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Tsalidis, Georgios Archimidis

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Human Health and Ecosystem Quality Benefits with Life Cycle Assessment Due to Fungicides Elimination in Agriculture

Georgios Archimidis Tsalidis 1,20

- Biotechnology Department, Applied Sciences Faculty, Delft University of Technology, 2629 HZ Delft, The Netherlands; g.a.tsalidis@tudelft.nl or gatsalidis@athenarc.gr
- Athena—Research and Innovation Center in Information, Communication and Knowledge Technologies, 67100 Xanthi, Greece

Abstract: Industrial agriculture results in environmental burdens due to the overuse of fertilizers and pesticides. Fungicides is a class of pesticides whose application contributes (among others) to human toxicity and ecotoxicity. The European Union aims to increase organic agriculture. For this reason, this work aims to analyze climate change, freshwater ecotoxicity, terrestrial ecotoxicity, human toxicity, (terrestrial) acidification, and freshwater eutrophication impacts of fungicides and calculate expected benefits to human health (per European citizen) and ecosystem quality (terrestrial) with life cycle assessment (LCA) during crop production. The Scopus database was searched for LCA studies that considered the application of fungicides to specific crops. The analysis shows how many systemic and contact fungicides were considered by LCA studies and what was the applied dosage. Furthermore, it shows that fungicides highly contribute to freshwater ecotoxicity, terrestrial ecotoxicity, human toxicity, and freshwater eutrophication for fruits and vegetables, but to a low extent compared to all considered environmental impacts in the case of cereals and rapeseed. Expected benefits to human health and ecosystem quality after fungicides elimination are greater for fruits and vegetables, ranging between 0 to 47 min per European citizen in a year and 0 to 90 species per year, respectively.

Keywords: conventional agriculture; azoxystrobin; mancozeb; disability adjusted life year; timeintegrated species loss; toxicity; vegetables; fruit; cereals

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1. Introduction

The most environmentally damaging form of human consumption is eating [1] because industrial agriculture results in environmental burdens due to the overuse of machinery, fertilizers, insecticides, herbicides, and fungicides [2]. The latter are toxic to humans and the environment [3]. This study reviews the contribution to toxicity of fungicides in fruits, vegetables, cereals, and rapeseed production to show the benefits for human health and ecosystem quality after their elimination.

The primary goals of agriculture are the production of nutritional food for humans and feed for animals, and the increased economic development of stakeholders. In addition, the UN 2030 Agenda on Sustainable Development also identified bioeconomy as being in line with goals for energy efficiency (Goal 7) [4]. These goals resulted in a significant growth of the bioeconomy, and the number of publications about bioeconomy increased from 1000 in 2017 to approximately 3500 in 2021 [5]. Furthermore, during the period 2021 to 2030, global agricultural production is projected to increase by 1.4% per year [6]. Maize, rice, wheat, oilseeds, and oil products are major commodities globally [6]. Due to the introduction of synthetic pesticides between 1960 and 1989, pest control and agricultural output have significantly improved [7].

Intensive, industrial agricultural practices employ large quantities of inputs, such as fuels, electricity, water for irrigation, fertilizers, and pesticides. Such intensive agricultural Sustainability **2022**, 14, 846 2 of 12

practices are held responsible for increased energy use and accompanied elevated emissions of greenhouse gases [8], and adverse effects on human health and ecosystem quality [3]. In contrast, in organic agricultural practice, the use of fertilizers is significantly reduced while the consumption of pesticides is avoided [9].

The term pesticide covers a wide range of compounds, including insecticides, fungicides, herbicides, etc. The benefits of pesticides are the consequences of their effects: increased productivity, protection of crop losses, vector disease control, and quality of food [7]. These effects occur due to active ingredients in pesticides, which are chemicals that kill, control, or repel pests. The World Health Organization [10] and a study [11] focusing on toxicity stated that insecticides tend to be more toxic to humans than fungicides and herbicides by one and four orders of magnitude, respectively. However, three active fungicides ingredients—(1) tebuconazole, (2) epoxiconazole, and (3) prochloraz—were more toxic than herbicides and insecticides to mitochondrial activities, membrane degradations, and caspases 3/7 activities [12].

Fungicides are the class of pesticides that target fungi. Fungicides are classified as systemic and contact. Systemic fungicide can be defined as a fungicide that, when sprayed on top surfaces of the plant, is translocated to the plant tissues, transferring its toxicity to the targeted fungus [13]. Examples of systemic fungicides are Benomyl, Crypoconazol, and Imazalil. Contact fungicides remain on the plant surface after application and do not penetrate plant tissues. These fungicides are typically employed to control foliar diseases. A contact fungicide must be present on the plant surface before the disease penetrates the tissue [14]. Examples of contact fungicides are Mancozeb and Thiram. Pre-harvest fungicide use is greatly limited or absent in organic agroecosystems [15]. According to studies [16–18], approximately 55% of applied fungicide is found in air, freshwater, and soil while the rest remains in or on the crop, depending on fungicide class.

Stakeholders from corporate, academic, civil society, and governmental sectors highlighted a shift to organic agriculture as a sustainable solution to the negative effects of industrial agriculture [19]. In particular, European Union (EU) strategies regarding agricultural development encourage eco-friendly practices [20]. Europe's Farm to Fork (F2F) strategy aims to boost the development of its organic farming area to 25% of total farmland by 2030. However, a recent study [21] concluded that increasing organic cultivation with the F2F strategy may result in less sustainable food systems because EU law strictly prohibits the use of genetically modified organisms in organic cultivation, which can result in failing to achieve hunger reduction (Sustainable Development Goal 2).

Life cycle assessment (LCA) is a tool to assess environmental sustainability during the entire life cycle of a product and it is standardized by ISO 14040 [22] and 14044 [23]. Energy and material flows, and environmental releases are quantified and converted to environmental impacts during the life cycle inventory and life cycle impact assessment phases, respectively. Environmental impacts exist at midpoint and endpoint levels. Midpoint indicators focus on single environmental impacts, such as climate change. The ReCiPe is an environmental impact model which calculates midpoint indicators and converts them into endpoint indicators based on an individualist, hierarchist, or egalitarian perspective. ReCiPe's endpoint indicators are human health, ecosystem quality, and resource scarcity [24].

A separate impact assessment modeling step is required for fungicides because reference life cycle inventory datasets of fungicides reflect only production-related impacts, not impacts that occur after application [3]. This review aims to analyze the environmental impacts of fungicides due to their production and application, and calculate expected benefits to human health and ecosystem quality due to their elimination based on ReCiPe's midpoint to endpoint conversion factors.

2. Materials and Methods

This study considered 39 papers. They are the results of searching in the Scopus database with certain combinations of keywords with the "AND" Boolean operator. Figure 1 shows all the steps followed to identify the considered LCA studies. To summarize,

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considered studies needed to apply LCA in agricultural products, mention fungicides or at least include them in the class of pesticides during the contribution analysis of life cycle interpretation stage, and calculate toxicity. In 2009, van Zelm et al. [25] published their work which calculated freshwater ecotoxicological effect factors for 397 pesticides. Therefore, we considered this study the starting point for this research. All considered studies can be found in the Supplementary Materials.

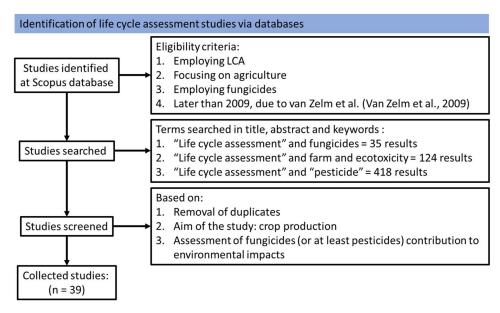


Figure 1. Steps followed for review and inclusion/exclusion criteria.

Analysis of reviewed midpoint impact values to design box plots was performed with MS Excel. Furthermore, outliers were analyzed with Grubbs' test with a significance level of 0.05 (two-sided) in the GraphPad Outlier calculator.

Midpoint impacts were converted to endpoint damages to human health and ecosystem quality (terrestrial) with the ReCiPe environmental impact model [26]. Midpoint to endpoint conversion factors were used to convert average midpoint results (per type of crop) to human health and ecosystem quality (terrestrial) endpoints based on the individualist, hierarchist, and egalitarian perspectives, and show the effect of fungicide elimination on a European level. The following formula was used:

where i stands for endpoint human health or ecosystem quality (terrestrial), j stands for midpoint indicator, and k stands for the type of crop. In the case of human health, the endpoint value was divided per European population in 2019 (513.5 million people) to normalize it per person in Europe. In the case of ecosystem quality (terrestrial), the endpoint value is presented to show species loss in one year.

3. Results and Discussion

3.1. Active Ingredient

Various types of fungicides and active ingredients are used in agriculture. The same systemic or contact fungicide is used in more than one fruit, vegetable, and cereal and rapeseed crop. Table 1 shows how many systemic and contact fungicides are applied per agricultural product. Systemic fungicides are applied more than contact fungicides in terms of different active ingredients. LCA studies that focus on fruits use a smaller number of fungicides than vegetables, cereals, and rapeseed. This does not necessarily mean that the quantity of active ingredients (dosage) is smaller in fruits than vegetables, cereals, or

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rapeseed. In contrast, vegetables employ the largest variety of fungicides, with systemic pesticides dominating, especially azoxystrobin [27–29] and boscalid [27,30,31]. For both fruits and vegetables, the most common contact fungicide is mancozeb [8,11,19,28,29,32]. Lastly, LCA studies of cereals and rapeseed considered almost only systemic fungicides, with azoxystrobin [33,34], propiconazole [34,35], and epoxiconazole dominating the list [34,36]. A detailed version of Table 1 can be found in the Supplementary Materials.

Table 1. Applied fungicides per agricultural product.

Product	Systemic Fungicides (Number)	Contact Fungicides (Number)
Fruits	6	4
Vegetables	16	8
Cereals and rapeseed	10	1

3.2. Dosage

The dosage of fungicides per functional unit is presented based on their active ingredients. These active ingredients are the cause of environmental impacts due to their release to the soil, water, and/or air, and residues in agricultural product. Figure 2 shows the dosage in mass per mass of fruit, vegetable, or cereal and rapeseed. LCA studies of fruits, cereals, and rapeseed consider the highest dosage of fungicides, while vegetables' dosage is much lower. For fruits, grape cultivation in Spain presents the highest dosage of copper-based fungicides [37]. For vegetables, the authors do not disclose what are the fungicides or the active ingredients for cucumber [38] and bean [39] cultivations in Iran and Greece, respectively. Regarding cereals and rapeseed, fungicides used in cotton cultivation in Mali [40] are outliers among reviewed studies because furathiocarb, metalaxyl, and chlorothalonil are supplied almost one order of magnitude more than the second-largest applied fungicide. Rice cultivation in China has the second-largest value because applied fungicides are presented with insecticides and herbicides as pesticides [41]. Lastly, climate and soil type may have an effect on the dosage of applied fungicides, but these two parameters are rarely mentioned in LCA studies.

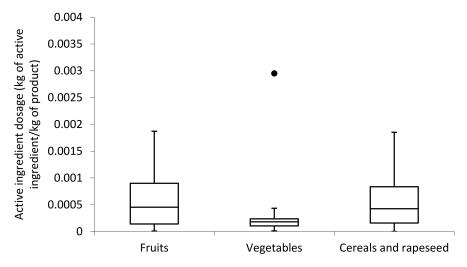


Figure 2. Box plot with median and outlier of active ingredients dosage (of fungicide) per 1 kg of product.

3.3. Fungicides Contribution to Environmental Impacts

Active ingredients of fungicides contribute to various environmental impacts depending on their constituents and release in terms of mass, and release in terms of environmental medium. Figures 3–5 show the extent to which active ingredients contribute to climate change, freshwater ecotoxicity, terrestrial ecotoxicity, human toxicity [3], (terrestrial) acidification [42], and freshwater eutrophication [43]. Among these environmental impacts, only

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climate change is not affected during fungicide application but derives from the production their stage due to greenhouse gases emissions. The other impacts occur during fungicides application in the cultivation stage.

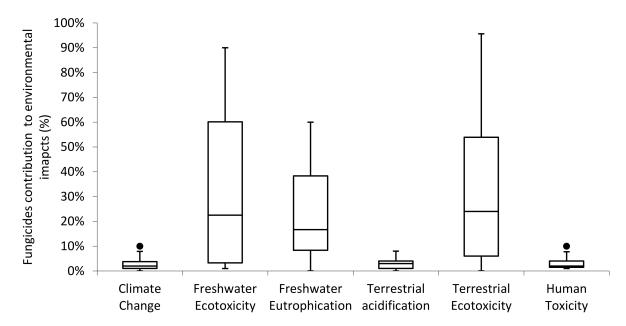


Figure 3. Box plot with median and outliers of fungicides contribution to environmental impacts of 1 kg of fruits.

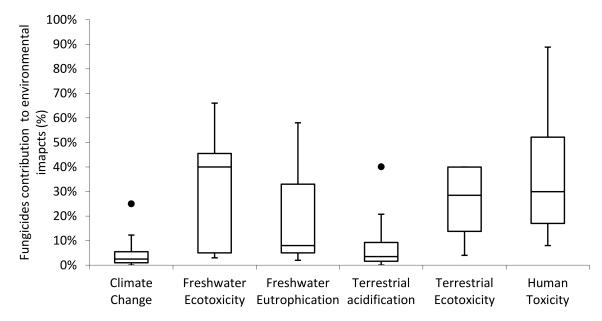


Figure 4. Box plot with median and outliers of fungicides contribution to environmental impacts of 1 kg of vegetables.

The most commonly assessed food product in reviewed LCA studies was grape, and various fungicides were applied during its cultivation. Regarding fruits, terrestrial ecotoxicity, freshwater ecotoxicity, and freshwater eutrophication depend on fungicides based on the reviewed studies (see Figure 3). The range of contribution is large for these environmental impacts and different fungicides are applied. For instance, contribution to terrestrial ecotoxicity increased from 8% to 96% for grape production in South Africa [32] and cherry production in Portugal [44] due to an order of magnitude difference in fungicide dosage during cultivation and the fact that fungicides are grouped with insecticides (in the

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life cycle interpretation stage), respectively. Furthermore, Beauchet et al. [45] presented a 40% contribution to terrestrial ecotoxicity and a 60% contribution to freshwater ecotoxicity and freshwater eutrophication in grape production in France, but these authors only mentioned that they considered a synthetic fungicide and grouped it with insecticides. In total, eight studies assessed freshwater ecotoxicity, which varied between 1% and 90% contribution. The contribution of 1% regards grape production in Iran and freshwater ecotoxicity is dominated by the use of poultry manure [46], while the name of the applied fungicide is not mentioned. In contrast, large quantities of applied fungicides (i.e., Fosetyl-Al and Imazalil) in orange production in Spain resulted in a 90% contribution to freshwater ecotoxicity [11]. Applied fungicides in fruit production contribute to a small extent to climate change [32,44–48], terrestrial acidification [32,44,45,47,48], and human toxicity [11,46–48] due to low characterization factors of fungicides for these impacts and other contributing environmental releases.

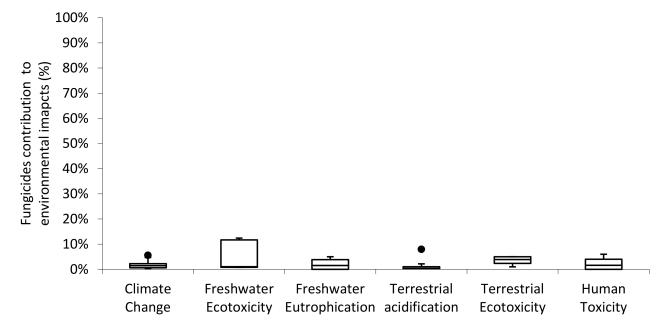


Figure 5. Box plot with median and outliers of fungicides contribution to environmental impacts of 1 kg of cereals and seeds.

Finally, the only distinction between systemic and contact fungicides can be observed for the contact fungicide mancozeb, which is a key contributor to freshwater ecotoxicity and freshwater eutrophication based on [11,32].

The most commonly assessed vegetable product in reviewed LCA studies is tomato. Concerning vegetables, human toxicity, freshwater ecotoxicity, terrestrial ecotoxicity, and freshwater eutrophication depend on fungicides but show a large range (see Figure 4). High contribution to toxicity is found in one study [49] for tomato production in China due to the application of iprodione (>58% regarding human toxicity) and chlorothalonil (90% regarding aquatic ecotoxicity). Similarly, tomato [50,51] and endive [31] production also show a high contribution to freshwater ecotoxicity, from 40% to 66%, respectively, but only Yelboğa [51] distinguished field emissions among various pesticides and reported organo-phosphorus and triazine compounds (i.e., a fungicide) as having the highest and lowest effects, respectively. The range of contribution of freshwater eutrophication varied from 2% to 58%, but only three studies [31,36,50] were identified and all authors considered different fungicides and vegetables. Human toxicity varied between 8% and 89%. The 8% contribution is derived from potato production in Spain, where a mix of fungicides rich in mancozeb and chlorothalonil is applied [36]. On the other hand, the 89% contribution is derived from tomato production in China, where iprodione and chlorothalonil were applied [49]. The range of terrestrial ecotoxicity is smaller than freshwater ecotoxicity, Sustainability **2022**, 14, 846 7 of 12

freshwater eutrophication, and human toxicity, and varied between 4% and 40%. Two studies presented a 40% contribution, but only Yelboğa [51] stated that applied fungicides acetamide anillide, phthalimide, and organo-phosphorus compounds contribute 4.4%, 4.8%, and 14.8%, respectively. Applied fungicides in vegetables production contribute to a small extent to climate change [19,29,30,36,38,49] and terrestrial acidification [28,30,31,36,38].

In general, most applied fungicides are systemic, with boscalid and azoxystrobin being the most common, while the most common contact fungicide is mancozeb. Boscalid contributes to freshwater ecotoxicity and human toxicity, whereas mancozeb contributes to climate change due to its production stage, and terrestrial acidification. Finally, it should be noted that even though the EU expects an improved environmental performance due to increasing organic production, Pedretti et al. [30] reported no statistical difference due to pesticides application between organic and conventional production of spinach leaf.

The most commonly assessed cereal product in reviewed LCA studies was rice. Wheat and rapeseed were also popular choices among researchers. Figure 5 shows the contribution of fungicides to environmental impacts due to cereals and rapeseed production. The contribution of fungicides to climate change, human toxicity, freshwater ecotoxicity, terrestrial ecotoxicity, and freshwater eutrophication was very low compared to fruits and vegetables. Contribution ranged from 0% to 6% for most impacts, except for freshwater ecotoxicity. In freshwater ecotoxicity, the application of systemic fungicides (such as epoxiconazole, propiconazole, and azoxystrobin) in spring and winter rapeseed production in Latvia [34] was the source of the largest contribution of 12% and thiophanate-methyl. In contrast, prochloraz (also systemic fungicide) in rapeseed production in Chile [52] was the source of the lowest contribution of 1%. Fungicides' contribution to freshwater ecotoxicity was higher than other environmental impacts irrespective of the dosage because the latter was the same order of magnitude in the cases of fruits and vegetables, and several authors grouped fungicides with pesticides during life cycle interpretation. A reason for the lower contributions of fungicides in cereals and rapeseed production can be the larger contribution of field emissions of chemical fertilizers because chemical fertilizers contribute to climate change, human toxicity, and ecotoxicity [35].

Finally, no distinction can be made regarding the impact of systemic and contact fungicides based on the reviewed studies. However, it should be pointed out that cereals and rapeseeds consisted of the only LCA studies that employed tebuconazole, epoxiconazole, and prochloraz, which are more toxic to mitochondrial activities, membrane degradations, and caspases 3/7 activities than insecticides and herbicides, according to [12].

3.4. Endpoint Impacts Due to Fungicides Application

In 2019, approximately 42,000, 55,430, and 255,870 ktonnes of fruits, vegetables [53], and cereals [54], respectively, were produced in Europe with conventional farming [55,56]. It should be mentioned that non-edible parts of these agricultural products and rapeseed can be used for the production of biofuels [57,58] depending on the availability of money and consumers' habits [59]. Tables 2 and 3 show minimum and maximum benefits in human health per person and ecosystems quality (terrestrial), respectively, if fungicides are eliminated in fruits, vegetables, and cereals and rapeseed production in Europe. Outliers of climate change (in Figures 3 and 4) were not considered in endpoint calculations based on Grubbs' test. These outliers were derived from studies that grouped fungicides emissions with farm emissions.

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Table 2. Human health and ecosystem quality (terrestrial) based on low contribution of fungicides to climate change, human toxicity, terrestrial ecotoxicity, and terrestrial acidification.

	Disability Adjusted Life Years per Person $(y \cdot p^{-1})$		Time Integrated Species Loss (Species·yr)		
	Climate Change	Human Toxicity	Climate Change	Terrestrial Ecotoxicity	Terrestrial Acidification
		Individualist perspecti	ve		
Fruits	4.4×10^{-7}	1.8×10^{-6}	0	0	0
Vegetables	4.6×10^{-7}	2.5×10^{-7}	0	7.7×10^{-1}	0
Cereals and rapeseed	$1.8 imes 10^{-7}$	$1.3 imes 10^{-5}$	0	$6.2 imes 10^{-1}$	7.1
		Hierarchical perspecti	ve		
Fruits	0	1.8×10^{-6}	0	0	0
Vegetables	0	2.5×10^{-7}	0	7.7×10^{-1}	0
Cereals and rapeseed	0	1.3×10^{-5}	0	6.2×10^{-1}	7.1
		Egalitarian perspectiv	re		
Fruits	0	1.8×10^{-6}	0	0	0
Vegetables	0	2.5×10^{-7}	0	$7.7 imes 10^{-1}$	0
Cereals and rapeseed	0	1.3×10^{-5}	0	6.2×10^{-1}	7.1

Table 3. Human health and ecosystem quality (terrestrial) based on high contribution of fungicides to climate change, human toxicity, terrestrial ecotoxicity, and terrestrial acidification.

	Disability Adjusted Life Years per Person $(y \cdot p^{-1})$		Time Integrated Species Loss (Species·yr)		
	Climate Change	Human Toxicity	Climate Change	Terrestrial Ecotoxicity	Terrestrial Acidification
		Individualist perspecti	ve		
Fruits	4.4×10^{-7}	1.8×10^{-5}	1.5	18.4	1.8
Vegetables	4.6×10^{-7}	2.7×10^{-6}	1.6	7.7	5.8
Cereals and rapeseed	1.8×10^{-7}	5.3×10^{-6}	0.6	3.1	56.5
		Hierarchical perspecti	ve		
Fruits	5.0×10^{-6}	1.8×10^{-5}	7.7	18.4	1.8
Vegetables	5.3×10^{-6}	2.7×10^{-6}	8.2	7.7	5.8
Cereals and rapeseed	2.0×10^{-6}	5.3×10^{-6}	3.1	3.1	56.5
		Egalitarian perspectiv	re		
Fruits	7.0×10^{-5}	1.8×10^{-5}	69.0	18.4	1.8
Vegetables	7.4×10^{-5}	2.7×10^{-6}	73.1	7.7	5.8
Cereals and rapeseed	2.8×10^{-5}	5.3×10^{-6}	28.1	3.1	56.5

Potential benefits for human health are greater in fruits than vegetables, cereals, and rapeseed. In addition, the fact that midpoint to endpoint conversion factors ranges minimally between individualist and hierarchist perspectives results in similar values for both perspectives for both low and high fungicides contributions to climate change and human toxicity. In contrast, concerning the egalitarian perspective, the climate change midpoint to endpoint conversion factor increases by at least one order of magnitude compared to the individualist perspective and results in adverse effects for all crops (Table 3). This increase in climate change conversion factor results in vegetables damaging human health more than cereals and rapeseed with the egalitarian perspective. Human toxicity is the midpoint indicator that influences human health results for all perspectives. Climate change insignificantly affects human health from individualist and hierarchist perspectives but increases its effect with the egalitarian perspective due to the increase

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in midpoint to endpoint contribution. Lastly, the production of cereals is an order of magnitude higher than fruits and vegetables, but the very low contribution of fungicides in human toxicity of cereals and rapeseed does not result in higher toxicity per person than fruits.

The elimination of fungicides in cereals and rapeseed results in greater potential benefits in ecosystem quality (terrestrial) than vegetables and fruits for individualist and hierarchist perspectives. This occurs mainly due to production yields of cereals and rapeseed and larger terrestrial acidification score of cereals and rapeseed than other crops, especially with respect to the lowest amount of expected ecosystem benefits (Table 2). In contrast, terrestrial ecotoxicity is the largest contributor to benefits in ecosystem quality for the cases of fruits and vegetables based on the individualist and hierarchist perspectives. The individualist and hierarchist perspectives result in a greater difference in benefits among considered crops, but similar benefits are found with the egalitarian perspective. The egalitarian perspective results in more species loss due to the increase in midpoint to endpoint conversion factor for climate change and production yields for all crops.

Lastly, it should be mentioned that calculations for ecosystem quality (terrestrial) result in unrealistically high values of species loss. According to the International Union for Conservation of Nature, as of 2015, 36 species became extinct in Europe [60]. Thus, the term "species" here seems to be inappropriate, but "animals" could be used because the U.S. Fish and Wildlife Service estimates that approximately 67 million birds die from pesticide poisoning each year [61].

4. Conclusions

This review paper aimed to collect LCA studies focusing on the use of fungicides in fruits, vegetables, and cereals and rapeseed, and calculate damages to human health and ecosystem quality (terrestrial). It was found that LCA practitioners often do not distinguish the contribution of insecticides, herbicides, and fungicides to considered environmental impacts. Several LCA studies mentioned explicitly what fungicides are used and what is their dosage in the life cycle inventory stage, but grouped them with other pesticides in the life cycle interpretation stage.

It was found that fungicides contribute more to environmental impacts in the cultivation of fruits and vegetables than in the cultivation of cereals and rapeseed. Furthermore, fungicides dosage does not vary among the produced crops. Therefore, the specific chemical is crucial to be mentioned to select characterization factors to assess toxicity. If current conventional agriculture practices become organic, European citizens will benefit with up to 47 min per year of increased life quality due to fungicides elimination in fruits and vegetables cultivation. In addition, species loss benefits are also expected up to 90 species per year due to fungicides elimination in fruits and vegetables cultivation.

It is recommended that LCA practitioners increase transparency in their studies by showing what input materials are contributing to selected environmental impacts, and that new LCA studies are designed to assess explicitly the effect of different fungicides on human toxicity and ecotoxicity.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su14020846/s1, Excel file.

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Data Availability Statement: The data presented in this study are available in Tables 2 and 3 within this article, and Supplementary Materials.

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