	Other Crops	Percent of Total
Atrazine	2.1E-2	4.1E-3	4.0E-3	-	1.9E-3	70%
Metolachlor	3.7E-3	7.6E-4	7.7E-4	-	3.9E-4	13%
Cyfluthrin	-	-	-	2.2E-3	1.6E-4	6%
Acetochlor	7.4E-4	1.3E-4	1.3E-4	-	6.2E-5	2%
Other pesticides	1.3E-3	2.6E-4	2.6E-4	8.3E-4	1.3E-3	9%
Percent of total	60%	12%	12%	7%	9%	100%

Table S11. Top flow contributions for human toxicological impacts (DALY/kg FPCM from field and dairy farm), arranged in decreasing order along columns (sources of inventory flow) and rows (substances emitted).

Flow	Feed Mix	Corn Grain	Corn Silage	Soybean Meal	Other Crops	Percent of Total
Atrazine	1.1E-9	2.1E-10	1.9E-10	-	1.0E-10	61%
Chlorpyrifos	2.4E-10	4.4E-11	3.6E-11	8.4E-11	7.9E-11	19%
Dimethoate	-	-	-	-	1.6E-10	6%
Metolachlor	7.8E-11	1.6E-11	1.1E-11	7.5E-13	7.9E-12	4%
Other pesticides	7.9E-11	1.4E-11	1.2E-11	5.8E-11	8.2E-11	10%
Percent of total	57%	11%	10%	5%	17%	100%

The combination of ecotoxicity across the milk production life cycle is shown in Figure 3e. Impacts, due to atrazine, metolachlor, and other pesticides, account for close to half of the feed production impact. Likewise, cyfluthrin and other pesticides applied to cows or to dairy facilities account for the other half of the milk production impact. The other half of the feed production and milk production ecotoxicological impacts are due to emissions of metals.

Figure 3f, in the main text, shows the human health impacts across the milk life cycle, with a combined human health impact on the order of $1 \cdot 10^{-6}$ DALY / kg FPCM consumed. The majority of impacts are caused by respiratory inorganic emissions of ammonia, nitrogen oxides, and sulfur dioxides. These substances are important across all stages of life cycle, with ammonia emissions largely due to feed production or manure management, and the other substances associated with fuel use with respect to human health.

In the consumer stage of the life cycle, there is a small human health impact due to the presence of dieldrin in milk that is consumed. This compound is not present due to application to crops, introduction during the processing, or any other life cycle stage. Rather, this is a globally distributed, legacy pollutant that has been detected in milk by the USDA (USDA AMS 2006). In general, residues of pesticides detected in milk are observed infrequently, are associated with legacy pesticides, and have limited human toxicity impacts.

8 Normalization

Milk production and consumption: Dairy production data are available from USDA [61]; these are corrected for FPCM based on state data from the dairy survey [65] to yield a total production of 8.035×10^{10} kg FPCM in 2007.

Milk consumption data are available from USDA ERS [79] and population from the US Census intercensal estimates [80]. 2007 per capita consumption value (59 kg milk / pers) is multiplied by population (3.016×10^8 pers) to yield a total consumption of 1.8×10^{10} kg milk consumed. We assume that consumed milk, representing a mix of whole, skim, etc., is equivalent to FPCM.

Normalization values: For most impact categories, normalization factors for the United States were used [81]. Exceptions to this approach are described below.

8.1 Calculation of aquatic eutrophication damage factors

Based on the fate and effect models described in the main text, the U.S. average damage factor for P emissions to freshwater is $192.7 \text{ PDF} \cdot \text{m}^2 \cdot \text{yr} / \text{kg P}$, while that for N emissions to marine ecosystems is $12.5 \text{ PDF} \cdot \text{m}^2 \cdot \text{yr} / \text{kg N}$. Table S12 provides a summary of the calculation approach for calculating normalization values. For each substance, the transfer fraction relates an emission in a given compartment to delivery to the compartment of interest; this is used to calculate a damage factor. For example, for phosphorus emissions to soil, the transfer of 13% is applied to the damage factor above, $192.7 \text{ PDF} \cdot \text{m}^2 \cdot \text{yr} / \text{kg P}$. Total U.S. emissions of each substance are based on Lautier et al. [81] and are used for normalization. Some substances are included in Table S12 even though emissions data for the substances may be lacking or may have been combined to yield overall data for other substances; these are common substances and will provide a useful frame of reference for users. In the last column of the table, freshwater P eutrophication and marine in eutrophication have been summed. These values are the overall aquatic ecosystem damage factors; they are used as input in the recalculation of normalization factors, discussed in the next section.

Table S12: Damage normalization calculation for aquatic eutrophication. Dashes indicate no known inventory values.

Compartment of Emission	Substances	Unit	Transfer Factors	Damage Factor	Emissions	Damage Normalization Values
Freshwater (P) eutrophication			kg P to freshwater / kg P compartment	[PDF·m ² ·yr/kg]	[kg/yr]	[PDF·m ² ·yr / kg]
Soil	Phosphorus	kg	0.13 ^a	192.7	0.0E+00	0.0E+00
Water	Phosphorus	kg	1.00	192.7	3.0E+08	5.7E+10
Air	Phosphorus	kg	1.00	192.7	5.4E+07	1.0E+10
Water	COD, Chemical Oxygen Demand	kg	1.00	1.4	0.0E+00	0.0E+00
Air	Phosphate	kg	1.00	63.0	0.0E+00	0.0E+00
Soil	Phosphate	kg	1.00	63.0	0.0E+00	0.0E+00
					SUM	6.7E+10
Marine (N) eutrophication			kg N to marine water / kg Substance to compartment ^b	[PDF·m ² ·yr/kg]	[kg/yr]	[PDF·m ² ·yr / kg]
Air	Ammonia	kg	0.12	1.5	4.1E+09	6.1E+09
Soil	Nitrogen	kg	0.25	3.1	0.0E+00	0.0E+00
Marine water	Nitrogen	kg	1.00	0.0	0.0E+00	0.0E+00
Water	Nitrogen	kg	0.99	12.3	2.8E+09	3.5E+10
Air	Nitrogen oxides (as NO ₂)	kg	0.04	0.6	1.5E+10	8.2E+09
Air	Nitrogen dioxide	kg	0.04	0.6	0.0E+00	0.0E+00
					SUM	4.9E+10

^a for emissions to soil, the average ratio of P applied and P loss to water in the NNLSC/EPIC database for corn grain, corn silage, hay, legume (alfalfa) hay and soybean was used [33,34].

^b transfer factors for delivery of N to marine water for various types of emissions are taken from U.S. averages provided in TRACI [41].

8.2 Recalculation of normalization factors for ecosystem quality

For terrestrial ecotoxicity, the approach taken in IMPACT 2002+ [39], and in other LCA methods, is problematic for two reasons. First, there were not – and to a large extent, still are not – specific terrestrial ecotoxicity effect data available. Therefore, results for terrestrial species were extrapolated from aquatic ecotoxicity tests, assuming that only the dissolved fraction in water is bioavailable to terrestrial species, with a correction for adsorption to soil. Second, for an LCA in the agricultural context, the land use impact on ecosystems accounts for the loss of biodiversity in the terrestrial ecosystem, suggesting a risk of double counting. Therefore, in keeping with choices in USEtox [82], terrestrial ecotoxicity has not been considered in the present impact analysis.

Accounting for eutrophication at endpoint level and not considering terrestrial ecotoxicity means that new normalization values at the damage level need to be recalculated for IMPACT 2002+. Tables S13 and S14 present this recalculation. First, Table S13 shows the approach taken to account for species richness in terrestrial vs. aquatic ecosystems. In previous impact assessments, land and water were weighted equally on an area basis. However, this does not take into account the relative abundance of species in aquatic systems, given the relative scarcity

of aquatic habitat. Estimates for total numbers of described species and ecosystem areas are used to calculate the species richness per area. Then, these values are normalized against the terrestrial ecosystem species richness. As shown in the last column of Table S13, this approach indicates that at a global scale, there are 2.4 times as many species per unit area freshwater systems as opposed to terrestrial ecosystems.

Table S13: Derivation of aquatic ecosystem species correction factor.

System	Total Global Number of Described Species [40]	Total Global Area [83]	Species per Area	Correction Factor for Aquatic Species
	species	m ²	species / m ²	[-]
Terrestrial	1.50E+06	1.29E+14	1.16E-08	1.00
Freshwater	1.00E+05	3.63E+12	2.75E-08	2.4
Marine	2.50E+05	2.20E+13	1.13E-08	0.98

The damage normalization values for freshwater and marine eutrophication from Table S12 are used in the second column of Table S14. Values for other classes come from the IMPACT 2002+ method. These total damages are divided by the U.S. population to create a population-normalized damage (column three). Finally, the species richness correction from Table S13 is used to create a corrected value, shown in the final column. Two sums are shown; one excludes terrestrial ecotoxicity, and the final one includes this category. As discussed above, inclusion of terrestrial ecotoxicity is problematic in LCA; therefore, in this study, the normalization value of 5200 PDF·m²·yr / pers.yr was used.

Table S14. Ecosystem quality normalization values, with and without terrestrial ecotoxicity.

Classes	Total Damage Normalization Value (PDF.m ² .yr/yr)	Damage / pers.yr (PDF.m ² .yr / pers.yr)	Aquatic Species Correction Factor	Corrected Value (excl. Terrestrial Ecotoxicity) (PDF.m ² .yr / pers.yr)
Aquatic ecotoxicity	1.6E+10	53	2.37	130
Aquatic eutrophication (P-limited watershed)	6.7E+10	220	2.37	530
Marine eutrophication	4.9E+10	160	0.98	160
Land occupation	1.1E+12	3800	1.00	3800
Terrestrial acidification / nitrification	1.6E+11	530	1.00	530
		SUM (excl. terrestrial)		5200
Terrestrial ecotoxicity	1.1E+12	3800	1.00	3800
		SUM (incl. terrestrial)		9000

Finally, Table S15 summarize the normalization values (and optional weighting factors) used.

Table S15. U.S. specific normalization values used for each area of protection.

Area of Protection	Impact Category	Unit	Normalization Value for U.S.	Stepwise Weighting Factor Weidema [84]
Human health	Total Human Health impacts per person	DALY/pers/yr	1.2E-01	74,000 €/DALY
Ecosystem Quality	Total Human Health impacts per person	PDF·m ² ·yr/pers/yr	2.2E+04	0.14 €/ PDF·m ² ·yr
Resources	Total Human Health impacts per person	MJ primary/pers/yr	2.9E+05	0.0043 €/MJ
Water scarcity		m ³ world avg/per/yr	3.1E+04	-

8.3 Normalized damages per kg fluid milk consumed

Figure S6 presents normalized impact per kg FPCM_{consumed} in log scale (see Figure 5, main text for arithmetic scale), showing five orders of magnitude variation across the different normalized impact category values.

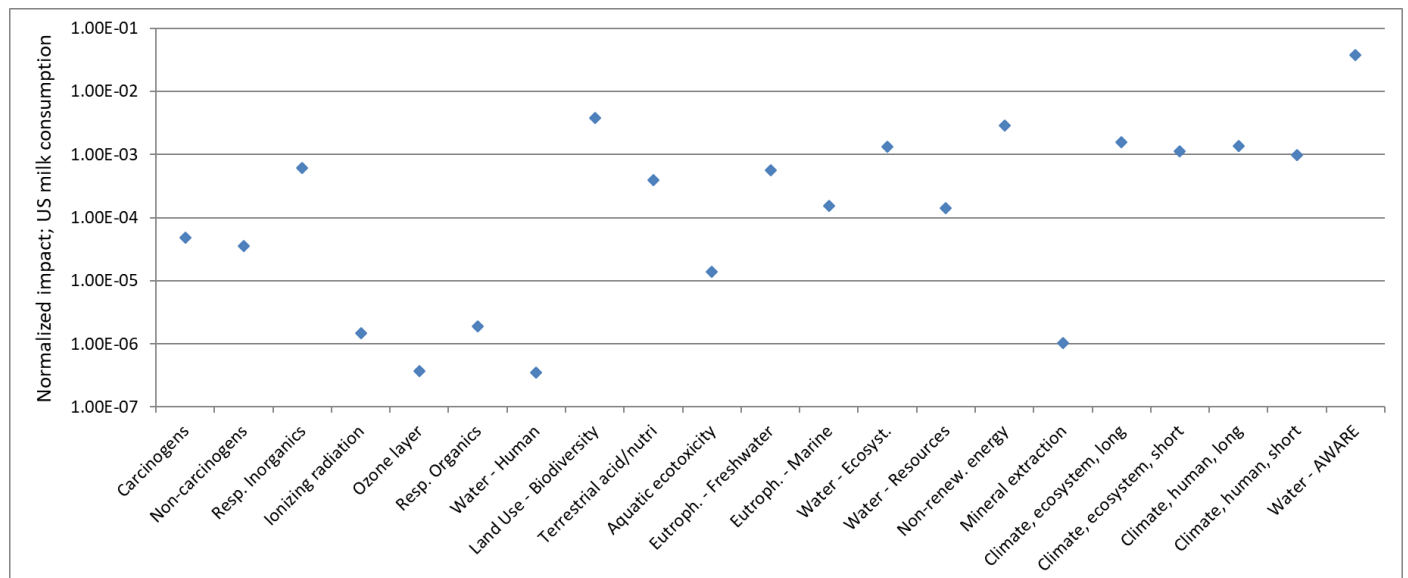


Figure S6. Normalized impacts for the annual consumption of 58 kg of U.S. fluid milk per person according to the customized IMPACT 2002+ method (in log scale).

8.4 Normalized damages per kg fluid milk produced

Figure S7 presents the normalized impact per kg FPCM_{farm} in regular scale, showing the contribution of milk production to the average impacts of a U.S. person. For the main contributing impact categories, milk production contributes to 0.5% to 1.5% for fine particulate impacts on human health and carbon footprint, and to 2% to 6.5% for water and land use impacts on ecosystems.

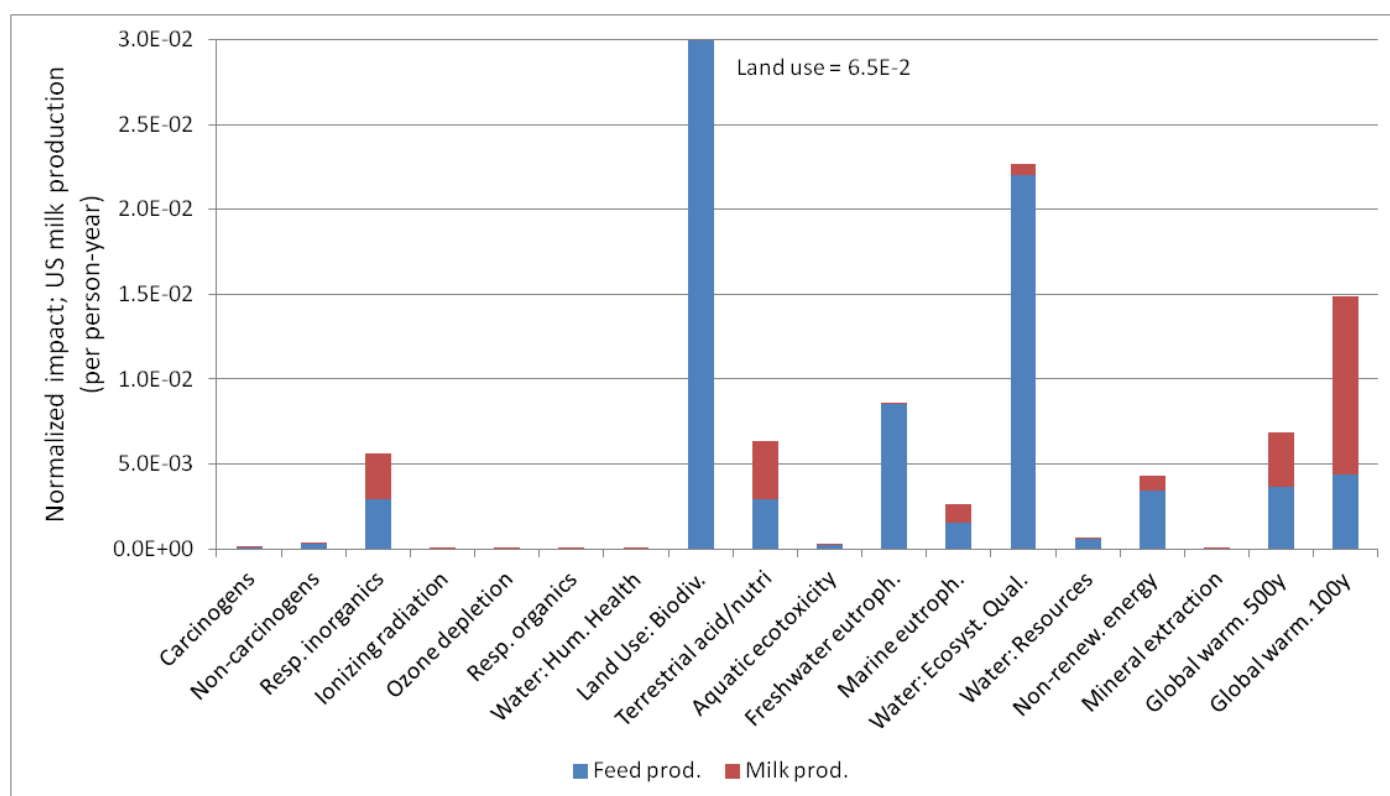


Figure S7. Normalized impacts for the annual production of 290 kg of U.S. milk or 276 kg FPCM per person according to the customized IMPACT 2002+ method.

9 Comparison with other impact assessment methods

Impact assessment results from the customized IMPACT 2002+ method per kg fluid milk consumed are compared with other methods in order to assess influence of the impact assessment method on the results. ReCiPe [40] (Figure S8) produces results that are largely similar to the customized IMPACT 2002+ (Figure 5, main text). Differences include the fact that climate change impacts are not tallied separate in ReCiPe but are translated into contributions to human health and, to a lesser extent, ecosystem impacts. On-farm emissions also dominate climate change and particulate matter impacts (called respiratory inorganics in IMPACT 2002+). For the ecosystem area of protection, agricultural land use impacts for feed production are even more dominant with ReCiPe than with IMPACT 2002+. Freshwater eutrophication is also dominated by feed production but is not relevant in the normalized impacts. This finding is expected, given that this study developed new eutrophication impact assessment methods.

TRACI [41] (Figure S9) also mostly identifies the same main processes with high contributions to the different impact categories: on farm and manure emissions for global warming, acidification, and marine eutrophication; feed production is important for freshwater eutrophication and ecotoxicity. One main difference is the dominating influence of consumer use on carcinogenic and non-carcinogenic impacts, due to TRACI's characterization of lead emissions to water associated with waste treatment of plastic milk containers.

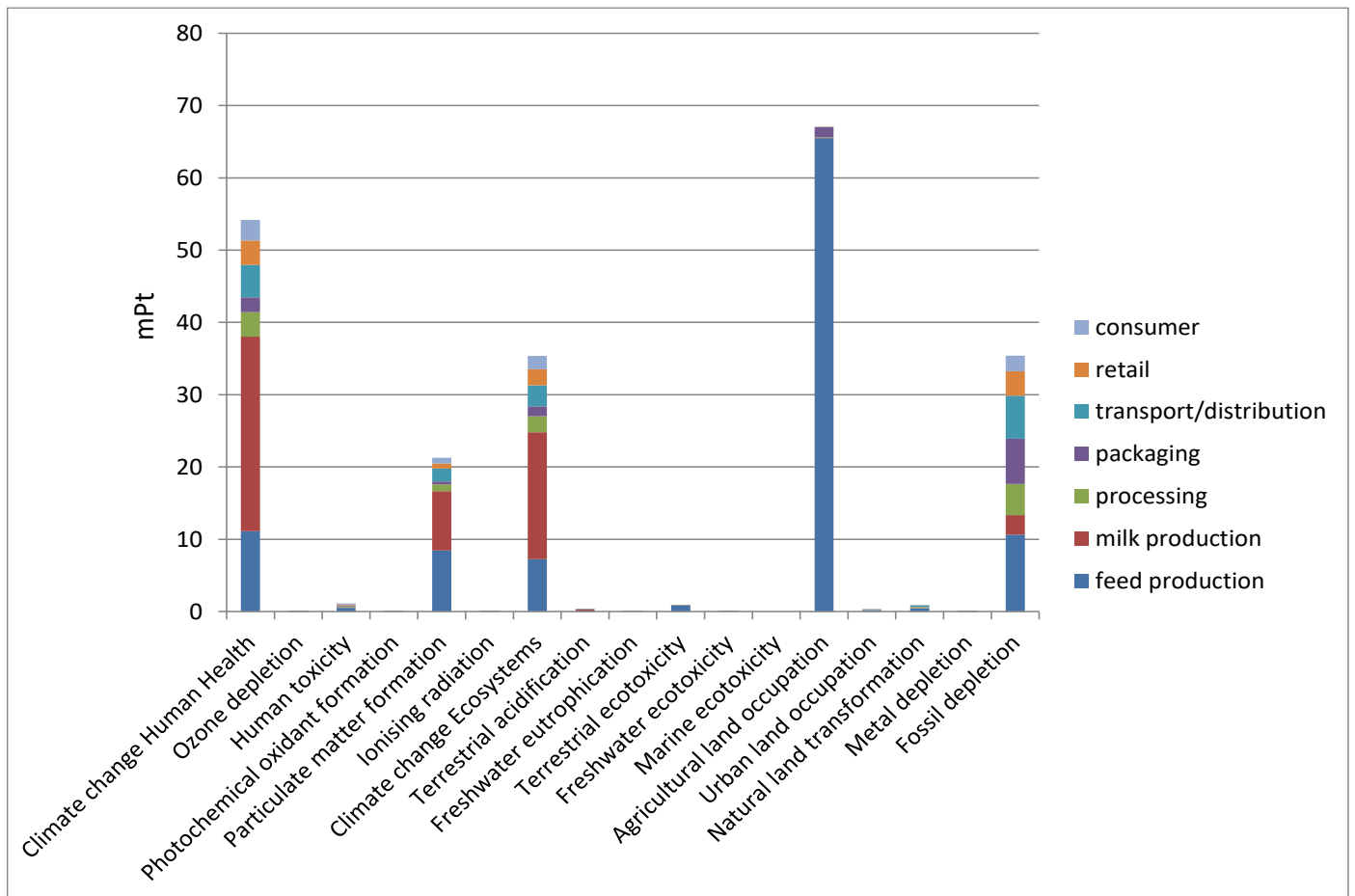


Figure S8. Weighted damage assessment of U.S. fluid milk consumed using ReCiPe (hierarchist perspective).

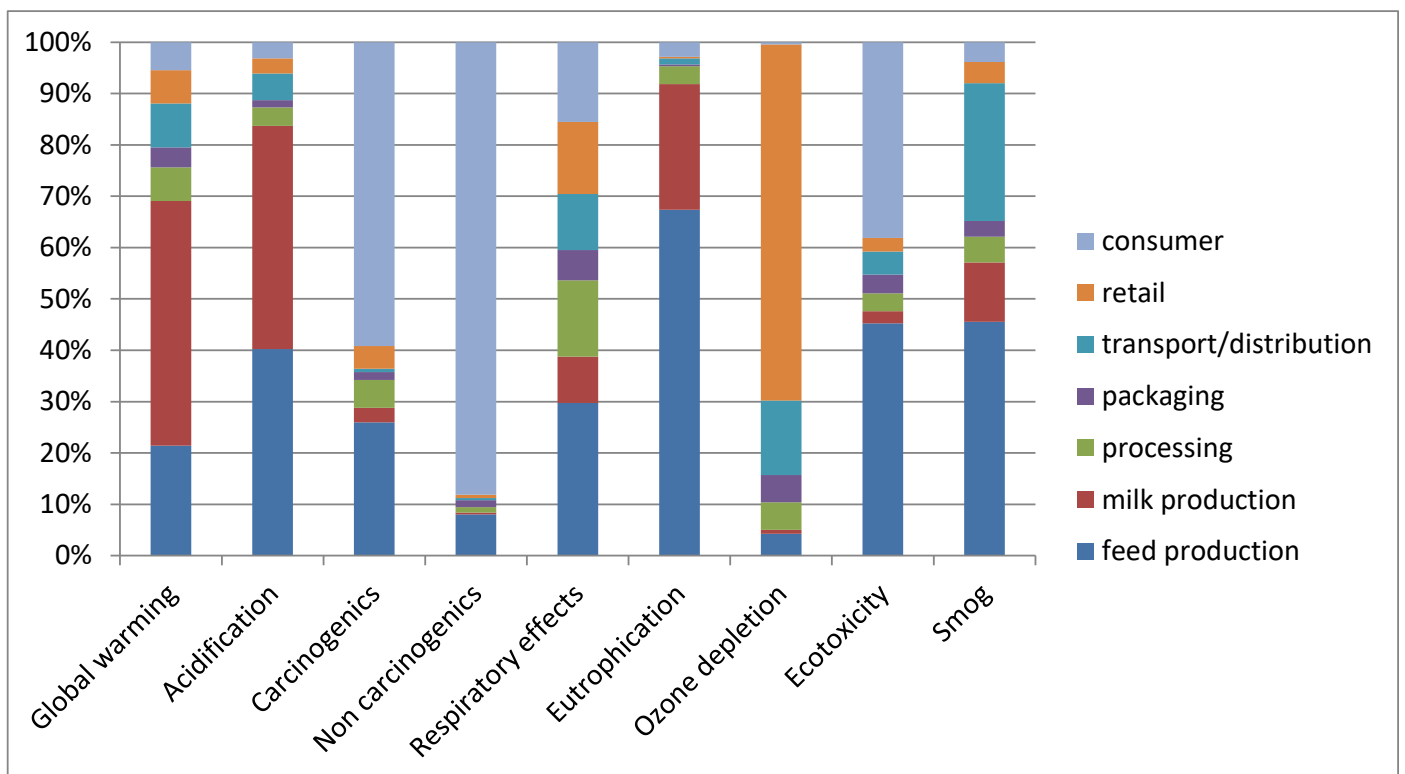


Figure S9. Characterization of U.S. fluid milk consumed using TRACI.

10 Matrices

Provided as separate Microsoft Excel files at <https://zenodo.org/record/7378838>.

References

See main text.