Supporting Information
“Changes in environmental impacts of major crops in the U.S.”

Yi Yang and Sangwon Suh*
Bren School of Environmental Science and Management, University of California, Santa Barbara,
CA 93106-5131

* The author to whom correspondence should be made
Email: suh@bren.ucsb.edu
Office Phone: (805) 893-7185
Fax: (805) 893-7612

Table 21
Page 21
1. Impact categories selected from TRACI 2.0 for analysis

| Impact category          | Media      | Unit                   | Explanation                                                                 |
|--------------------------|------------|------------------------|-----------------------------------------------------------------------------|
| Acidification            | air        | moles of H+ eq         | Increases in hydrogen ion (H+) within a local environment                   |
| Eutrophication           | water      | kg N eq                | Nutrient buildup in aquatic ecosystems that stimulates excessive biological productivity such as algal growth |
| Smog formation           | air        | kg O₃ eq               | Photochemical reactions between nitrogen oxides (NOₓ) and volatile organic compounds (VOCs), resulting in a range of respiratory issues |
| Freshwater ecotoxicity   | multimedia | CTUe                   | Estimates of the potentially affected fraction of species (PAF), expressed in comparative toxic units (CTUe) |
| Human health respiratory | air        | kg PM₁₀ eq             | Criteria pollutants such as particulate matter that cause respiratory illness and death. |
| Human health cancer      | multimedia | CTUh                   | Increases in morbidity in the total human population, expressed in comparative toxic units (CTUe) |
| Human health noncancer   | multimedia | CTUh                   | Increases in morbidity (e.g., reproductive, developmental, and neurotoxic effects) in the total human population, expressed in comparative toxic units (CTUe) |
2. Numerical information on inputs applied to major crops in the U.S.

Tables S2-S5 present inputs data that were used for LCA calculation in this study. It is worth noting that a large number of pesticides were applied to the four crops in the years investigated: 84 for corn, 113 for cotton, 81 for soybean, and 65 for wheat. But these are not the complete list of pesticides applied because USDA, the department that collects such data, often withholds information on certain pesticides to avoid disclosing data from individual farms. The pesticides unpublished are usually applied in a small number of states and in small amounts, which get lumped in the total amount of herbicides, insecticides, fungicides, or other chemicals disclosed for all the states surveyed. The amount of pesticides published and specified in general accounts for 95% of the total applied.

There are a few pesticides not consistently published for the years investigated; they are denoted “NA” in Tables S2-S5. The inconsistency reflects, in most cases, changes in the number of states applying the pesticides and, as a result, changes in the amount applied. For example, bromoxynil was published for corn production in years 2001 and 2005 but not for that in 2010. A closer examination of the state-level pesticide data shows that the pesticide was applied in 12 states in 2001 and 9 states in 2005 but only in 1 state in 2010.

It was hypothesized that given their small amounts, the unpublished pesticides (i.e., NAs) would have little effect on the overall freshwater ecotoxicity impact. To rigorously test this hypothesis, we run the following analysis. First, we replaced all the NAs for given years with maximal rates (kg ha\(^{-1}\)) identified from other years. In the case of bromoxynil applied to corn, for example, we replaced the NA for 2010 with 0.005773 from 2001. This would be considered a worst-case, unlikely scenario given the number of states dropped significantly that applied this pesticide.

Next, we recalculated the freshwater ecotoxicity impact and compared how much it had changed for each crop. The results generally confirmed our hypothesis. The new ecotoxicity impact per ha corn produced would increase by <2% for all years; the same goes for soybean. The new impact per ha wheat produced would increase by <6% for all years. And the new impact per ha cotton produced would increase by <2% for 2000 and 2003 but by around 13% for 2007. This is because the unpublished pesticides are generally in small amounts relative to other pesticides and their freshwater ecotoxicity potentials as assigned by TRACI 2.0 are also relatively small. The results also identified several pesticides worth of further analysis.

For 2007 cotton, we identified cyanazine, fenpropathrin, and profenofos as the major contributor to the large difference (i.e., 13% increase) if their NAs (kg ha\(^{-1}\)) were replaced with maximal rates. We took a further look into the state-level data and estimated their 2007 application rates (kg ha\(^{-1}\)) based on the number of states that applied these pesticides in that year relative to the number of state that applied the pesticides in 2003 given no further information available. All other pesticides with NAs were treated as 0s in our LCA analysis considering their insignificant contributions as demonstrated above.
Table S2. Inputs use in U.S. corn production (ha⁻¹)

| Energy       | 2001 | 2005 | 2010 |
|--------------|------|------|------|
| diesel       | kg   | 49   | 45   | 46   |
| gasoline     | kg   | 12   | 13   | 13   |
| lpg          | kg   | 22   | 16   | 17   |
| electricity  | kwh  | 103  | 60   | 64   |
| natural gas  | mj   | 982  | 902  | 1052 |

| Nutrient     | 2001 | 2005 | 2010 |
|--------------|------|------|------|
| nitrogen     | kg   | 155  | 159  | 161  |
| phosphate    | kg   | 54   | 56   | 56   |
| potash       | kg   | 66   | 65   | 58   |
| sulfur       | kg   | 0    | 2    | 2    |

| Pesticide    | 2001 | 2005 | 2010 |
|--------------|------|------|------|
| 2,4-d        | kg   | 0.039607 | NA   | NA   |
| 2,4-d, 2-ehe | kg   | 0.008282 | 0.018215 | 0.021629 |
| 2,4-d, bee   | kg   | 0.000584 | 0.001979 | 0.004182 |
| 2,4-d, dieth. salt | kg | 0 | 0.000393 | 0 |
| 2,4-d, dimeth. salt | kg | NA | 0.012546 | 0.016774 |
| 2,4-d, isoprop. salt | kg | 0 | 0.000597 | 0.000351 |
| acetochlor   | kg   | 0.548841 | 0.467966 | 0.408313 |
| acifluorfen, sodium | kg | 0 | 0 | 0.000307 |
| alachlor     | kg   | 0.05404 | 0.024527 | 0.006025 |
| ametryn      | kg   | 0.001426 | 0.000283 | 0 |
| atrazine     | kg   | 1.069847 | 0.901167 | 0.747703 |
| azoxystrobin | kg   | 0 | NA | 0.001492 |
| bentazon     | kg   | 0.004364 | NA | 0 |
| bifenthrin   | kg   | 0.001151 | 0.001131 | 0.000994 |
| bromoxynil   | kg   | 0.005773 | 0.003125 | NA |
| bromoxynil heptan. | kg | 0 | 0.000879 | NA |
| bromoxynil octanoate | kg | NA | 0.001335 | NA |
| carbofuran   | kg   | 0.008179 | 0.001774 | NA |
| carfentrazone-ethyl | kg | 0.0001137 | 7.85E-05 | 5.85E-05 |
| chlorpyrifos | kg   | 0.062941 | 0.032143 | 0.00699 |
| clopyralid   | kg   | 0.012406 | 0.00705 | 0.004797 |
| cyanazine    | kg   | 0.009433 | 0.006108 | 0 |
| cyfluthrin   | kg   | 0.000275 | 0.000597 | 0.000219 |
| dicamba      | kg   | 0.029572 | 0.002418 | 0.000336 |
| dicamba, digly salt | kg | 0 | 0.00592 | 0.00196 |
| dicamba, dimet. salt | kg | 0.00555 | 0.004742 | 0.007019 |
| dicamba, pot. salt | kg | 0.020121 | 0.008746 | 0.001696 |
| dicamba, sodium salt | kg | 0.000344 | 0.00548 | 0.003838 |
| diflufenzopyr-sodium | kg | 0.002337 | 0.001931 | 0.001155 |
| dimethenamid | kg   | 0.131278 | 0.010756 | 0.007604 |
| dimethenamid-p | kg | NA | 0.037246 | 0.038066 |
| dimethoate   | kg   | 0.002818 | 0.001068 | 0.00076 |
| epc          | kg   | 0.054247 | NA | NA |
| esfenvalerate| kg   | 1.72E-05 | 0.000126 | NA |
| fipronil     | kg   | 0.00445 | 0.001382 | NA |
| flufenacet   | kg   | 0.015344 | 0.011384 | 0.004095 |
| Name                         | Unit | Value 1   | Value 2   | Value 3   |
|------------------------------|------|-----------|-----------|-----------|
| flumetsulam                  | kg   | 0.005189  | 0.003141  | 0.001828  |
| foramsulfuron                | kg   | 0         | 0.000502  | 0.000132  |
| glufosinate-ammonium         | kg   | 0.007268  | 0.021795  | 0.007531  |
| glyphosate                   | kg   | 0.118013  | 0.015027  | 0.076848  |
| glyphosate amm. salt         | kg   | 0         | 3.14E-05  | 0.000673  |
| glyphosate iso. salt         | kg   | 0         | 0.36064   | 0.841398  |
| glyphosate pot. salt         | kg   | 0         | 0         | 0.022258  |
| halosulfuron                 | kg   | 0.000292  | 0.000345  | 4.39E-05  |
| imazapyr                    | kg   | 6.87E-05  | 0.00022   | NA        |
| imazethapyr                 | kg   | 0.000326  | 0.000675  | NA        |
| isoxaflutole                 | kg   | 0.007543  | 0.003659  | 0.005835  |
| lambda-cyhalothrin           | kg   | 0.000395  | 0.000393  | 0.000351  |
| linuron                      | kg   | NA        | 0.01727   | 0.003013  |
| mcpa                         | kg   | 0.000739  | 0         | 0         |
| mcpa, sodium salt            | kg   | 0         | 0.004051  | 0.00253   |
| mesotrine                    | kg   | 0.00012   | 0.029097  | 0.024758  |
| methyl parathion             | kg   | 0.006633  | 0.001288  | 0         |
| metolachlor                  | kg   | 0.127996  | 0.030353  | 0.007985  |
| metribuzin                   | kg   | 0.00201   | 0.000471  | NA        |
| nicosulfuron                 | kg   | 0.002784  | 0.00256   | 0.000424  |
| paraquat                     | kg   | 0.008781  | 0.006187  | 0.006844  |
| pendimethalin                | kg   | 0.045122  | 0.020555  | 0.020254  |
| permethrin                   | kg   | 0.004055  | 0.001821  | 0.001053  |
| petroleum distillate         | kg   | 0.000962  | NA        | 0         |
| phorate                      | kg   | 0.001254  | 0         | 0         |
| primisulfuron                | kg   | 0.001718  | 0.000597  | 7.31E-05  |
| propargite                   | kg   | 0.002681  | 0.004538  | 0.001594  |
| propiconazole                | kg   | NA        | NA        | 0.002545  |
| prosulfuron                  | kg   | 0.000309  | 0.00126   | 1.46E-05  |
| pyraclostrobin               | kg   | 0         | 0         | 0.005586  |
| pyridate                     | kg   | 0.001735  | NA        | 0         |
| rimsulfuron                  | kg   | 0.0011    | 0.001696  | 0.000702  |
| saflufenacil                 | kg   | 0         | 0         | 0.000585  |
| simazine                     | kg   | 0.028489  | 0.038063  | 0.032114  |
| s-metolachlor                | kg   | 0.305771  | 0.371396  | 0.319253  |
| spiroxamine                  | kg   | 0         | 0         | 0.000863  |
| sulfosate                    | kg   | 0.013265  | 0.008432  | 0         |
| tebupirimphos                | kg   | 0.006375  | 0.008998  | 0.002852  |
| tefluthrin                   | kg   | 0.008007  | 0.010003  | 0.003539  |
| tembotrione                  | kg   | 0         | 0         | 0.001506  |
| terbufos                     | kg   | 0.042803  | 0.005198  | 0.002003  |
| thiencarbazone-methy         | kg   | 0         | 0         | 0.0006    |
| thifensulfuron               | kg   | 3.44E-05  | 0.00033   | 0.000161  |
| topramezone                  | kg   | 0         | 0         | 0.000132  |
| trifloxystrobin              | kg   | 0         | 0         | 0.001024  |
| trifluralin                  | kg   | NA        | 0.005951  | 0.003758  |
| vernolate                    | kg   | 0.005241  | 0.005245  | 0         |
| zeta-cypermethrin            | kg   | 0         | 0.000173  | 2.92E-05  |
| unspecified                  | kg   | 0.137344  | 0.130566  | 0.139643  |
Table S3. Inputs use in U.S. cotton production (ha⁻¹)

|            | 2000   | 2003   | 2007   |
|------------|--------|--------|--------|
| **Energy** |        |        |        |
| diesel     | kg     | 172    | 146    | 121    |
| gasoline   | kg     | 32     | 27     | 22     |
| lpg        | kg     | 0      | 0      | 0      |
| electricity| kwh    | 139    | 118    | 97     |
| natural gas| mj     | 496    | 422    | 347    |
| **Nutrient**|       |        |        |
| nitrogen   | kg     | 97     | 95     | 97     |
| phosphate  | kg     | 39     | 39     | 34     |
| photash    | kg     | 53     | 53     | 42     |
| sulfur     | kg     | 0      | 0      | 6      |
| **Pesticide**|      |        |        |
| 2,4-D      | kg     | 0.0158 | 0.037292 | NA     |
| 2,4-D, 2-ehe| kg     | 0      | 0      | 0.014339 |
| 2,4-D, dimeth. salt| kg | 0 | NA | 0.039292 |
| abamectin  | kg     | 0.000277 | 0.000296 | 0.000226 |
| acephate   | kg     | 0.119005 | 0.250294 | 0.267368 |
| acetamiprid| kg     | 0      | 0.002269 | 0.002936 |
| aldicarb   | kg     | 0.229417 | 0.197907 | 0.14012 |
| amitraz    | kg     | 0.002125 | 0      | 0      |
| arsenic acid| kg   | 0      | 0.003453 | 0      |
| azinphos-methyl| kg | 0.013213 | NA | 0      |
| azoxystrobin| kg   | 0      | 0.002072 | 0.000452 |
| barban     | kg     | 0      | 0      | 0.000565 |
| bifenthrin | kg     | 0.001756 | 0.000592 | 0.004065 |
| bromoxynil | kg     | 0.046013 | 0.001381 | 0      |
| buprofezin | kg     | 0.000832 | 0.001184 | NA     |
| cacodylic acid| kg | 0.009979 | NA | 0.002936 |
| carbofuran | kg     | 0.015892 | 0.002072 | NA     |
| carboxin   | kg     | 0      | 9.87E-05 | NA     |
| carfentrazone-ethyl| kg | 0 | 0.002664 | 0.002145 |
| chlorpyrifos| kg  | 0.060888 | 0.024171 | 0.002032 |
| clethodim  | kg     | 0.005913 | 0.001381 | 0.000452 |
| clomazone  | kg     | 0.008778 | 0.001579 | 0.000226 |
| cyanazine  | kg     | 0.100803 | 0.00513 | **0.002565** |
| cyclanilide| kg     | 0.017647 | 0.017068 | 0.01863 |
| cyfluthrin | kg     | 0.011272 | 0.006413 | 0.004065 |
| cypermethrin| kg  | 0.007299 | 0.008189 | 0.005871 |
| deltamethrin| kg | 0.003234 | 0.000493 | NA     |
| dicamba    | kg     | NA     | 0.001776 | 0.001581 |
| dicamba, digly. salt| kg | 0 | 0 | 0.010388 |
| Chemical Name                  | Unit | Value 1 | Value 2 | Value 3 |
|-------------------------------|------|---------|---------|---------|
| dicamba, dimet. salt          | kg   | 0       | 0       | 0.005758 |
| dicamba, sodium salt          | kg   | 0       | 0       | 0.00079  |
| dichloropropene               | kg   | 0.118266| NA      | NA      |
| dicrotofos                    | kg   | 0.01774 | 0.009274| 0.004968 |
| dicofol                       | kg   | 0.033632| 0.080603| 0.140233 |
| diflubenzuron                 | kg   | 0.000277| NA      | NA      |
| dimethipin                    | kg   | 0.008223| 0.002466| NA      |
| dimethoate                    | kg   | 0.006745| 0.003354| 0.004178 |
| disulfoton                    | kg   | 0.013028| 0.004341| 0       |
| diuron                        | kg   | 0.105146| 0.171466| 0.149604 |
| dsma                          | kg   | 0.03086 | 0.001776| 0       |
| emamectin benzoate            | kg   | 0.000185| NA      | NA      |
| endosulfan                    | kg   | 0.019865| 0.009964| NA      |
| endothall                     | kg   | 0.00647 | 0.00197 | 0.000565 |
| esfenvalerate                 | kg   | 0.000462| 0.000592| 0.001242 |
| ethephon                      | kg   | 0.495146| 0.696521| 0.998452 |
| etoxazole                     | kg   | 0       | 0       | 0.000452 |
| etridiazole                   | kg   | 0.005451| 0.004045| 0.000903 |
| fenoxaprop                    | kg   | 0.000185| 0       | 0       |
| fenpropathrin                 | kg   | 0.001294| 0.001085| 0.001085 |
| flonicamid                    | kg   | 0       | 0       | 0.00079  |
| fluazifop-p-butyl             | kg   | 0.002587| NA      | NA      |
| flumiclorac-pentyl            | kg   | 0       | NA      | 0.000226 |
| flumioxazin                   | kg   | 0       | NA      | 0.004629 |
| fluometuron                   | kg   | 0.182573| 0.074486| 0.031276 |
| fomesafen                     | kg   | NA      | 0       | 0.00542  |
| glufosinate-ammonium          | kg   | 0       | 0       | 0.008694 |
| glyphosate                    | kg   | 0.880433| 1.246535| 0.085924 |
| glyphosate amm. salt          | kg   | 0       | 0       | 0.009597 |
| glyphosate dia. salt          | kg   | 0       | 0.023184| 0       |
| glyphosate iso. salt          | kg   | 0       | 0       | 1.859043 |
| imidacloprid                  | kg   | 0.001571| 0.001973| 0.004629 |
| indoxacarb                    | kg   | 0.004158| 0.003256| 0.001468 |
| iprodione                     | kg   | 0.001016| 0.001776| NA      |
| lactofen                      | kg   | 0.000832| 0.000592| NA      |
| lambda-cyhalothrin            | kg   | 0.00425 | 0.003552| 0.002371 |
| linuron                       | kg   | 0.001571| 0.007005| 0.0035  |
| malathion                     | kg   | 2.94953 | 0.611774| 0.213398 |
| mefenoxam                     | kg   | 0.001016| 0.001973| 0.000113 |
| mepiquat chloride             | kg   | 0.013582| 0.039562| 0.023485 |
| mepiquat pentaborate          | kg   | 0       | 0       | 0.004855 |
| metalaxyl                     | kg   | 0.000554| 0.000592| NA      |
| methamidophos                 | kg   | 0.007761| 0.000395| 0.000452 |
| Chemical            | Unit | ppp | ccc | ccc                        |
|---------------------|------|-----|-----|----------------------------|
| methomyl            | kg   | 0.00425 | 0.002368 | NA                        |
| methoxyfenozide     | kg   | 0    | 0.000493 | 0                          |
| methyl parathion     | kg   | 0.075302 | 0.014503 | 0.0035                    |
| metolachlor          | kg   | 0.023376 | 0.026539 | 0.024953                  |
| monocarbaamide dihyd.| kg  | 0.219069 | 0.238553 | 0.214753                  |
| msma                | kg   | 0.221933 | 0.114146 | 0.042905                  |
| naled               | kg   | 0.00462 | 0.007498 | NA                        |
| norflurazon         | kg   | 0.031692 | 0.002861 | 0.002371                  |
| novaluron           | kg   | 0    | 0    | 0.002936                  |
| oxamyl              | kg   | 0.066709 | 0.013121 | 0.01513                   |
| oxyfluorfen         | kg   | 0.005359 | 0.001085 | 0.001807                  |
| paraquat            | kg   | 0.055529 | 0.063141 | 0.086601                  |
| pcnb                | kg   | 0.048785 | 0.023678 | 0.004968                  |
| pendimethalin       | kg   | 0.229879 | 0.178866 | 0.163831                  |
| permethrin          | kg   | 9.24E-05 | 0.000296 | 0.000113                  |
| petroleum distillate| kg   | NA   | 0.02466  | 0.37599                   |
| phorate             | kg   | 0.040561 | 0.030485 | 0.005081                  |
| profenofos          | kg   | 0.011457 | 0.014305 | **0.004087**              |
| prometryn           | kg   | 0.125935 | 0.115922 | 0.072262                  |
| propargite          | kg   | 0.005451 | 0.012727 | NA                        |
| pyraflufen-ethyl    | kg   | 0    | 0    | 0.000339                  |
| pyridate            | kg   | 0.00231 | NA   | 0                          |
| pyriproxyfen        | kg   | 0.000277 | 0.000493 | 0.000226                  |
| pyrithiobac-sodium  | kg   | 0.009701 | 0.012234 | 0.006436                  |
| quizalofop-ethyl    | kg   | 9.24E-05 | 0    | 0                          |
| sethoxydim          | kg   | 0.001478 | NA   | 0.000452                  |
| s-metolachlor       | kg   | 0.006745 | 0.031768 | 0.069778                  |
| sodium chlorate     | kg   | 0.142473 | 0.121644 | 0.157621                  |
| spinosad            | kg   | 0.008316 | 0.002072 | 0.000113                  |
| spirodicifen        | kg   | 0    | 0    | 0.000452                  |
| sulfosate           | kg   | NA   | 0.045876 | 0.016598                  |
| tebufenozide        | kg   | 0.002864 | 0.000691 | 0                          |
| thiamethoxam        | kg   | 0    | 0.004341 | 0.005081                  |
| thidiazuron         | kg   | 0.039083 | 0.033839 | 0.03105                   |
| thifensulfuron      | kg   | NA   | 0.000592 | 0                          |
| thiodicarb          | kg   | 0.001016 | NA   | 0                          |
| tralomethrin        | kg   | 0.000554 | 0.000592 | 0                          |
| tribufos            | kg   | 0.336965 | 0.2351  | 0.251448                  |
| trifloxysulfuron-sod| kg   | 0    | 0    | 0.000226                  |
| trifluralin         | kg   | 0.406446 | 0.41002  | 0.311967                  |
| zeta-cypermethrin   | kg   | 0.001848 | 0.003749 | 0.001016                  |
| unspecified         | kg   | 0.069758 | 0.079024 | 0.283966                  |
Table S4. Inputs use in U.S. soybean production (ha⁻¹)

| Energy          | 2002 | 2006 | 2012 |
|-----------------|------|------|------|
| diesel          | kg   | 32   | 27   | 22   |
| gasoline        | kg   | 9    | 8    | 6    |
| lp gas          | kg   | 2    | 2    | 1    |
| electricity     | kwh  | 19   | 16   | 13   |
| natural gas     | mj   | 135  | 114  | 94   |

| Nutrient        | 2002 | 2006 | 2012 |
|-----------------|------|------|------|
| nitrogen        | kg   | 5    | 3    | 5    |
| phosphate       | kg   | 14   | 12   | 20   |
| potash          | kg   | 29   | 23   | 34   |
| sulfur          | kg   | 0    | 0    | 1    |
| biofixed N      |      | 187  | 185  | 183  |

| Pesticide       | 2002 | 2006 | 2012 |
|-----------------|------|------|------|
| 2,4-d           | kg   | 0.021374 | NA | 0.000383 |
| 2,4-d, 2-ehe    | kg   | 0.005216 | 0.038899 | 0.062745 |
| 2,4-d, bee      | kg   | 0 | 0.001056 | 0.001041 |
| 2,4-d, dimeth. salt | kg | 0 | 0.014799 | 0.028019 |
| 2,4-db, dimeth. salt | kg | 4.77E-05 | NA | NA |
| acephate        | kg   | NA | 0.008479 | 0.015143 |
| acetochlor      | kg   | 0 | NA | 0.009723 |
| acifluorfen, sodium | kg | 0.005168 | 0.00073 | 0.003215 |
| alachlor        | kg   | 0.012754 | 0.007531 | NA |
| azoxystrobin    | kg   | 0.000398 | 0.001568 | 0.005696 |
| bentazon        | kg   | 0.018098 | 0.001087 | NA |
| beta-cyfluthrin | kg   | 0 | 0 | 6.12E-05 |
| bifenthrin      | kg   | 0 | 0 | 0.002343 |
| carbaryl        | kg   | 0.001161 | 0.001413 | NA |
| carfentrazozone-ethyl | kg | 1.59E-05 | 0.000155 | 1.53E-05 |
| chlorimuron-ethyl | kg | 0.001209 | 0.000807 | 0.002863 |
| chlorpyrifos    | kg   | 0.002974 | 0.025824 | 0.032 |
| clethodim       | kg   | 0.002306 | 0.00295 | 0.008023 |
| clomazone       | kg   | 0.001686 | 0 | NA |
| cloransulam-methyl | kg | 0.001129 | 0.000264 | 0.001271 |
| cyfluthrin      | kg   | 0 | 0.000155 | 0.000674 |
| cypermethrin    | kg   | NA | NA | 0.00153 |
| dicamba, digly. salt | kg | 0 | 0.000248 | 0.000276 |
| dicamba, dimet. salt | kg | 0 | NA | 0.001056 |
| diflubenzuron   | kg   | 7.95E-05 | 0.000155 | 9.19E-05 |
| dimethenamid-p  | kg   | 0 | NA | 0.003598 |
| dimethoate      | kg   | 0.000747 | 0 | 0.004226 |
| esfenvalerate   | kg   | 0.000175 | 0.001087 | 0.000153 |
| Chemical Name          | Unit   | Amount 1 | Amount 2 | Amount 3 | Amount 4 |
|-----------------------|--------|----------|----------|----------|----------|
| ethalfuralin          | kg     | 0.001097 | NA       | 0        |
| fenoxaprop            | kg     | 0.003419 | 0.00014  | 0        |
| fenoxaprop-p-ethyl    | kg     | 0        | 0        | 0.000107 |
| fluazifop-p-butyl     | kg     | 0.001702 | 0.000668 | 0.002986 |
| flubendiamide         | kg     | 0        | 0        | 0.000322 |
| flufenacet            | kg     | 0.00035  | 0.001242 | NA       |
| flumetsulam           | kg     | 0.000191 | 0.000124 | 0.000214 |
| flumiclorac-pentyl    | kg     | 0.000254 | 0.000264 | 0.000536 |
| flumioxazin           | kg     | 0        | 0        | 0.000153 |
| fluthiacet-methyl     | kg     | 0.008683 | 0.005124 | 0.020624 |
| gamma-cyhalothrin     | kg     | 0        | 4.66E-05 | 9.19E-05 |
| glufosinate-ammonium  | kg     | 0        | NA       | 0.019185 |
| glyphosate            | kg     | 0.953578 | 0.044116 | 0.100119 |
| glyphosate amm. salt  | kg     | 0        | 0.002205 | NA       |
| glyphosate dia. salt  | kg     | 0.06129  | 0        | 0        |
| glyphosate dim. salt  | kg     | 0        | 0        | 0.037068 |
| glyphosate iso. salt  | kg     | 0        | 1.380528 | 0.452442 |
| glyphosate pot. salt  | kg     | 0        | 0        | 1.084421 |
| imazamox              | kg     | 0.000747 | 0.00014  | 9.19E-05 |
| imazaquin             | kg     | 0.001304 | 0.001025 | 0.000352 |
| imazaquin, mon. salt  | kg     | 0        | 0        | 0.000168 |
| imazaquin, sod. salt  | kg     | 3.18E-05 | 0        | 0        |
| imazethapyr           | kg     | 0.005423 | 0.001553 | 0.003139 |
| imazethapyr, ammon.   | kg     | 0        | 7.76E-05 | 0.000245 |
| imidacloprid          | kg     | 0        | 0        | 0.000199 |
| lactofen              | kg     | 0.000954 | 0.000357 | 0.002944 |
| lambda-cyhalothrin    | kg     | 0.000493 | 0.001506 | 0.002159 |
| methoxyfenozide       | kg     | 0.000207 | 0.00014  | 0.001999 |
| methyl parathion      | kg     | 0.006075 | 0.001025 | NA       |
| metolachlor           | kg     | 0.009112 | 0        | 0.004471 |
| metribuzin            | kg     | 0.006997 | 0.006786 | 0.010335 |
| paraquat              | kg     | 0.009733 | 0.005202 | 0.012448 |
| pendimethalin         | kg     | 0.097517 | 0.029411 | 0.02387 |
| permethrin            | kg     | 0.000859 | 0.000186 | NA       |
| propiconazole         | kg     | 0        | 0.000435 | 0.001914 |
| pyraclostrobin        | kg     | 0        | 0.002919 | 0.006078 |
| quizalofop-p-ethyl    | kg     | 0.000398 | 0.000217 | 0.001807 |
| rimsulfuron           | kg     | 0        | NA       | 6.12E-05 |
| saflufenacil          | kg     | 0        | 0        | 0.001225 |
| sethoxydim            | kg     | 0.007315 | 0.000155 | 0.000965 |
| s-metolachlor         | kg     | 0.021549 | 0.012997 | 0.082542 |
| sulfentrazone         | kg     | 0.008445 | 0.001087 | 0.016505 |
| Chemical       | Unit | 2000     | 2004     | 2009   |
|----------------|------|----------|----------|--------|
| sulfosate      | kg   | 0.025047 | 0.015063 | 0      |
| tebuconazole   | kg   | 0        | 0.000559 | NA     |
| tetraconazole  | kg   | 0        | NA       | 0.00026|
| thiamethoxam   | kg   | 0        | 0        | 0.000291|
| thifensulfuron | kg   | 6.36E-05 | 4.66E-05 | 0.000475|
| thiodicarb     | kg   | 0.001081 | 0.000606 | NA     |
| tribenuron-methyl| kg  | 3.18E-05 | 7.76E-05 | 0.000153|
| trifloxystrobin| kg   | 0        | 0.000109 | 0.001118|
| trifluralin    | kg   | 0.069735 | 0.022578 | 0.019996|
| zeta-cypermethrin| kg  | 0.00027  | 0.000217 | 6.12E-05|
| unspecified    | kg   | 0.019529 | 0.007531 | 0.011652|

Table S5. Inputs use in U.S. wheat production (ha⁻¹)

| Energy        | 2000 | 2004 | 2009 |
|---------------|------|------|------|
| diesel        | 34   | 29   | 25   |
| gasoline      | 7    | 6    | 5    |
| lp gas        | 1    | 1    | 1    |
| electricity   | 37   | 32   | 27   |
| natural gas   | 0    | 0    | 0    |

| Nutrient      | 2000 | 2004 | 2009 |
|---------------|------|------|------|
| nitrogen      | 81   | 103  | 75   |
| phosphate     | 27   | 37   | 25   |
| potash        | 10   | 11   | 7    |
| sulfur        | 0    | 0    | 2    |

| Pesticide     | 2000     | 2004     | 2009     |
|---------------|----------|----------|----------|
| 2,4-d         | 0.124277  | 0.097425 | NA       |
| 2,4-d, 2-ehe  | 0         | 0.012619 | 0.121297 |
| 2,4-d, bee    | 0         | 0.012169 | NA       |
| 2,4-d, dieth. salt | 0     | 0        | 0.000402 |
| 2,4-d, dimeth. salt | NA  | 0.005233 | 0.069916 |
| 2,4-d, isoprop. salt | 0    | 0        | 0.000981 |
| 2,4-dp, dimeth. salt | NA   | 0.026791 | 0        |
| atrazine      | NA       | 0.005584 | NA       |
| azoxystrobin  | 0        | 0.000325 | 0.000704 |
| beta-cyfluthrin| 0       | 0        | 7.54E-05 |
| bromoxynil    | 0.024448 | 0.017402 | 0.000151 |
| bromoxynil heptan. | 0    | 0        | 0.021704 |
| bromoxynil octanoate | 0   | 0.024738 | 0.047633 |
| carfentrazone-ethyl | 0   | 2.5E-05  | 5.03E-05 |
| chlorpyrifos  | 0.012248 | 0.011192 | 0.014511 |
| chlorsulfuron | 0.000946 | 0.000801 | 0.000679 |
| Chemical                  | Unit 1   | Unit 2   | Unit 3   |
|--------------------------|----------|----------|----------|
| clodinafop-propargil     | NA       | 0.003856 | 0.00161  |
| clopyralid               | 0.004341 | 0.001953 | 0.007972 |
| cyfluthrin               | 0        | 0        | 5.03E-05 |
| dicamba                  | 0.014383 | 0.012669 | 0.000553 |
| dicamba, dimet. salt     | NA       | NA       | 0.011971 |
| dicamba, sodium salt     | 0        | 0.0002  | 0.002465 |
| diclofop-methyl          | 0.004123 | 0.002228 | NA       |
| dimethoate               | 0.000582 | 0.000701 | 0.002163 |
| diuron                   | 0.000437 | 0.000501 | 0     |
| ethyl parathion          | 0        | 0.00333 | 0       |
| fenoxaprop               | 0.007713 | 0.010191 | 0       |
| fenoxaprop-p-ethyl       | 0.000146 | 0        | 0.007972 |
| flucarbazone-sodium      | 0        | 0.000501 | 0.000629 |
| flufenacet               | 0        | 0.000225 | NA      |
| fluroxypyr               | 0.001965 | 0.003731 | 0.001811 |
| fluroxypyr 1-mhe         | 0        | 0.002404 | 0.011619 |
| glyphosate               | 0.086635 | 0.154412 | 0.017932 |
| glyphosate iso. salt     | 0        | 0        | 0.420578 |
| glyphosate pot. salt     | 0        | 0        | 0.006514 |
| imazamethabenz           | 0.002159 | 0.0001  | NA      |
| imazamox                 | 0        | 0.000225 | 0.000428 |
| lambda-cyhalothrin       | 4.85E-05 | 0.00015 | 0.000201 |
| mcpa                     | 0.066334 | 0.07474 | 0.000679 |
| mcpa, 2-ethylhexyl       | 0        | 0.002629 | 0.044993 |
| mcpa, dimethyl. salt     | 0        | 0.004181 | 0.007067 |
| mcpa, isooctyl ester     | 0        | 0        | 0.006539 |
| mesosulfuron-methyl      | 2.43E-05 | 7.51E-05 | 0.000176 |
| methyl parathion         | NA       | NA       | 0.002238 |
| methanone                | 0        | 0        | 0.002364 |
| metribuzin               | 0.001868 | 0.00338 | 0.000629 |
| metsulfuron-methyl       | 0.000315 | 0.000325 | 0.000428 |
| picloram, k salt         | 0.000146 | 7.51E-05 | NA      |
| pinoxaden                | 0        | 0        | 0.003295 |
| propiconazole            | 0.000606 | 0.000876 | 0.007721 |
| propoxycarbazone-sod     | 0        | 0        | 0.000679 |
| prosulfuron              | 7.28E-05 | 5.01E-05 | 0.000101 |
| prothioconazole          | 0        | 0.002178 | 0.002842 |
| pyraclostrobin           | 0        | 0.001753 | 0.003622 |
| pyroxsulam               | 0        | 0        | 0.000327 |
| sulfosulfuron            | 0.000121 | 0.000826 | 0.000226 |
| tebuconazole             | 0.001746 | 0.004432 | 0.005508 |
| thifensulfuron           | 0.001019 | 0.001402 | 0.001937 |
| tralkoxydim              | 0.005627 | 0.000476 | NA      |
| Compound               | 2002   | 2007   | 2012   |
|------------------------|--------|--------|--------|
| triallate              | 0.049332 | 0.006159 | NA     |
| triasulfuron           | 0.000873 | 0.000676 | 0.000503 |
| tribenuron-methyl      | 0.011188 | 0.000776 | 0.000931 |
| trifloxystrobin        | 0      | 0.000225 | 0.00083 |
| trifluralin            | 0.019549 | 0.005834 | 0.000377 |
| zeta-cypermethrin      | 0      | 0.00025  | NA     |
| unspecified            | 0.012806 | 0.016626 | 0.016121 |

Table S6. Average irrigation water use per ha crop produced in the U.S.

| Year | Corn | Cotton | Soybean | Wheat |
|------|------|--------|---------|-------|
| 2002 | 514  | 1400   | 180     | 326   |
| 2007 | 422  | 1180   | 234     | 344   |
| 2012 | 510  | 1256   | 267     | 280   |

Noted: Results are calculated based on \((\text{irrigation intensity for irrigated area} \times \text{area irrigated}) / \text{total area harvested}\).
3. Data on emission factors

Table S7. Nitrogen (N) and Phosphorus (P) runoff and leaching rates (% of the amount applied)

|         | Corn |       | Soybean |       | Wheat |       |
|---------|------|-------|---------|-------|-------|-------|
|         | N    | P     |         | N    | P     |
| CO      | 13   | 9     | AR      | 17   | 33    |
| GA      | 47   | 13    | IL      | 9    | 43    |
| IL      | 21   | 15    | IN      | 9    | 43    |
| IN      | 21   | 15    | IA      | 9    | 43    |
| IA      | 21   | 15    | KS      | 11   | 35    |
| KS      | 18   | 12    | KY      | 23   | 20    |
| KY      | 38   | 17    | LA      | 17   | 33    |
| MI      | 21   | 15    | MI      | 9    | 43    |
| MN      | 21   | 15    | MN      | 9    | 43    |
| MO      | 25   | 18    | MS      | 17   | 33    |
| NE      | 13   | 9     | MO      | 17   | 33    |
| NY      | 33   | 22    | NE      | 8    | 29    |
| NC      | 47   | 13    | NC      | 30   | 8     |
| ND      | 13   | 9     | ND      | 8    | 29    |
| OH      | 27   | 18    | OH      | 13   | 28    |
| PA      | 33   | 22    | SD      | 8    | 29    |
| SD      | 13   | 9     | TN      | 17   | 33    |
| TX      | 19   | 13    | VA      | 30   | 8     |
| WI      | 21   | 15    | WI      | 9    | 43    |

Note: Nutrient loss rates for each state are derived from regional (multistate) nutrient loss rates according to the location of the state relative to that of the region, mostly multi-states, modeled in the reference. If a state occupies parts of several regions, the average of the regional rates is used.
### Table S8. Speciated VOC emissions from natural gas- and diesel-engine farm machinery

| Substances                              | Natural Gas (kg/Mj) | Diesel kg/kg |
|-----------------------------------------|---------------------|--------------|
|                                         | 2-stroke lean-burn  | 4-stroke lean-burn | 4-stroke rich-burn |
| 1,1,2,2-Tetrachloroethane               | 2.9E-08             | 1.7E-08       | 1.1E-08            | 0.0E+00 |
| 1,1,2-Trichloroethane                  | 2.3E-08             | 1.4E-08       | 6.6E-09            | 0.0E+00 |
| 1,1-Dichloroethane                     | 1.7E-08             | 1.0E-08       | 4.9E-09            | 0.0E+00 |
| 1,2,3-Trimethylbenzene                 | 1.5E-08             | 9.9E-09       | 0.0E+00            | 0.0E+00 |
| 1,2,4-Trimethylbenzene                 | 4.8E-08             | 6.2E-09       | 0.0E+00            | 0.0E+00 |
| 1,2-Dichloroethane                     | 1.8E-08             | 1.0E-08       | 4.9E-09            | 0.0E+00 |
| 1,2-Dichloropropane                    | 1.9E-08             | 1.2E-08       | 5.6E-09            | 0.0E+00 |
| 1,3,5-Trimethylbenzene                 | 7.7E-09             | 1.5E-08       | 0.0E+00            | 0.0E+00 |
| 1,3-Butadiene                          | 3.5E-07             | 1.2E-07       | 2.9E-07            | 7.2E-07 |
| 1,3-Dichloropropene                    | 1.9E-08             | 1.1E-08       | 5.5E-09            | 0.0E+00 |
| 2,2,4-Trimethylpentane                 | 3.6E-07             | 1.1E-07       | 0.0E+00            | 0.0E+00 |
| 2-Methylnaphthalene                    | 9.2E-09             | 1.4E-08       | 0.0E+00            | 0.0E+00 |
| Acenaphthene                            | 5.7E-10             | 5.4E-10       | 0.0E+00            | 2.6E-08 |
| Acenaphthylene                          | 1.4E-09             | 2.4E-09       | 0.0E+00            | 9.3E-08 |
| Acetaldehyde                            | 3.3E-06             | 3.6E-06       | 1.2E-06            | 1.4E-05 |
| Acrolein                               | 3.3E-06             | 2.2E-06       | 1.1E-06            | 1.7E-06 |
| Anthracene                             | 3.1E-10             | 0.0E+00       | 0.0E+00            | 3.4E-08 |
| Benz(a)anthracene                      | 1.4E-10             | 0.0E+00       | 0.0E+00            | 3.1E-08 |
| Benzene                                | 8.3E-07             | 1.9E-07       | 6.8E-07            | 1.7E-05 |
| Benzo(a)pyrene                          | 2.4E-12             | 0.0E+00       | 0.0E+00            | 0.0E+00 |
| Benzo(b)fluoranthene                   | 3.7E-12             | 7.1E-11       | 0.0E+00            | 1.8E-09 |
| Benzo(e)pyrene                          | 1.0E-11             | 1.8E-10       | 0.0E+00            | 3.5E-09 |
| Benzo(g,h,i)pyrrole                    | 1.1E-11             | 1.8E-10       | 0.0E+00            | 9.0E-09 |
| Benzo(k)fluoranthene                    | 1.8E-12             | 0.0E+00       | 0.0E+00            | 2.9E-09 |
| Biphenyl                               | 1.7E-09             | 9.1E-08       | 0.0E+00            | 0.0E+00 |
| Butane                                 | 2.0E-06             | 2.3E-07       | 0.0E+00            | 0.0E+00 |
| Butyraldehyde                           | 1.9E-07             | 4.3E-08       | 2.1E-08            | 0.0E+00 |
| Carbon Tetrachloride                    | 2.6E-08             | 1.6E-08       | 7.6E-09            | 0.0E+00 |
| Chlorobenzene                           | 1.9E-08             | 1.3E-08       | 5.6E-09            | 0.0E+00 |
| Chloroethane                            | 0.0E+00             | 8.0E-10       | 0.0E+00            | 0.0E+00 |
| Chloroform                              | 2.0E-08             | 1.2E-08       | 5.9E-09            | 0.0E+00 |
| Chrysene                                | 2.9E-10             | 3.0E-10       | 0.0E+00            | 6.5E-09 |
| Cyclohexane                             | 1.3E-07             | 9.8E-08       | 0.0E+00            | 0.0E+00 |
| Cyclopentane                            | 4.1E-08             | 0.0E+00       | 0.0E+00            | 0.0E+00 |
| Dibenz(a,h)anthracene                   | 0.0E+00             | 0.0E+00       | 0.0E+00            | 1.1E-08 |
| Ethane                                  | 3.1E-05             | 4.5E-05       | 3.0E-05            | 0.0E+00 |
| Ethylbenzene                            | 4.6E-08             | 1.7E-08       | 1.1E-08            | 0.0E+00 |
| Ethylene Dibromide                      | 3.2E-08             | 1.9E-08       | 9.2E-09            | 0.0E+00 |
| Compound                  | Diesel        | Gasoline      | LPG           | NG             |
|---------------------------|---------------|---------------|---------------|----------------|
|                           | Farming Tractor | Farming Tractor | Commercial Boiler | Stationary Reciprocating Engine |
| Fluoranthene              | 1.6E-10       | 4.8E-10       | 0.0E+00       | 1.4E-07        |
| Fluorene                  | 7.3E-10       | 2.4E-09       | 0.0E+00       | 5.4E-07        |
| Formaldehyde              | 2.4E-05       | 2.3E-05       | 8.8E-06       | 2.2E-05        |
| Indeno(1,2,3-c,d)pyrene    | 4.3E-12       | 0.0E+00       | 0.0E+00       | 6.9E-09        |
| Isobutane                 | 1.6E-06       | 0.0E+00       | 0.0E+00       | 0.0E+00        |
| Methanol                  | 1.1E-06       | 1.1E-06       | 1.3E-06       | 0.0E+00        |
| Methylcyclohexane         | 1.5E-07       | 5.3E-07       | 0.0E+00       | 0.0E+00        |
| Methylene Chloride        | 6.3E-08       | 8.6E-09       | 1.8E-08       | 0.0E+00        |
| n-Hexane                  | 1.9E-07       | 4.8E-07       | 0.0E+00       | 0.0E+00        |
| n-Nonane                  | 1.3E-08       | 4.7E-08       | 0.0E+00       | 0.0E+00        |
| n-Octane                  | 3.2E-08       | 1.5E-07       | 0.0E+00       | 0.0E+00        |
| n-Pentane                 | 6.6E-07       | 1.1E-06       | 0.0E+00       | 0.0E+00        |
| Naphthalene               | 4.1E-08       | 3.2E-08       | 4.2E-08       | 1.6E-06        |
| Perylene                  | 2.1E-12       | 0.0E+00       | 0.0E+00       | 0.0E+00        |
| Phenanthrene              | 1.5E-09       | 4.5E-09       | 0.0E+00       | 5.4E-07        |
| Phenol                    | 1.8E-08       | 1.0E-08       | 0.0E+00       | 0.0E+00        |
| Propane                   | 1.2E-05       | 1.8E-05       | 0.0E+00       | 0.0E+00        |
| Propylene                 | 0.0E+00       | 0.0E+00       | 0.0E+00       | 4.8E-05        |
| Pyrene                    | 2.5E-10       | 5.9E-10       | 0.0E+00       | 8.8E-08        |
| Styrene                   | 2.4E-08       | 1.0E-08       | 5.1E-09       | 0.0E+00        |
| Tetrachloroethane         | 0.0E+00       | 1.1E-09       | 0.0E+00       | 0.0E+00        |
| Toluene                   | 4.1E-07       | 1.8E-07       | 2.4E-07       | 7.5E-06        |
| Vinyl Chloride            | 1.1E-08       | 6.4E-09       | 3.1E-09       | 0.0E+00        |
| Xylene                    | 1.2E-07       | 7.9E-08       | 8.4E-08       | 5.2E-06        |

Note: For natural gas, average emission factors for different modes are used for analysis in this study.

Table S9. Emission factors for fuel combustion (mg MJ⁻¹)

| Compound  | Diesel Farming Tractor | Diesel Farming Tractor | Diesel Commercial Boiler | Diesel Stationary Reciprocating Engine |
|-----------|------------------------|------------------------|--------------------------|--------------------------------------|
| CO        | 344.2                  | 516.4                  | 10.2                     | 324                                  |
| NOx       | 649.3                  | 454                    | 80.6                     | 1137                                 |
| PM10      | 58.8                   | 23.7                   | 2.3                      | 5.2                                  |
| PM2.5     | 52.8                   | 21.7                   | 2.3                      | 5.2                                  |
| SOx       | 7.6                    | 1.1                    | 0                        | 0.3                                  |
Table S10. Estimation of pesticide releases into different compartments

| Compartment | Fraction | Note                                      |
|-------------|----------|-------------------------------------------|
| Air         | 95%      | p(vapor pressure) >10 (mPa)               |
|             | 50%      | 1<p<10                                    |
|             | 15%      | 0.1<p<1                                   |
|             | 5%       | 0.01<p<0.1                                |
|             | 1%       | p<0.01                                    |
| Water       | 0.5%     | Generic runoff and leaching rate          |
| Soil        | 1-air-water | Capped at 85%                            |

Table S11. Average heavy metal concentrations in U.S. phosphate rock and fertilizer

|                | As | Cd | Cr<sup>b</sup> | Pb | Hg | Ni | V   |
|----------------|----|----|----------------|----|----|----|-----|
| Phosphate rock | 12 | 11 | 109            | 12 | 0.05 | 37 | 82  |
| P2O5 fertilizers | 34 | 32 | 310            | 35 | 0.1 | 105 | 235 |

Note: Cr is assumed to exist in the form of Cr (III) in agricultural soil, because Cr (VI), being a strong oxidizing agent, is likely to be reduced to Cr (III) through reaction with soil organic matter or other reducing agents. For 1 kg As emitted, 0.5 kg is assumed to exist in As (III) and the other half in As (V) due to a lack of more precise insights into the issue.

Table S12. Emission factors for NH<sub>3</sub> and NO<sub>X</sub> from nitrogen

|                | NH<sub>3</sub> | NO<sub>X</sub> |
|----------------|---------------|----------------|
| Emission factor | 3.5%          | 2.5%           |
4. Estimation of biological nitrogen fixation for soybean

Nitrogen input through biological nitrogen fixation (BNF), due to nitrogen-fixing bacteria hosted symbiotically by the legume crops, can also be a source of nitrogen pollution\(^1\). Here, we estimated the total amount of BNF for soybean production in the major states covered, based on a USDA study that quantified nutrient inputs from multiple sources to major U.S. crops in a single year.\(^1\) Their estimates of BNF ranged from 91 to 243 kg ha\(^{-1}\) for 7 geographic regions into which the study classified the continental United States. We first derived state-level estimates from the regional estimates based on the location of the state relative to the regions classified in the USDA study. We assumed that the rates remained unchanged over the period investigated given that there is no strong evidence that the environmental factors\(^2\) affecting BNF changed significantly in the past decade or. Based on the state rates, we further calculated the average rate for per ha soybean in the U.S., which is about 185 kg ha\(^{-1}\). The total amount of BNF calculated for the 19 to 20 states major soybean-producing states covered in this study is around 5.4 Tg yr\(^{-1}\), which is close to a recent estimate of 5.7 Tg calculated for the entire country.\(^3\)

5. Results

Table S13. Life-cycle, cradle-to-gate impact per ha crop produced (TRACI 2.0)

|       | Corn  |       |       | Cotton |       |       |
|-------|-------|-------|-------|--------|-------|-------|
|       | 2001  | 2005  | 2010  | 2000   | 2003  | 2007  |
| ACD   | moles of H+ eq | 1584 | 1602 | 1630 | 1262 | 1185 | 1155 |
| EUT   | kg N eq | 64.4 | 66.7 | 66.3 | 72.8 | 71.9 | 68.4 |
| SF    | kg O3 eq | 416  | 417  | 427  | 383  | 352  | 332  |
| HHR   | kg PM10 eq | 2.7  | 2.7  | 2.7  | 3.3  | 2.9  | 2.6  |
| FET   | CTUe  | 32844 | 24219 | 17331 | 72935 | 48146 | 28830 |
| HHC   | CTUh  | 5.3E-05 | 5.3E-05 | 5.3E-05 | 4.1E-05 | 3.8E-05 | 3.6E-05 |
| HHNC  | CTUh  | 4.4E-04 | 4.4E-04 | 4.4E-04 | 3.8E-04 | 4.1E-04 | 3.8E-04 |

|       | Soybean |       |       | Wheat  |       |       |
|-------|---------|-------|-------|--------|-------|-------|
|       | 2002    | 2006  | 2012  | 2000   | 2004  | 2009  |
| ACD   | moles of H+ eq | 1307 | 1270 | 1280 | 802  | 995  | 728  |
| EUT   | kg N eq | 41.6  | 38.1  | 48.6  | 20.7  | 27.5  | 18.7  |
| SF    | kg O3 eq | 298  | 287  | 285  | 205  | 248  | 182  |
| HHR   | kg PM10 eq | 0.9  | 0.8  | 0.9  | 1.4  | 1.6  | 1.2  |
| FET   | CTUe  | 2154  | 5281  | 8790  | 2153  | 2958  | 3050  |
| HHC   | CTUh  | 6.7E-06 | 5.4E-06 | 7.8E-06 | 2.5E-05 | 3.1E-05 | 2.3E-05 |
| HHNC  | CTUh  | 1.0E-04 | 8.8E-05 | 1.5E-04 | 2.1E-04 | 2.8E-04 | 2.0E-04 |
6. Comparison between TRACI 2.0, IMPACT 2002+, and CML 2001

To test the robustness of our result on freshwater ecotoxicity impact, we applied two additional characterization models, IMPACT 2002+ and CML 2001, to evaluating the inventory data compiled. Because pesticide releases were identified as the major contributor to freshwater ecotoxicity (see figure 3 in the manuscript), the other two models were used to characterize pesticide releases only. The impact categories in IMPACT 2002+ and CML 2001 that correspond to freshwater ecotoxicity in TRACI 2.0 are aquatic ecotoxicity. Estimation of pesticide releases to different compartments was kept the same for all three methods (see section 2.4. in the manuscript).

Results are presented below. Two factors lead to the differences in the magnitude of change calculated by the three characterization models (see also figure 5 in the manuscript). First, different models cover different numbers of pesticides. TRACI 2.0, whose freshwater ecotoxicity impact category is adopted from USEtox,4 covers the largest number of pesticides, with 63/84 for corn, 89/113 for cotton, 55/81 for soybean, and 48/65 for wheat. Compare this with CML 2001 (19/84 for corn, 23/113 for cotton, 16/81 for soybean, and 13/65 for wheat) and IMPACT 2002+ (63/84 for corn, 89/113 for cotton, 55/81 for soybean, and 48/65 for wheat). Second, different characterization models give different ecotoxicity potentials to different pesticides. In TRACI 2.0, for example, cyfluthrin shows the largest freshwater ecotoxicity potential, which is 300 times that of atrazine. In IMPACT 2002+, however, lambda-cyhalothrin shows the largest aquatic ecotoxicity potential, and that of cyfluthrin is only 17 times that of atrazine.

Tables S14-S17 present impact results based on CML 2001 and contributions of key pesticides identified.

Table S14. Aquatic ecotoxicity impact per ha corn produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

|          | 2001 | 2005 | 2010 |
|----------|------|------|------|
| atrazine | 22%  | 22%  | 25%  |
| chlorpyrifos | 14%  | 9%   | 3%   |
| metolachlor | 15%  | 4%   | 1%   |
| permethrin | 7%   | 4%   | 3%   |
| simazine | 4%   | 6%   | 7%   |
| s-metolachlor | 36%  | 51%  | 60%  |
| other pesticides | 2%   | 4%   | 1%   |
| **total** | **1599** | **1348** | **993** |

Table S15. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

|          | 2000 | 2003 | 2007 |
|----------|------|------|------|
| aldicarb | 66%  | 78%  | 83%  |
| cypermethrin | 6%   | 9%   | 10%  |
| malathion | 23%  | 7%   | 3%   |
| other pesticides | 5%   | 6%   | 4%   |
| **total** | **25935** | **19058** | **12729** |
Table S16. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

|                  | 2002 | 2006 | 2012 |
|------------------|------|------|------|
| chlorpyrifos     | 11%  | 71%  | 50%  |
| metolachlor      | 10%  | 0%   | 2%   |
| permethrin       | 7%   | 1%   | 0%   |
| s-metolachlor    | 23%  | 10%  | 38%  |
| trifluralin      | 25%  | 6%   | 3%   |
| zeta-cypermethrin| 16%  | 10%  | 2%   |
| other pesticides | 9%   | 1%   | 6%   |
| total            | **347** | **456** | **804** |

Table S17. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg 1,4-dichlorobenzene eq.)

|                  | 2002 | 2006 | 2012 |
|------------------|------|------|------|
| chlorpyrifos     | 66%  | 33%  | 68%  |
| ethyl parathion  | 0%   | 18%  | 0%   |
| methanone        | 0%   | 0%   | 8%   |
| triallate        | 22%  | 1%   | 1%   |
| trifluralin      | 5%   | 1%   | 0%   |
| zeta-cypermethrin| 0%   | 42%  | 23%  |
| other pesticides | 7%   | 5%   | 0%   |
| total            | **67** | **125** | **78** |

Tables S18-21 present impact results based on IMPACT 2002 + and contributions of key pesticides identified.

Table S18. Aquatic ecotoxicity impact per ha corn produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

|        | 2001 | 2005 | 2010 |
|--------|------|------|------|
| atrazine | 98%  | 98%  | 99%  |
| other pesticides | 2%   | 2%   | 1%   |
| total   | **962290** | **808538** | **667197** |
Table S19. Aquatic ecotoxicity impact per ha cotton produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

|                | 2000 | 2003 | 2007 |
|----------------|------|------|------|
| aldicarb       | 18%  | 21%  | 21%  |
| chlorpyrifos   | 5%   | 2%   | 0%   |
| cyanazine      | 4%   | 0%   | 0%   |
| cyfluthrin     | 4%   | 3%   | 3%   |
| diuron         | 5%   | 10%  | 13%  |
| lambda-cyhalothrin | 4% | 4% | 4% |
| malathion      | 3%   | 1%   | 0%   |
| methomyl       | 4%   | 3%   | 0%   |
| prometryn      | 17%  | 21%  | 19%  |
| tribufos       | 17%  | 16%  | 24%  |
| trifluralin    | 4%   | 5%   | 5%   |
| other pesticides | 16% | 13% | 10% |
| **total**      | 69240| 51911| 36533|

Table S20. Aquatic ecotoxicity impact per ha soybean produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

|                | 2002 | 2006 | 2012 |
|----------------|------|------|------|
| chlorpyrifos   | 5%   | 42%  | 30%  |
| glyphosate     | 32%  | 1%   | 2%   |
| lambda-cyhalothrin | 9% | 28% | 23% |
| metribuzin     | 13%  | 13%  | 11%  |
| pendimethalin  | 7%   | 2%   | 1%   |
| s-metolachlor  | 9%   | 5%   | 19%  |
| trifluralin    | 13%  | 4%   | 2%   |
| other pesticides | 13% | 4% | 12% |
| **total**      | 3273 | 3250 | 5658 |

Table S21. Aquatic ecotoxicity impact per ha wheat produced and contributions of key pesticides (unit: kg triethylene glycol. eq.)

|                | 2002 | 2006 | 2012 |
|----------------|------|------|------|
| atrazine       | 47%  | 71%  | 60%  |
| bromoxynil     | 19%  | 8%   | 0%   |
| chlorpyrifos   | 16%  | 9%   | 23%  |
| triallate      | 7%   | 1%   | 0%   |
| other inputs   | 12%  | 11%  | 17%  |
| **total**      | 4173 | 6890 | 3307 |
References

1. USDA. Model Simulation of Soil Loss, Nutrient Loss, and Change in Soil Organic Carbon Associated with Crop Production. Natural Resource Conservation Service, US Department of Agriculture; 2006.

2. Zahran HH. Rhizobium-Legume Symbiosis and Nitrogen Fixation under Severe Conditions and in an Arid Climate. Microbiol Mol Biol Rev. 1999 Dec 1;63(4):968–89.

3. Herridge DF, Peoples MB, Boddey RM. Global inputs of biological nitrogen fixation in agricultural systems. Plant Soil. 2008;311(1-2):1–18.

4. Rosenbaum RK, Bachmann TM, Gold LS, Huijbregts MAJ, Jolliet O, Juraske R, et al. 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 Nov 1;13(7):532–46.