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POLYTECHNIQUE MONTRÉAL

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Modélisation de l'influence du changement climatique sur l'écotoxicité terrestre du cuivre dans le cadre de l'analyse du cycle de vie prospective : application à la filière vitivinicole

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Thèse présentée en vue de l'obtention du diplôme de *Philosophiæ Doctor*

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présentée par **Ivan VIVEROS SANTOS**

en vue de l'obtention du diplôme de *Philosophiæ Doctor*

a été dûment acceptée par le jury d'examen constitué de :

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DÉDICACE

À ma famille

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RESUME

In vino veritas, dans le vin se trouve la vérité, disait Pline l'Ancien. En effet, la précocité des dates des vendanges, des changements dans la composition des raisins, ainsi que des pertes de rendement en viticulture ont été attribuées aux impacts des changements climatiques déjà manifestés dans quelques régions viticoles. De plus, dans son sixième rapport, le Groupe d'experts intergouvernemental sur l'évolution du climat (GIEC) affirme avec un degré de certitude élevé que plus de la moitié de l'élévation observée de la température moyenne de la planète depuis 1979 est causée par les émissions de gaz à effet de serre (GES) générées par les activités humaines. Donc, la vérité sur l'origine anthropique du changement climatique n'est plus contestée. Le changement climatique entraine des impacts sur l'humain, sur les écosystèmes et en particulier sur les systèmes vulnérables aux variations du climat, comme c'est le cas de la filière vitivinicole. De plus, il est projeté que le changement climatique influera sur l'ampleur de l'impact des polluants dans des conditions futures en raison de son incidence sur le sort, l'exposition et le niveau d'effet des substances chimiques.

Actuellement, la filière vitivinicole fait face à des enjeux environnementaux liés à l'émission de GES, ainsi qu'à l'utilisation d'engrais, à l'application de pesticides, et plus particulièrement à l'application de fongicides à base de cuivre, dont l'utilisation extensive a donné lieu à des concentrations élevées de ce métal dans les sols viticoles; de plus, cela a causé également la pollution des eaux de surface par ruissellement. En outre, en raison des changements climatiques, les vignerons seront amenés à mettre en œuvre des stratégies d'adaptation. En conséquence, il devient nécessaire d'empêcher que les impacts environnementaux des stratégies d'adaptation excédent ceux des mesures d'atténuation au changement climatique.

L'analyse du cycle de vie (ACV) est un outil d'évaluation environnementale qui a été déjà utilisé dans le secteur vitivinicole afin de définir des pistes d'amélioration du profil environnemental de la production de raisins et du vin. Dans le contexte du changement climatique et au vu de ces impacts projetés sur la filière vitivinicole, l'utilisation de l'ACV dans la définition de stratégies d'adaptation au changement climatique devient encore plus pertinente, dans le but d'éviter le déplacement d'impacts d'une catégorie à une autre, ou encore, un déplacement d'impact dans l'espace et dans le temps. Néanmoins, il existe quelques défis méthodologiques que l'ACV doit surmonter afin de mieux évaluer des scénarios prospectifs.

Cette thèse vise donc à développer une approche d'analyse du cycle de vie prospective permettant d'inclure l'influence du changement climatique à la fois sur l'inventaire du cycle de vie et sur l'évaluation des impacts écotoxiques terrestres du cuivre. La méthodologie générale développée est appliquée à l'étude de cas de la viticulture où le changement climatique et les impacts écotoxiques terrestres causés par l'application de fongicides à base de cuivre sont deux enjeux environnementaux importants. Pour parvenir à cet objectif, trois étapes de modélisation sont proposées, lesquelles ont donné lieu à trois articles scientifiques.

La première étape est portée sur l'évaluation de la variabilité spatiale de l'écotoxicité terrestre causée par l'utilisation de fongicides à base de cuivre. Le manque de facteurs de caractérisation (FC) pour l'écotoxicité terrestre du cuivre tenant compte de la spéciation du métal dans une méthode d'évaluation des impacts du cycle de vie (ÉICV), a mené d'abord au calcul de FCs. Les FCs obtenus avec le modèle WHAM s'étendent sur 5,5 ordres de grandeur pour l'ensemble des sols non calcaires du monde. En outre, l'étude de cas incluant quatre vignobles européens a montré l'influence de la régionalisation l'inventaire, car le classement du score d'impact a été déterminé par la quantité de fongicide utilisée.

La deuxième étape méthodologique est portée sur la prise en compte de l'influence du changement climatique sur l'inventaire de cycle de vie de scénarios prospectifs en viticulture pour deux vignobles en France (Val de Loire et Languedoc-Roussillon) selon deux scénarios d'émissions de GES et quatre périodes. Dans un premier niveau d'analyse, les impacts du changement climatique sur le rendement des vignobles ont été intégrés à l'aide d'une méthode de dissociation statistique des séries temporelles de rendement et de climat. Dans le cas des vignobles analysés, l'influence des changements climatiques projetés donne lieu à des conclusions opposées quant aux impacts environnementaux projetés. Par exemple, selon le scénario à fortes émissions de GES (SSP5-8.5), l'empreinte carbone (kg $CO_2eq \cdot kg\ raisins^{-1}$) du vignoble en Languedoc-Roussillon augmenterait d'environ 29% d'ici la fin du siècle, tandis que celle du vignoble en Val de Loire diminuerait d'environ 10%. Néanmoins, dans un deuxième niveau d'analyse, lorsque les impacts des événements extrêmes sont modélisés en fonction de leur probabilité et leur niveau de dommage, les conclusions pointent vers la même direction: les impacts environnementaux augmentent de manière importante en raison des impacts entrainés par les événements extrêmes à la fois sur le rendement et sur l'adoption des stratégies d'adaptation contre des extrêmes climatiques. Ainsi, à la fin du siècle, selon le scénario SSP5-8.5, l'empreinte carbone du vignoble en Languedoc-

Roussillon serait quatre fois son empreinte actuelle, tandis qu'elle serait multipliée par un facteur trois dans le cas du vignoble en Val de Loire.

La troisième étape méthodologique correspond au développement des FCs futurs tenant compte de la spéciation du métal et de l'influence du changement climatique. Ce chapitre montre que bien que la variabilité spatiale des FCs pour l'écotoxicité terrestre soit plus importante (1,96 ordre de grandeur) que la variabilité temporelle (0,78 ordre de grandeur) que l'omission de l'influence du changement climatique sur l'évaluation de l'écotoxicité terrestre représente, de manière générale, une sous-estimation de ces impacts dans l'évaluation de scénarios prospectifs pour la plupart des régions viticoles en Europe. Ainsi, selon le scénario RCP8.5, les FCs pour l'écotoxicité terrestre des vignobles analysés dans le second article augmenteraient de 60% et 7% respectivement dans les vignobles en Val de Loire et Languedoc-Roussillon. Compte tenu des projections d'augmentation de la quantité de fongicides à base de cuivre utilisée à l'horizon 2050 de 33.5% et 12.5%, respectivement dans ces deux vignobles, si l'on combine les inventaires prospectifs de l'article 2 et les FCs futurs de l'article 3, cela entrainerait une augmentation des impacts écotoxiques terrestres de 113.5% et 20% respectivement par rapport à un scénario qui ne tiendrait compte de l'influence des changements climatiques ni sur les pratiques agricoles ni sur l'écotoxicité du cuivre.

Les principales limites de ce projet correspondent au fait que les FCs calculés sont valables uniquement pour les sols non calcaires, en raison du domaine d'application des modèles TBLM utilisés pour calculer des facteurs d'effet (FE). De plus, les FCs futurs ne tiennent pas compte de l'influence du changement climatique sur le FE, car les résultats rapportés dans la littérature ne permettent pas de départager la contribution des changements dans la température de celle des changements associés à des modifications de la spéciation. De plus, l'approche pour estimer l'impact des événements extrêmes ne permet pas de tenir compte des impacts synergistes qui peuvent survenir, par exemple, lorsque des périodes de sècheresse et des ondes de chaleurs surviennent simultanément.

Ce projet de recherche a permis de prendre en considération des dimensions spatiales et temporelles dans l'application de l'ACV prospective. Plus particulièrement, au vu des résultats, la méthode générale a montré la pertinence d'inclure l'influence du changement climatique à la fois sur l'inventaire du cycle de vie et sur l'évaluation de l'écotoxicité terrestre du cuivre.

ABSTRACT

In vino veritas, in wine, there is truth, the Roman author Pliny the Elder wrote. The truth is that the advancement of harvest dates, the changes in grape composition, and the losses of yield in viticulture have been attributed to the impacts of climate change already manifested in some wine-growing regions. Moreover, the sixth report of the Intergovernmental Panel on Climate Change (IPCC) states with a high confidence level that more than half of the observed rise in average temperature on Earth since 1979 has been caused by greenhouse gas (GHG) emissions generated by human activities. Thus, the truth about the anthropogenic origin of climate change is no longer doubtful. Climate change impacts humans, ecosystems, and particularly systems vulnerable to climate variations, as is the case with viticulture. Furthermore, projected climate change can also affect the extent of pollutants' impacts under future conditions because of its influence on the fate, exposure, and effect level of chemicals.

Currently, the wine industry faces some environmental challenges related to the generation of GHG emissions, the application of fertilizers and pesticides, and particularly the use of copper-based fungicides. The extensive use of copper-based fungicides has led to high concentrations of this metal in vineyard soils and surface water pollution by runoff. In addition, winegrowers will need to implement some adaptation strategies to cope with projected climate change. Consequently, it becomes imperative to prevent the environmental impacts of adaptation strategies from exceeding those aimed at mitigating climate change.

Life Cycle Assessment (LCA) is an environmental assessment method that has been used in the wine sector to identify ways to improve the environmental profile of grape and wine production. Considering the projected impacts of climate change on the wine sector, applying LCA in the definition of climate change adaptation strategies becomes even more relevant, to avoid potential problems shifting from one impact category to another or even a problem shift in space and time. Nonetheless, there are some methodological challenges that LCA needs to overcome to better assess prospective scenarios.

Therefore, this thesis aims to develop a prospective LCA approach to account for the influence of climate change on the life cycle inventory and on the characterization of copper terrestrial ecotoxicity. The general methodology developed in this thesis is applied to the case study of viticulture, since climate change and copper terrestrial ecotoxicity impacts are two environmental

concerns of this agricultural sector. To achieve the main goal, three modelling steps are proposed, which have led to three scientific articles.

The first modelling step addressed the evaluation of the spatial variability of terrestrial ecotoxicity caused by the application of copper-based fungicides. The lack of characterization factors (CF) accounting for metal speciation to assess copper terrestrial ecotoxicity integrated into life cycle impact assessment (LCIA) methods led first to the calculation of CFs. The CFs obtained with WHAM, a geochemical speciation model, extend over 5.5 orders of magnitude for non-calcareous soils of the world. Furthermore, the case study on four European vineyards showed the influence of considering regionalization at the inventory level, since the impact score ranking was determined by the amount of copper-based fungicides applied.

The second methodological step focused on accounting for the influence of climate change on the life cycle inventory of prospective scenarios in viticulture for two vineyards in France (Loire Valley and Languedoc-Roussillon) according to two GHG emissions scenarios and four periods. In the first level of analysis, the impacts of climate change on grape yield were considered by employing a detrending method on time series of grapes yield against climate variables (average temperature and total precipitation during the grape-growing season). For the analyzed vineyards, the influence of projected climate change leads to opposite conclusions regarding the projected environmental impacts. For instance, according to the high GHG emissions scenario (SSP5-8.5), the carbon footprint (kg CO_2eq kg grapes^{-1}) of the vineyard from Languedoc-Roussillon would increase by around 29% by the end of the century, whereas the carbon footprint of the vineyard from the Loire Valley would decrease by approximately 10%. Nevertheless, in the second level of analysis, when the impacts of extreme events are accounted for according to their probability and their level of damage, the conclusions point in the same direction: the environmental impacts increase drastically due to the impacts caused by extreme events both on grape yield and on the implementation of adaptation strategies aiming at coping with climate change. By the end of the century, according to the SSP5-8.5 scenario, the carbon footprint of the vineyard from Languedoc-Roussillon is expected to rise fourfold related to the current carbon footprint, whereas it will increase threefold for the vineyard from the Loire Valley.

The third methodological step corresponds to the development of future CFs considering metal speciation and the influence of climate change. This chapter shows that although the spatial variability of CFs for copper terrestrial ecotoxicity is more important (1.96 orders of magnitude) than the temporal variability (0.78 orders of magnitude), omitting the influence of climate change on the assessment of copper terrestrial ecotoxicity would lead to a potential underestimation of these impacts in the assessment of prospective scenarios for most wine-growing regions in Europe. For instance, according to the RCP8.5 scenario, the CFs for terrestrial ecotoxicity of the vineyards analyzed in the second article would increase by 60% and 7%, respectively for the vineyards in the Loire Valley and Languedoc-Roussillon. Given the projected increases in the quantity of copper-based fungicides used by 2050 of 33.5% and 12.5%, respectively in these two vineyards, if we combine the prospective inventories of article 2 and the future CFs of article 3, this would lead to an increase in terrestrial ecotoxicity impacts of 113.5% and 20% respectively, compared to a scenario that neglects the influence of climate change on both agricultural practices and copper terrestrial ecotoxicity.

The main limitations of this project correspond to the fact that the calculated CFs are only valid for non-calcareous soils, due to the range of the application of the TBLM models used to calculate effect factors. In addition, future CFs do not account for the influence of climate change on effect factors, because the toxicity levels reported in the literature do not allow to separate the contribution of changes in temperature from that of the changes associated with modifications of metal speciation. In addition, the methodological approach for estimating the impact of extreme events does not allow modelling the synergistic impacts that may result, for example, when periods of drought and heat waves occur simultaneously.

This research project made it possible to take into consideration spatial and temporal dimensions in the application of prospective LCA. More particularly, given the results, the general method showed the relevance of including the influence of climate change both on the life cycle inventory and on the assessment of the terrestrial ecotoxicity of copper.

TABLE DES MATIÈRES

LISTE DES TABLEAUX

LISTE DES FIGURES

LISTE DES SIGLES ET ABRÉVIATIONS

ACF	Accessibility factor
ACV	Analyse du cycle de vie
AFOLU	Agriculture, Forestry and Other Land Use
AMI	Assessment of mean impact
AOP	Adverse Output Pathway
BF	Bioavailability factor
CDD	Consecutive dry days
CF	Characterization factor
CFD	Consecutive frost days
CITS	Climate-induced toxicant sensitivity
CMIP	Coupled Model Intercomparison Project
DALY	Disability-adjusted life years
DOC	Dissolved organic matter
EF	Effect factor
ÉICV	Évaluation des impacts du cycle de vie
EQ	Ecosystem quality
ESDAC	European Soil Data Centre
EU	European Union
FAO	Food and Agriculture Organization of the United Nations
FC	Facteur de caractérisation
FF	Fate factor
FIAM	Free ion activity model
FU	Functional unit
GCM	Global climate model
GES	Gaz à effet de serre
GHG	Greenhouse gases
GIEC	Groupe d'experts intergouvernemental sur l'évolution du climat
GIS	Geographic information system
HH	Human health
HWSD	Harmonized World Soil Database

ICV	Inventaire du cycle de vie
IDW	Inverse distance weighting (interpolation technique)
IPCC	Intergovernmental Panel on Climate Change
ISO	International Organization for Standardization
JJA	June-July-August
Kd	Coefficient de partition sol-eau
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
MAM	March-April-May
MLR	Multiple linear regressions
MO	Matière organique
NPP	Net primary production
NUTS	Nomenclature of Territorial Units for Statistics
OIV	International Organisation of Vine and Wine
OM	Organic matter
PAF	Potentially affected fraction of species
PDF	Potentially disappeared fraction of species
POP	Polluant organique persistant
PTO	Pathway of technical operations
RCP	Representative concentration pathways
SETAC	Society of Environmental Toxicology and Chemistry
SOC	Soil organic carbon
SSP	Shared Socioeconomic Pathways
TBLM	Terrestrial biotic ligand model
TICS	Toxicant-induced climate sensitivity
UNEP	United Nations Environment Programme
VBA	Visual Basic for Applications
WSDI	Warm-spell duration index
WW	Warm and wet days
YLR	Yield loss rate

LISTE DES ANNEXES

CHAPITRE 1 INTRODUCTION

Les systèmes agroalimentaires causent des impacts environnementaux non négligeables en raison de l'utilisation de terres, de l'émission de gaz à effet de serre et de l'application de pesticides. Dans le cas de la filière vitivinicole, il a été estimé qu'elle contribue d'environ 0,3% à l'empreinte carbone de l'ensemble des activités anthropiques. En Europe, par exemple, il a été rapporté que 13% du volume total des pesticides utilisé est appliqué en viticulture, bien qu'elle occupe seulement 3% de la surface des terres cultivées (Rugani et al., 2013). Plus particulièrement, l'utilisation extensive des fongicides à base de cuivre en viticulture pour lutter contre le mildiou a contribué à l'accumulation de ce métal dans les sols viticoles, entrainant des impacts écotoxiques terrestres et de pollution d'eau par lixiviation par et par ruissellement.

Le changement climatique est un défi auquel fait face la filière vitivinicole. L'avancée des dates des vendanges et des modifications de la qualité et quantité des raisins ont été associées à l'augmentation des températures observées dans quelques régions viticoles européennes. En conséquence, pour maintenir la production et la typicité du vin il sera nécessaire d'y mettre en œuvre des stratégies d'adaptation. De plus, selon l'ampleur du changement climatique projeté, celui-ci affectera le devenir, l'exposition et l'écotoxicité des polluants dans le futur. C'est le cas de l'impact écotoxique terrestre des métaux, car des modifications des conditions des sols telles que la teneur en matière organique, l'érosion du sol, et des changements dans le régime de précipitations affectent à leur tour leur spéciation, biotransformation, biodisponibilité et le niveau d'effet de ces substances.

Ces dernières années, l'analyse du cycle de vie (ACV) a été amplement recommandée et utilisée pour supporter la transition vers des systèmes agroalimentaires plus durables. Toutefois, il existe quelques défis méthodologiques que cet outil doit surmonter. D'abord, les facteurs de caractérisation disponibles dans les logiciels d'ACV pour l'évaluation de l'écotoxicité terrestre du cuivre ne tiennent pas compte du lieu d'émission ni de la spéciation du métal. Pour cette raison, quelques études d'ACV portant sur la production de vin ont décidé de négliger la contribution du cuivre à l'impact écotoxique, ce qui nuit la crédibilité des résultats. D'autre part, dans le contexte du changement climatique, les stratégies d'adaptation à mettre en place dans la filière vitivinicole pourraient entrer en compétition avec des stratégies de mitigation, il sera donc nécessaire d'évaluer les impacts environnementaux associés auxdites stratégies d'adaptions. En outre, le changement climatique affectera le niveau d'impact des émissions de polluants dans des scénarios futurs.

L'objectif général de ce projet doctoral est de développer une approche d'analyse du cycle de vie prospective permettant d'inclure l'influence du changement climatique à la fois sur l'inventaire du cycle de vie et sur l'évaluation des impacts écotoxiques terrestres du cuivre.

Le deuxième chapitre de cette thèse présente la revue de la littérature ayant mené à l'hypothèse de recherche. Ensuite, le troisième chapitre décrit l'hypothèse de recherche, ainsi que l'objectif général et les objectifs spécifiques, ainsi que la description de la méthodologie proposée pour répondre à ces objectifs. Les chapitres 4, 5 et 6 correspondent aux manuscrits issus de l'application de la méthodologie développée dans le cadre de cette thèse. Plus spécifiquement, le chapitre 4 présente les résultats de l'évaluation régionalisée de l'impact d'écotoxicité terrestre causé par l'application de fongicides à base de cuivre en viticulture. Ceci a mené à la rédaction du premier article intitulé « *Regionalized Terrestrial Ecotoxicity Assessment of Copper-Based Fungicides Applied in Viticulture* » soumis à la revue *Sustainability*. Le chapitre 5 quant à lui porte sur la prise en compte du changement climatique dans la définition de l'inventaire du cycle de vie de la viticulture sous des scénarios prospectifs. Ceci a mené à la rédaction du deuxième article intitulé « *Prospective life cycle assessment of viticulture under climate change scenarios, application on two case studies in France* » soumis à la revue *Science of the Total Environment*. Le chapitre 6 vise le développement de facteurs de caractérisation futurs pour l'écotoxicité terrestre du cuivre intégrant l'influence du changement climatique sur le sort, l'exposition et la biodisponibilité du métal. Ceci a donné lieu à la rédaction du troisième article intitulé « *Modelling the influence of climate change on characterization factors for copper terrestrial ecotoxicity* » soumis à la revue *Journal of Cleaner Production.* Finalement, la thèse conclut par une discussion sur les contributions, l'identification des limites du projet doctoral et la proposition de quelques recommandations pour des pistes de recherche futures.

CHAPITRE 2 REVUE DE LA LITTÉRATURE

Ce chapitre présente la revue de la littérature ayant mené à l'hypothèse de recherche et aux objectifs de cette thèse. D'abord, le phénomène du changement climatique est introduit (2.1), lequel est suivi de la description de quelques enjeux de la filière vitivinicole, dont l'adaptation au changement climatique et l'utilisation de fongicides à base de cuivre (2.2). Ce dernier enjeu mène à la description de la méthodologie d'analyse du cycle de vie qui est utilisé pour évaluer les impacts entraînés par ces fongicides. Des plus, des limites sur la prise en compte des dimensions spatiale et temporelle autant sur l'inventaire que sur l'évaluation des impacts environnementaux potentiels sont discutées (2.3). Ensuite, la section 2.4 présente les limites de l'évaluation de l'écotoxicité terrestre des métaux et l'influence du changement climatique sur cette catégorie d'impact (2.4). Finalement, une synthèse de la problématique générale issue de la revue de littérature est présentée à la section 2.5.

2.1 Le changement climatique

Le climat a toujours joué un rôle décisif dans l'histoire de l'humanité. Depuis les tribus nomades, qui cherchaient des aliments saisonniers, jusqu'à l'établissement des premières grandes civilisations agricoles en Mésopotamie, Égypte et Chine, l'homme a été toujours dépendant des avantages et des contraintes imposées par le climat (Jones & Webb, 2010). Bien que le climat ait toujours fluctué dans l'histoire de la planète, depuis le siècle dernier, un dérèglement du système climatique global sans précédent a été détecté par la communauté scientifique. Dans son sixième rapport, le Groupe d'experts intergouvernemental sur l'évolution du climat (GIEC) souligne que chacune des quatre dernières décennies a été successivement plus chaude à la surface de la Terre que toutes les décennies précédentes depuis 1850. De plus, le rapport souligne qu'il est extrêmement probable (90-100%) que plus de la moitié de l'augmentation observée de la température moyenne à la surface du globe depuis 1979 soit due à l'augmentation des émissions de gaz à effet de serre (GES) issues des activités anthropiques. En outre, de nombreux changements dans le système climatique se sont amplifiés, tels que des augmentations de la fréquence et l'intensité des températures extrêmes, des vagues de chaleur, des répartitions très variables des précipitations, ainsi que la réduction du manteau neigeux et du pergélisol (IPCC, 2021).

Le changement climatique provoque des impacts sur l'humain, sur les écosystèmes et notamment sur le domaine agricole. C'est le cas de la vigne, laquelle est un bon marqueur du climat, car elle réagit à ces changements et plus particulièrement à l'augmentation de température dans la plupart des régions viticoles (voir section 2.2.2) (Quénol, 2014). Selon les conditions climatiques futures prévues, ainsi que les impacts projetés par ces variations climatiques sur la production et la qualité de la récolte, les vignerons devront instaurer des leviers d'adaptation au changement climatique, lesquels pourraient dépasser les mesures d'atténuation (voir section 2.2.3). En outre, le changement climatique affecte le devenir et l'ampleur des impacts des émissions de polluants (voir section 2.4.2). Un groupe de travail Pellston organisé par la *Society of Environmental Toxicology and Chemistry* (SETAC) a abordé la question concernant l'influence du changement climatique sur la modélisation et l'application l'écotoxicologie à des scénarios prospectifs. La conclusion principale tirée par ce groupe de travail est que le changement climatique devient un facteur de stress qui modifie la capacité des organismes à résister à la présence de polluants dans l'environnement, et inversement, la présence de polluants peut dérégler des processus d'accoutumance aux changements climatiques (Stahl Jr. et al., 2013).

2.1.1 Le climat du futur : les scénarios du GIEC

Des modèles climatiques sont utilisés pour analyser l'évolution du climat, mais aussi pour étudier comment il pourrait changer dans le futur. Ces modèles climatiques simulent la physique, la chimie, et la biologie de l'atmosphère, de la surface terrestre et des océans. Quant à la projection du climat futur, elle est basée sur des informations découlant de scénarios des émissions de GES. Ces scénarios tiennent compte de plusieurs facteurs tels que la croissance économique et démographique, les modes de vie et des comportements, l'évolution de la consommation d'énergie, de l'utilisation des terres, de la technologie, ainsi que des politiques climatiques. La panoplie des scénarios d'émissions de GES s'est transformée au fil des *Projets d'inter-comparaison de modèles couplés* (*CMIP, Coupled Model Intercomparison Project)* et des divers rapports du GIEC (IPCC, 2014, 2021).

Dans la cinquième itération du CMIP, l'ensemble des scénarios de GES est désigné sous le terme de « *Profils représentatifs d'évolution de concentration* » (RCP pour « Representative concentration pathways »), lesquels permettent aussi d'évaluer les coûts associés aux réductions des émissions de GES selon les stratégies d'atténuation à mettre en place. Ces scénarios s'étalent d’un scénario d’atténuation stricte (RCP2.6) à un scénario d’émissions très élevées de GES (RCP8.5), en incluant deux scénarios intermédiaires (IPCC, 2014). Dans le contexte du CMIP6 et du sixième rapport du GIEC, la dernière version des scénarios correspond aux « *Profils socioéconomiques partagés »* (SSP pour *« Shared Socieconomic Pathways »*). Les scénarios de SSP sont les plus complexes développés jusqu’à présent et à l’instar des scénarios RCP, ils s’étalent d’un scénario de mesures de mitigation très ambitieuses à un scénario des émissions croissantes de GES (IPCC, 2021). Les scénarios SSP sont employés pour estimer des politiques sur le changement climatique, tandis que les scénarios RCP décrivent l’évolution des concentrations des GES. Ainsi, les scénarios SSP sont utilisés en association avec une récente version améliorée des scénarios RCP. Cette combinaison permet d’évaluer l’impact des divers choix de politique climatique selon les scénarios SSP, et ainsi déterminer le degré de difficulté de parvenir au forçage radiatif à la fin du siècle établi par un RCP donné (O’Neill et al., 2016).

À leur tour, les sorties des projections du climat futur sont utilisées pour évaluer leur impact sur des systèmes vulnérables à ces changements. Par exemple, les projections de température et de précipitation sont utilisées pour étudier leur influence sur les systèmes agricoles. Par exemple, la viticulture a été analysée à l’aide des indicateurs bioclimatiques est des modèles phénologiques, c’est-à-dire, des modèles décrivant le développement de la vigne, pour prévoir l’incidence du climat futur sur cette culture, mais aussi pour identifier des leviers d’adaptation au changement climatique (Marco Moriondo et al., 2015; Sacchelli et al., 2017; David Santillán et al., 2019). Plus largement, des indicateurs agroclimatiques génériques ont été développés afin d’évaluer l’impact des changements climatiques sur l’interaction plante-climat à l’échelle de la planète pour orienter les actions d’adaptation et d’atténuation en agriculture (Copernicus, 2021). En outre, les projections du climat permettent d’évaluer leur influence sur des conditions environnementales futures, telles que la teneur en carbone organique dans les sols, ainsi que sur le taux d’érosion des sols (Panagos et al., 2021; Yigini & Panagos, 2016).

2.2 Des enjeux environnementaux de la filière vitivinicole

La part de l’empreinte environnementale associée au secteur agroalimentaire devient de plus en plus élevée en raison de l’augmentation de la population, qui atteint les huit milliards. Les impacts environnementaux du système agroalimentaire incluent des impacts globaux comme l’utilisation des terres, la disponibilité d’eau, la perte de biodiversité et le réchauffement climatique, ainsi que des impacts toxiques et écotoxiques (Notarnicola et al., 2017; Springmann et al., 2018). Si des modifications ne sont pas apportées aux systèmes agricoles et aux habitudes de consommation, les impacts environnementaux de ces systèmes deviendront de plus en plus sévères au risque de dépasser des limites planétaires, car la production des aliments devrait augmenter de plus de 60% d’ici 2050. Relativement à cette problématique, l’analyse du cycle de vie (ACV) est considérée comme une méthodologie qui peut contribuer à la transition vers des systèmes agricoles plus durables (Notarnicola et al., 2016). De manière générale, les principales applications de l’ACV dans l’évaluation des systèmes agricoles comportent (Sala et al., 2016):

- L’identification des étapes les plus contributrices aux impacts environnementaux (points chauds) de la chaine d’approvisionnement.
- L’optimisation de la chaine d’approvisionnement à travers la comparaison de différents scénarios.
- L’évaluation des scénarios futurs qui tient compte de l’évolution technologique et des changements dans les conditions environnementales futures.
- L’évaluation des impacts sociaux des modes de production et de consommation.

Bien que la viticulture ne soit pas strictement une culture indispensable à la survie de l’humanité, elle représente une pratique agricole importante et un héritage culturel dans plusieurs pays (David Santillán et al., 2019). En effet, en 2019, la culture de la vigne couvrait une surface de 7,3 millions d’hectares avec une production de 85 millions de tonnes de raisins (OIV, 2022). Néanmoins, comme toute activité économique, la viticulture entraine aussi des impacts environnementaux non négligeables. En Europe, par exemple, la viticulture utilise environ 13% en masse des pesticides synthétiques appliqués, bien qu’elle n'occupe que 3% des terres cultivées (Christel Renaud-Gentié et al., 2015). L’utilisation de pesticides à long terme est responsable d’une augmentation de la concentration de ces substances dans le sol et dans l’eau, entrainant des impacts toxiques et écotoxiques (Komárek et al., 2010). De plus, il a été estimé que l’empreinte carbone du secteur vitivinicole correspond à environ 0,3% de l’empreinte carbone totale de l’ensemble des activités anthropiques (Rugani et al., 2013).

La variabilité spatiale et temporelle des systèmes agricoles soulève des problèmes lors de la définition de l’inventaire du cycle de vie de ces systèmes et l’utilisation de données moyennes peut affecter significativement les résultats (Sala et al., 2016). Par exemple, une étude d'ACV portant sur la production d'une bouteille de vin en Galice (Espagne) a montré la pertinence de considérer l'année de la récolte des raisins. En analysant la période de 2007 à 2010, des différences d'environ 15% ont été trouvées pour des catégories d'impact comme le potentiel d'appauvrissement abiotique, le potentiel d'acidification et le potentiel de réchauffement global; tandis que des différences jusqu'à 50% ont été calculées pour l'écotoxicité. Ces différences ont été attribuées à l'influence des conditions météorologiques sur la présence des nuisibles de la vigne (Vázquez-Rowe, Villanueva-Rey, Moreira, et al., 2012). Une autre étude qui a analysé neuf systèmes de production dans trois pays européens a montré que l'empreinte carbone d'une bouteille de vin se trouve entre 500 et 2500 g de CO_2 équivalents. Des différences ont été trouvées en fonction de l'appellation et du type de vin, lesquelles sont le résultat de la contribution de plusieurs facteurs tels que le rendement, la quantité, le type des pesticides appliqués, les engrais répandus, le niveau d'optimisation des activités agricoles, ainsi que le temps de vieillissement du vin (Vázquez-Rowe et al., 2013).

2.2.1 L'utilisation de fongicides à base de cuivre en viticulture

En 1863, le *Phylloxéra*, un insecte homoptère, abat près de la moitié du vignoble européen. Pour remédier à ce problème, les vignerons importent des porte-greffes plus résistants à ces insectes. Toutefois, les vignes deviennent vulnérables au champignon *Plasmopara viticola* qui cause la maladie fongique dénommée mildiou. Des conditions humides et chaudes sont particulièrement favorables au développement de cette maladie fongique. En 1884, Alexis Millardet découvre l'effet protecteur du cuivre contre le mildiou en appliquant un mélange de sulfate de cuivre et de chaux. Ce mélange, connu comme *bouillie bordelaise*, est de nos jours largement utilisé en viticulture (Navel, 2011). Cependant, dans quelques pays, l'utilisation de sulfate de cuivre n'est pas recommandée en raison de ses impacts potentiels sur les travailleurs et l'environnement. À la place du sulfate de cuivre, l'hydroxyde de cuivre ($Cu(OH)_2$) et l'oxychlorure de cuivre ($Cu_3Cl_3(OH)_4$), moins concentrés, sont utilisés de manière intensive en viticulture (Mackie et al., 2012).

L'utilisation intensive de fongicides à base de cuivre durant des dizaines d'années a contribué à l'accumulation de ce métal dans les sols viticoles. Alors que la concentration médiane en cuivre dans des utilisations de sols autres que la viticulture est d'environ 11.6 $mg \cdot kg^{-1}$; les sols viticoles ont la concentration médiane la plus élevée en Europe, laquelle est d'environ 26 $mg \cdot kg^{-1}$. De plus, des teneurs supérieures à 100 $mg{\cdot}kg^{-1}$ en cuivre ont été mesurées dans environ 15% de la surface du vignoble européen (Ballabio et al., 2018; Droz et al., 2021). Des concentrations élevées en cuivre peuvent entrainer des problèmes d'écotoxicité terrestre, et de pollution de l'eau souterraine par lixiviation, ainsi que la contamination des eaux superficielles par ruissellement. En outre, la combinaison de la pollution en métaux avec l'érosion et le labourage des sols entraine une réduction de la qualité des sols. Ainsi, beaucoup de vignobles européens se sont donc retrouvés à l'abandon et particulièrement ceux établis sur des pentes raides (Komárek et al., 2010).

En Europe, afin de borner l'accroissement de la teneur en cuivre dans les sols et donc de limiter ses impacts écotoxiques, la dose d'application autorisée est récemment passée de 6 à 4 $kg\ Cu{\cdot}ha^{-1}{\cdot}an^{-1}$ (Droz et al., 2021). Des études se sont aussi penchées sur l'évaluation des solutions de remplacement des fongicides à base de cuivre contre le mildiou. Par exemple, l'effet protecteur des extraits de *Yucca schidigera*, de *Salvia officinalis* et de *Trichoderma harzianum* contre le *Plasmopara viticola* a été évalué. Cependant, ces extraits ont montré une faible action protectrice contre le mildiou, ainsi les fongicides à base de cuivre demeurent les plus efficaces contre ce champignon (Dagostin et al., 2011), et son utilisation est soutenue par son coût de marché relativement faible (Tamm et al., 2022).

Une étude d'ACV analysant l'influence de l'année de la récolte sur le profil environnemental d'une bouteille de vin produite en Galice (Espagne) durant la période de 2007 à 2010 a montré que malgré la variabilité interannuelle, le cuivre est le principal contributeur aux impacts écotoxicologiques (Vázquez-Rowe, Villanueva-Rey, Moreira, et al., 2012). En revanche, dans une étude comparant des systèmes de production conventionnelle, conventionnelle-biodynamique et biodynamique, les auteurs ont décidé de négliger la contribution du cuivre aux impacts écotoxicologiques du fait que le modèle de distribution des pesticides utilisé n'est pas adapté pour des composés inorganiques et deuxièmement parce que la caractérisation des impacts écotoxicologiques des métaux a été jugée trop incertaine (Pedro Villanueva-Rey et al., 2014). Bien que cette limite de l'étude soit communiquée, ceci remet en question la justesse de la comparaison de ces systèmes et peut entraîner de problèmes de crédibilité des résultats d'une ACV.

2.2.2 L'impact du changement climatique sur la filière vitivinicole

Le vignoble mondial se répartit principalement dans les régions comprises entre les isothermes annuelles de 12 à 22 °C pendant le cycle de croissance de la vigne. Cette période de croissance est comprise entre mars et septembre dans l'hémisphère nord, et entre octobre et février dans l'hémisphère sud (Figure 2.1) (Wolkovich et al., 2018). La vigne est particulièrement sensible à des variations de la température, en conséquence, le changement climatique peut occasionner des impacts sur le rendement, la qualité des raisins, et dans un cas plus sévère, la redistribution spatiale des vignobles (Mozell & Thach, 2014). Cependant, les impacts du changement climatique ne correspondent pas uniquement à des projections dans le futur. En fait, des études ont établi un lien entre la précocité des vendanges observée ces dernières décennies dans quelques régions viticoles en France, et plus largement en Europe, avec la variation climatique qui y a été observée (Chuine et al., 2004; Jones et al., 2005).

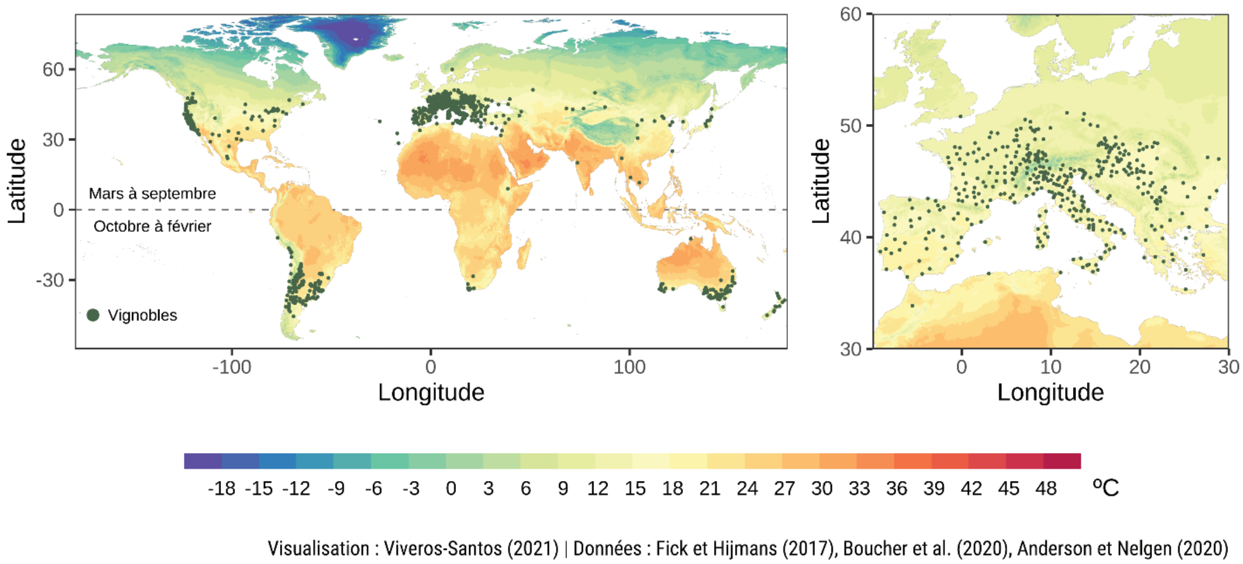


Figure 2.1 Répartition mondiale du vignoble dont l'essentiel est compris entre les isothermes annuelles de 12 °C et 22 °C. Tiré de Viveros Santos (2021).

Les impacts du changement climatique sur le rendement de la vigne seront variables selon les régions viticoles, et ces modifications pourraient pointer vers des baisses, mais aussi vers des hausses du rendement. D'une part, des périodes de sécheresse accrue dans les régions du sud de l'Europe causeront une forte baisse du rendement. En revanche, l'augmentation de la concentration en CO_2 atmosphérique pourrait potentiellement contrebalancer les effets de la sécheresse, ce qui donnerait lieu à une augmentation des rendements en Europe centrale et septentrionale (Helder Fraga et al., 2016). En outre, l'augmentation de la fréquence et de la variabilité des extrêmes climatiques tels que la grêle, le gel de printemps, des sécheresses et des vagues de chaleur pourraient affecter le rendement en viticulture (Leolini et al., 2018; David Santillán et al., 2019)

Les principales modifications de la composition chimique des raisins entrainées par le changement climatique correspondent à l'augmentation de la teneur en sucre, ainsi qu'à la diminution des concentrations en acides, en anthocyanes et en composés organiques volatils (Mozell & Thach, 2014). L'accroissement de la concentration en sucre provoque une augmentation du degré d'alcool, de même que des modifications de l'arôme (Mira de Orduña, 2010). Pour sa part, l'augmentation du pH accélère des réactions d'oxydation qui affectent la couleur, le goût et l'arôme du vin. Pour remédier à la diminution d'acide malique, il existe des options technologiques en vinification, comme l'ajout de l'acide tartrique, ainsi que l'addition de dioxyde de soufre moléculaire pour garantir la stabilité microbienne du vin. Quant à elle, la réduction de la teneur en anthocyanes soulève des problèmes particulièrement dans la vinification des vins rouges, car ces composés sont responsables de la couleur de ce type de vin (Mozell & Thach, 2014).

Le changement climatique peut causer des impacts indirects sur le rendement de la vigne et sur la pratique agricole en modifiant la distribution et l'abondance des insectes nuisibles. Une étude s'est penchée sur la prévision de l'occurrence de mildiou dans une région viticole au nord-ouest de l'Italie aux horizons 2030, 2050 et 2080 à partir de deux modèles de circulation générale et d'un modèle empirique décrivant l'apparition de mildiou. L'étude conclut que sous des conditions climatiques futures, plus chaudes et moins humides, la pression de mildiou augmentera potentiellement, laquelle causerait l'application de deux arrosages supplémentaires de fongicides à base de cuivre par rapport au scénario de référence (Salinari et al., 2006). Une autre étude, portant sur les pressions futures du phalène et de l'oïdium de la vigne dans la région centrale orientale de l'Italie, a utilisé des modèles phénologiques de la vigne et des nuisibles afin d'étudier leurs interactions. L'étude conclut que l'augmentation de la température future accroitra la pression du phalène, mais diminuera potentiellement celle de l'oïdium (Caffarra et al., 2012).

Un des impacts potentiels les plus inquiétants du changement climatique sur la viticulture est la modification de la carte du vignoble mondial. En considérant le scénario RCP8.5 à fortes émissions de GES, une étude a projeté une diminution importante de la surface du vignoble européen d'environ 25% à 73% à l'horizon 2050 (H. Lee et al., 2013). D'autre part, une autre étude a montré que bien que des régions du bassin méditerranéen seraient sévèrement affectées par les changements climatiques projetés, d'autres régions au nord de la France pourraient en bénéficier (M Moriondo et al., 2013). Néanmoins, les études précédentes n'ont pas tenu compte des stratégies d'adaptation qui pourraient atténuer l'impact du changement climatique sur la redistribution des aires viticoles dans le futur, et par voie de conséquence d'assurer la continuité de la production du vin. En effet, une étude a montré que même si la température pendant la période de croissance de la vigne est déjà au-dessus de la limite supérieure du développement optimal pour les principaux cépages dans trois régions viticoles européennes, la production de raisins y est maintenue en raison des modifications des pratiques apportées par les vignerons (van Leeuwen et al., 2013).

2.2.3 Les stratégies d'adaptation de la filière viticole au changement climatique

Comme la plupart des activités anthropiques, les systèmes agricoles génèrent des émissions de GES, mais ils peuvent aussi contribuer à l'atténuation du changement climatique à travers de l'amélioration de la gestion des terres cultivées, de la gestion des pâturages, et de la restauration des sols (IPCC, 2014). Cependant, en raison des impacts potentiels du changement climatique sur la qualité et la productivité des cultures, les systèmes agricoles seront amenés à mettre en place des stratégies d'adaptation au changement climatique projeté. En conséquence, l'évaluation environnementale des stratégies d'adaptation devient nécessaire afin d'éviter des déplacements entre des catégories d'impact ou entre des étapes du cycle de vie et d'empêcher ainsi que les impacts des actions d'adaptation dépassent ceux des actions d'atténuation (Tendall & Gaillard, 2015).

Dans le cas de la filière vitivinicole, les options d'adaptation au changement climatique comprennent à la fois des stratégies concernant la production de raisin et d'autres touchant la vinification. La gestion de la vendange et de la vinification constitue le premier niveau d'adaptation. En effet, les vignerons adaptent fréquemment leurs stratégies de vinification aux conditions du millésime (Quénol, 2014). Donc, ces stratégies d'adaptation seraient plutôt marginales.

En ce qui concerne les stratégies d'adaptation touchant les pratiques en viticulture, elles peuvent être classées selon leur fréquence d'application en pratiques pérennes et annuelles (Tableau 2.1), ou selon leur horizon de temps d'implantation à court ou à long terme. Les pratiques pérennes se réalisent lors de la plantation et compromettent les vignerons pour la durée de vie de la vigne, laquelle peut s'étaler sur deux générations de vignerons. Pour leur part, les pratiques annuelles peuvent être modifiées pour adapter la vigne aux variations climatiques interannuelles. Dans le court terme (moins de 10 ans), ce sont les pratiques annuelles qui seront principalement modifiées. Cependant, dans le moyen (10 à 30 ans) et le long terme (plus de 30 ans), les pratiques pérennes pourraient être affectées par le changement climatique (Quénol, 2014). Toutefois, la mise en place des stratégies d'adaptation dépendra des effets projetés sur chaque région viticole. De plus, selon l'estimation des événements extrêmes climatiques, d'autres options d'adaptation seront nécessaires telles que l'utilisation de filets anti-grêle, ou des ventilateurs et radiateurs contre le gel printanier, ainsi que l'implantation des systèmes d'irrigation pour contrer la sècheresse (Sacchelli et al., 2017).

Tableau 2.1 Options d'adaptation de viticulture au changement climatique

Pratiques pérennes	**Pratiques annuelles**
• Le choix du terrain (site, orientation, profondeur de sol, réserve hydrique) • Le choix du porte-greffe (lequel conditionne l'adaptation de la vigne au type de sol, selon la profondeur, humidité, teneur en calcaire actif) • Le choix de la variété • La date de plantation • Le système de conduite et la densité de plantation.	• La taille • L'entretien du sol (désherbage, travail du sol, paillage) • L'irrigation (volumes, rythme) • L'entretien de la partie aérienne (ébourgeonnage, rognage, éclaircissage, protection phytosanitaire).

2.3 L'analyse du cycle de vie

L'analyse du cycle de vie (ACV) est une méthodologie qui vise à évaluer les impacts environnementaux potentiels d'un produit, d'un procédé ou d'un service pour l'ensemble de son cycle de vie (ISO, 2006a). Du fait de sa nature holistique, l'ACV est utilisée dans la définition de stratégies d'amélioration de systèmes de produits, afin d'éviter le transfert d'impacts entre des étapes du cycle de vie ou entre des catégories d'impact (Stefanie & Llorenç, 2014). Bien que l'approche holistique de l'ACV soit son principal atout, cet attribut constitue à la fois une faiblesse. En effet, les résolutions spatiale et temporelle de l'ACV traditionnelle sont peu développées du fait qu'à l'origine cette méthodologie était « générique », car peu ou aucune information concernant le lieu et le moment d'émission était considérée (de Haes et al., 2004). Pour cette raison, au cours des dernières années, des travaux ont porté sur la prise en compte des dimensions spatiale et temporelle en ACV afin d'améliorer l'évaluation des impacts environnementaux et de diminuer l'incertitude des résultats.

2.3.1 Description générale de la méthodologie

L'ACV est une méthodologie itérative qui se réalise en quatre phases : 1) définition des objectifs et du champ de l'étude, 2) analyse de l'inventaire, 3) évaluation des impacts du cycle de vie et 4) interprétation (ISO, 2006a) (Figure 2.2).

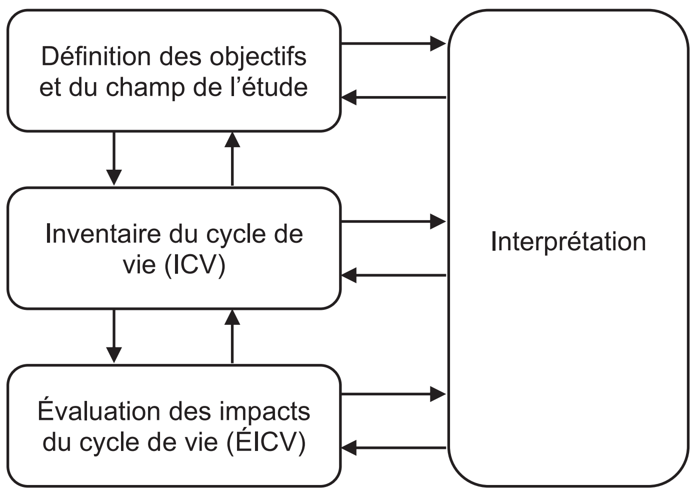


Figure 2.2 Étapes constituantes de l'ACV, adaptée de ISO (2006a)

L'étape de définition des objectifs et du champ de l'étude correspond à l'identification de la fonction étudiée et à la définition de la base de comparaison, aussi appelée unité fonctionnelle, sur laquelle le produit ou service sera analysé ou comparé dans le cas d'une ACV comparative. En outre, les frontières du système sont définies à ce stade.

L'analyse de l'inventaire est dédiée à la collecte et au traitement des flux de matières associés aux systèmes de produit analysés. Ces flux de matière se déclinent en flux élémentaires et flux économiques. Les flux élémentaires sont aussi appelés interventions environnementales, car ils correspondent à des échanges avec la biosphère. Ces flux incluent l'extraction de ressources primaires et l'émission de polluants, tandis que les flux économiques correspondent à des échanges avec la technosphère sous forme d'intrants ou de sortants.

La troisième étape de l'ACV est l'évaluation des impacts du cycle de vie (ÉICV) (Figure 2.2), laquelle est destinée au calcul des impacts environnementaux potentiels causés par les flux élémentaires associés au cycle de vie du produit ou service étudié. La conversion des flux élémentaires en des scores d'impact requiert de l'utilisation de facteurs de caractérisation qui sont obtenus en utilisant des modèles d'ÉICV décrivant des chaînes de cause à effet. Dans le cadre de cette thèse, le développement de facteurs de caractérisation est d'intérêt prépondérant, et plus particulièrement ceux pour l'évaluation de l'écotoxicité terrestre causée par le cuivre.

L'interprétation consiste à fournir une présentation compréhensible, complète et cohérente des résultats d'une ACV, ceci entraine un processus itératif de révision du champ de l'étude, de la qualité des données ainsi que de la méthodologie d'ÉICV choisie.

2.3.2 Dimension spatiale en ACV : du monde unitaire à la régionalisation des impacts

Au cours du cycle de vie d'un système de produits, les émissions qui y sont associées peuvent être générées et dispersées dans plusieurs régions et même dans différents pays. L'ampleur de l'impact environnemental des émissions dépend des propriétés physicochimiques des polluants, ainsi que du milieu d'émission et de réception. En conséquence, l'impact potentiel de ces émissions peut être local, régional, continental ou global (Hauschild, 2006; Mutel & Hellweg, 2009; Y. Yang, 2016). Néanmoins, lors de l'analyse de l'inventaire, les flux élémentaires du même type sont agrégés dans l'espace et dans le temps, et en conséquence, les informations sur le moment et le lieu d'émission sont perdues. Ultérieurement, pendant la phase d'ÉICV, les interventions environnementales sont traduites en des scores d'impact à l'aide de facteurs de caractérisation. Historiquement, ces facteurs étaient axés sur les propriétés physicochimiques des substances, en supposant que la complexité et la variabilité de l'environnement pouvaient être représentées par un « monde unitaire » moyen. Ceci impliquait que peu importe l'endroit d'émission et de réception, le facteur de caractérisation à utiliser pour évaluer l'impact potentiel d'un flux élémentaire donné était le même (Hauschild, 2006). Cependant, cette hypothèse du « monde unitaire » n'est pas valable dans le cas des catégories dont l'ampleur de l'impact est fortement influencée par la variabilité de l'environnement. C'est le cas de l'écotoxicité terrestre des métaux qui est influencé par la spéciation de ces substances en fonctions des propriétés des sols (Section 2.4.1.1).

L'inclusion de la dimension spatiale, ou régionalisation, constitue à la fois un défi et une opportunité en ACV, car il est reconnu que son intégration permet d'améliorer sa représentativité et fiabilité auprès des décideurs (Stefanie & Llorenç, 2014; Y. Yang, 2016; Y. Yang & Heijungs, 2016). La régionalisation en ACV implique la régionalisation de l'inventaire et de l'ÉICV. Dans la phase d'inventaire, il est nécessaire de régionaliser les processus afin d'améliorer leur représentativité pour une région donnée, et de spatialiser les flux élémentaires en indiquant le lieu d'émission avec le plus de détails possible. De plus, il est nécessaire que les méthodologies d'ÉICV développent des facteurs de caractérisation régionalisés qui tiennent compte de l'influence du lieu d'émission et de réception sur l'ampleur des impacts environnementaux potentiels (Patouillard et al., 2020). Ainsi, la méthode d'ÉICV régionalisée à l'échelle mondiale IMPACT World+ a été développée afin de combler ce dernier besoin en ACV (Bulle et al., 2019).

2.3.3 Dimension temporelle en ACV

Le cycle de vie d'un produit comporte, généralement, des étapes qui se réalisent à des moments différents et s'étalent sur des échelles de temps distinctes. Pourtant, cette variation dans le temps n'est pas prise en compte dans l'ACV traditionnelle (Beloin-Saint-Pierre et al., 2020; de Haes et al., 2004). Dans la phase d'inventaire, les interventions environnementales de la même catégorie sont agrégées dans un flux élémentaire ponctuel, ce qui implique de considérer que les impacts environnementaux et les interventions environnementales se produisent simultanément (Beloin-Saint-Pierre et al., 2020). En conséquence, des facteurs comme l'accumulation de polluants, des effets tampons, des seuils d'effet, l'interaction de polluants, et la dynamique environnementale sont négligés en ÉICV.

L'ÉICV est réalisée selon une approche marginale qui considère que l'émission de polluants entraine des perturbations faibles dans une condition environnementale *ceteris paribus* dont la réponse est linéaire. Cette hypothèse permet de négliger le moment d'émission et les variations des conditions environnementales au fil du temps (Mark A J Huijbregts et al., 2011; Reap et al., 2008). Toutefois, des effets saisonniers et la présence d'autres composés chimiques dans l'environnement peuvent modifier l'ampleur des impacts. Par exemple, une étude portant sur la caractérisation de l'oxydation photochimique a montré que la variation saisonnière s'avère plus influente par rapport au lieu d'émission pour cette catégorie d'impact (Shah & Ries, 2009).

En raison de l'incertitude associée au manque de résolution temporelle de l'ACV traditionnelle, ces dernières années, différentes approches ont été développées afin d'y intégrer différents aspects temporels qui portent sur le champ d'application temporelle, la dynamique des systèmes et l'augmentation de la représentativité temporelle (Beloin-Saint-Pierre et al., 2020). Notamment, le concept d'ACV dynamique a été introduit afin de tenir compte de la dynamique des systèmes étudiés, ainsi que de la différentiation temporelle des flux élémentaires (Levasseur et al., 2010). De plus, le choix de l'horizon de temps pour le calcul des facteurs de caractérisation a été analysé. En raison du principe d'équité intergénérationnelle, la plupart de méthodes d'ÉICV considèrent une intégration à l'infini de l'impact des émissions toxiques. Le choix d'un horizon de temps court permet d'évaluer les impacts à court terme, mais néglige potentiellement des impacts futurs (Reap et al., 2008). Dans le cas de la catégorie de réchauffement climatique, la sélection d'un horizon de temps est une des étapes critiques de l'évaluation de l'empreinte carbone. En effet, l'utilisation d'un horizon de temps court implique que les impacts les plus proches dans le temps ont plus d'importance, car les impacts générés après l'horizon de temps choisi sont exclus de l'analyse (Levasseur et al., 2010).

Une autre considération temporelle correspond au développement de l'ACV prospective. En effet, il existe un intérêt croissant dans l'évaluation de scénarios prospectifs à l'aide de l'ACV afin d'anticiper les impacts environnementaux potentiels de nouvelles technologies et produits (Bisinella et al., 2021; Sacchi et al., 2022), ainsi que dans l'évaluation des systèmes de produits dans des conditions environnementales futures modifiées par le changement climatique (Sala et al., 2017). Plus particulièrement, comme les systèmes agricoles sont fortement dépendants du climat, des études ont porté sur l'impact du changement climatique projeté sur la performance environnementale de certaines cultures, comme le maïs, le soja, et l'orge de printemps (Cosme & Niero, 2017; Garba et al., 2014; E. K. Lee et al., 2020). Ces études ont simulé l'impact du changement climatique dans l'inventaire du cycle de vie en considérant des modifications du rendement des cultures entrainées par les changements des variables climatiques, ainsi que dans les pratiques agricoles telles que le taux d'application de pesticides et des engrais. En outre, il est projeté que le devenir, l'exposition et les effets des polluants seront modifiés dans des conditions environnementales futures altérées par le changement climatique (Section 2.4.2). À ce sujet, des travaux ont porté sur le calcul de facteurs de caractérisation pour leur application dans l'évaluation de scénarios futurs. Spécifiquement, des facteurs de caractérisation ont été développés pour évaluer les impacts liés à l'utilisation de l'eau à court et moyen terme en Espagne (Núñez et al., 2015). De plus, des facteurs de caractérisation pour l'eutrophisation marine ont été paramétrés afin d'intégrer l'influence des conditions environnementales futures prédites par des scénarios de changement climatiques sur le devenir, l'exposition et l'effet des émissions d'azote (Cosme & Niero, 2017).

2.4 Catégorie d'impact « écotoxicité terrestre »

En ACV, l'indicateur d'écotoxicité sert à quantifier les impacts environnementaux potentiels des polluants chez des êtres vivants à part l'homme (P. Fantke et al., 2018). Cette catégorie est divisée selon l'écosystème affecté en écotoxicité terrestre, écotoxicité aquatique et écotoxicité marine, si des organismes du sol, de l'eau douce ou de la mer sont respectivement affectés. Ces catégories d'impact sont modélisées selon leur respective chaîne de cause à effet, laquelle constitue le cadre méthodologique pour calculer des facteurs de caractérisation utilisés lors de la phase d'ÉICV.

Les facteurs de caractérisation pour l'écotoxicité terrestre sont calculés selon la chaîne de cause à effet montrée à la Figure 2.3. Cette chaîne de cause à effet inclut :1) la modélisation du devenir, 2) la modélisation de l'exposition, 3) la modélisation de l'effet, et 4) la modélisation du dommage (EC-JRC, 2010; Rosenbaum, 2015). Cependant, l'inclusion de la modélisation de l'exposition est relativement récente, et généralement, les facteurs de caractérisation pour l'écotoxicité terrestre étaient calculés comme le produit d'un facteur de devenir (FF) et d'un facteur d'effet (EF).

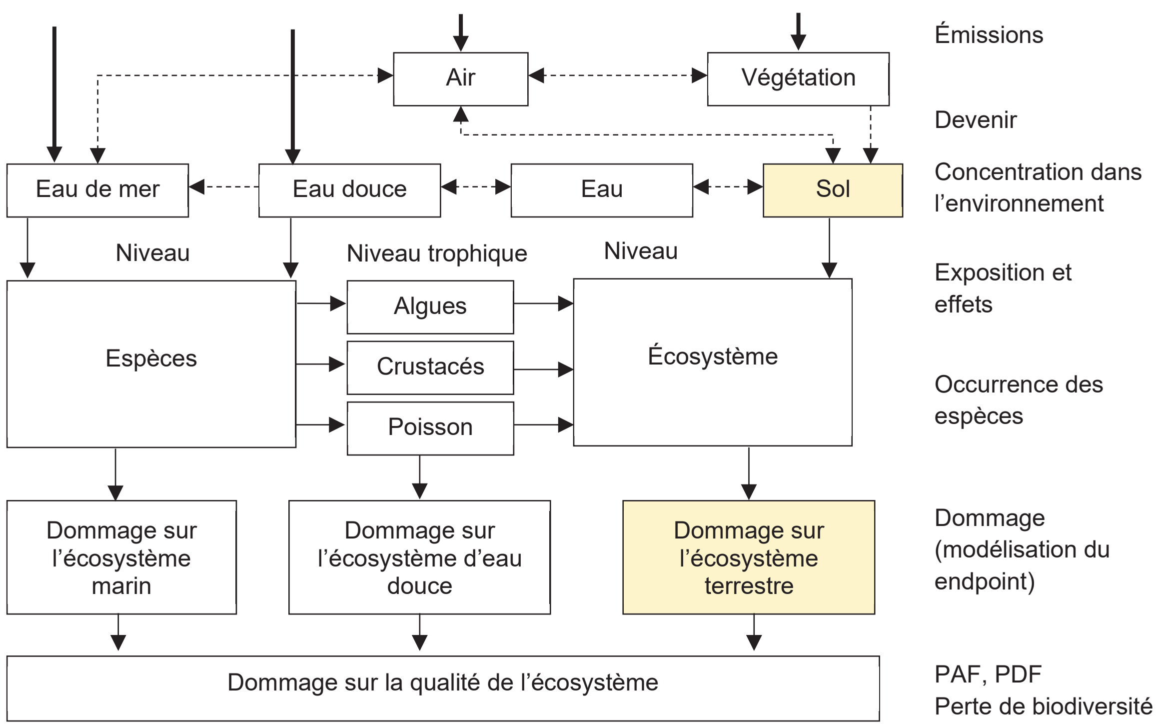


Figure 2.3 Chaine de cause à effet de l'écotoxicité. Adapté de EC-JRC (2010).

Le facteur de devenir correspond à la fraction de la substance transférée du compartiment d'émission au compartiment de réception. Ce facteur est calculé à l'aide de modèles de devenir qui estiment la distribution d'une substance dans l'environnement. Les modèles de devenir sont fondés sur des bilans de masse et incluent de processus de partition, de diffusion, de sorption, d'advection, de convection, de dégradation et de séquestration (dans des sédiments) (Rosenbaum et al., 2008). Parmi les modèles de devenir, USEtox est recommandé par l'Initiative du Cycle de Vie des Nations Unies. D'ailleurs, USEtox est le résultat de la collaboration entre les développeurs des modèles de devenir tels que Caltox, USES LCA, IMPACT 2002 et EDIP (P. Fantke et al., 2018).

Le facteur d'exposition tient compte du fait que seulement une fraction de la quantité totale d'une substance présente dans un compartiment environnemental donné entraîne des effets toxiques. Cette fraction est appelée « biodisponible ». En effet, plusieurs phénomènes peuvent modifier la fraction biodisponible d'une substance : la dissolution, la précipitation, l'adsorption et la spéciation (Rosenbaum, 2015). Dans USEtox, l'exposition est évaluée en fonction de la fraction dissoute (Henderson et al., 2011; Rosenbaum et al., 2008). D'autres études ont inclus l'exposition dans le calcul des facteurs de caractérisation à travers l'introduction d'un facteur de biodisponibilité pour les catégories d'écotoxicité aquatique (Diamond et al., 2010) et d'écotoxicité terrestre (Owsianiak et al., 2013; Plouffe et al., 2015a).

Le facteur d'effet, quant à lui, quantifie l'impact d'une substance sur un organisme par concentration d'exposition (Pennington et al., 2006). Pour l'écotoxicité terrestre, le manque de données peut être comblé par extrapolation des facteurs d'effet des organismes aquatiques en utilisant des coefficients de partition sol-eau (Kd) (Haye et al., 2007). Cependant, dans le cas des métaux, il a été montré que cette méthode d'extrapolation basée sur des Kd ne permet pas de modéliser adéquatement l'effet chez des organismes terrestres, et l'utilisation des modèles TBLM (*Terrestrial biotic ligand model*) a été proposée à la place (Tromson et al., 2017). Les méthodes d'impact IMPACT 2002+ et USEtox utilisent la méthode AMI (*Assessment of mean impact*) pour calculer le facteur d'effet d'une substance. Cette méthode utilise la moyenne géométrique ($HC50_{EC50}$) des EC_{50}, c'est-à-dire des concentrations induisant un effet chez 50% de la population d'une espèce donnée. Les données utilisées doivent couvrir au moins trois niveaux trophiques afin de tenir compte de la variabilité biologique de l'écosystème. La raison principale de l'utilisation de $HC50_{EC50}$ est d'éviter des biais dans la comparaison des polluants, car elle est moins sensible à des valeurs extrêmes de toxicité (faibles EC_{50}), et fournit donc une meilleure estimation de l'effet moyen d'une substance (P. Fantke et al., 2018). Finalement, le facteur d'effet ($PAF \cdot kg^{-1}$) est calculé en divisant 0.5 par la $HC50_{EC50}$. Ce facteur 0.5 correspond au point sur la courbe de sensibilité des espèces où 50% des espèces sont potentiellement affectées (Henderson et al., 2011).

2.4.1 Évaluation des impacts écotoxiques terrestres des métaux

Les métaux se trouvent de façon naturelle dans les sols normalement à des concentrations faibles qui varient en fonction de la roche mère, ainsi que des processus d'altération météorique (Cabral Pinto et al., 2017). Toutefois, des interventions anthropiques comme l'exploitation minière et l'utilisation des pesticides et des engrais en agriculture ont contribué à l'augmentation de la concentration en métaux dans les sols. Cet accroissement de la concentration en métaux dans les sols cause des impacts écotoxiques terrestres, de même que des impacts aquatiques à cause de la lixiviation et du ruissellement de ces substances. De plus, la consommation de l'eau polluée par lesdits processus peut entrainer des impacts toxiques (Ardestani et al., 2014; Mackie et al., 2012; Watmough & Orlovskaya, 2015).

Néanmoins, l'évaluation des impacts (éco)-toxiques des métaux constitue un défi en raison de leurs caractéristiques : ils se trouvent naturellement dans l'environnement, ils forment des mélanges dans le sol et leur effet est modifié par les conditions de l'environnement. En outre, chaque métal a un mode d'action qui dépend de ses propriétés et de la sensibilité des organismes exposés (Ardestani et al., 2014; Mackie et al., 2013). Quelques métaux, comme le fer, le cuivre, le manganèse, le zinc, le chrome et le cobalt sont essentiels à de faibles concentrations pour certains organismes. En conséquence, une carence en ces métaux peut engendrer des effets nocifs. Cependant, un excédent des métaux essentiels entraine aussi des effets néfastes chez des organismes (Ardestani et al., 2014). D'autre part, des métaux qui ne sont pas essentiels au bon fonctionnement des organismes, comme le mercure, le plomb, le cadmium et l'aluminium, sont toxiques à toute concentration d'exposition (Reiley, 2007).

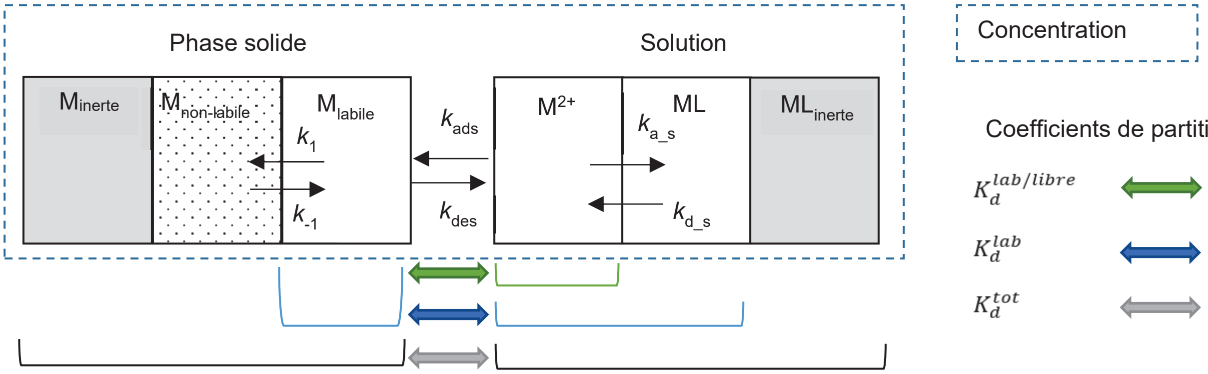
En ACV, les métaux font partie des substances pour lesquelles le facteur de caractérisation pour l'écotoxicité est très élevé relativement aux composés organiques. En effet, il a été montré que malgré les différences dans les modèles de caractérisation, les métaux ressortaient comme les principaux contributeurs aux impacts écotoxiques selon une comparaison menée sur huit méthodes d'ÉICV (Pizzol et al., 2011). Néanmoins, ces facteurs de caractérisation étaient déterminés en fonction de la concentration totale du métal, sans tenir compte des processus qui modifient leur biodisponibilité, tels que la spéciation. Ainsi, depuis le consensus d'un groupe de travail de la

UNEP/SETAC réuni à Clearwater qui visait à améliorer l'évaluation des impacts des métaux en milieu aquatique, d'autres travaux ont porté sur l'amélioration de la caractérisation des impacts écotoxiques des métaux. Ces travaux pionniers ont porté sur l'introduction d'un facteur de biodisponibilité dans le calcul des facteurs de caractérisation pour l'écotoxicité aquatique afin de tenir compte de la spéciation des métaux en fonction de la variabilité des milieux aquatiques (Diamond et al., 2010). Ultérieurement, d'autres études se sont penchées sur l'écotoxicité terrestre (Owsianiak et al., 2013; Plouffe et al., 2016).

2.4.1.1 La spéciation des métaux dans le sol : un processus qui modifie l'ampleur de leur impact écotoxique

À la différence des composés organiques, les métaux ne se dégradent pas dans le sol et sont donc potentiellement persistants à l'infini dans ce compartiment environnemental (M A J Huijbregts et al., 2001). Cependant, des facteurs tels que le pH de la solution du sol, la teneur en matière organique, la capacité d'échange cationique, l'humidité et le niveau d'aération du sol modifient l'impact écotoxique des métaux selon un processus de spéciation. Ce processus correspond à la distinction entre les différentes formes chimiques possibles du métal dans le sol (Plouffe et al., 2015a). Comme l'illustre la Figure 2.4, en fonction de leur spéciation, les métaux se trouvent dans la phase solide du sol ou dans la solution du sol. En effet, les propriétés physicochimiques du sol déterminent des processus plus ou moins réversibles qui déterminent la formation des formes organiques ou inorganiques des métaux (Degryse et al., 2009).

La distribution des métaux dans la phase solide et dans la solution du sol, tel que schématisé à la Figure 2.4, est déterminée expérimentalement à partir des extractions séquentielles. Ces extractions consistent à mettre en contact le sol avec des agents d'extraction de plus en plus forts qui sont censés attaquer des phases solides spécifiques (Degryse et al., 2009; Sauvé et al., 2000). La mobilité du métal est exprimée en fonction de coefficients de partition. Le K_d^{tot} ou coefficient de partition total sol-eau, correspond au ratio de la concentration du métal dans la phase solide divisée par sa concentration dans la solution du sol, ce coefficient est largement utilisé dans l'évaluation des risques (Sauvé et al., 2000) et dans le calcul des facteurs de devenir avec la méthode USEtox (Rosenbaum et al., 2008), mais il est possible de définir d'autres coefficients de partition comme le montre la Figure 2.4.



k_{ads} : constante de vitesse d'adsorption ; k_{des} : constante de vitesse de désorption; k_1 et k_{-1}: constantes de vitesse de transfert entre la phase labile et non-labile dans la phase solide ; k_{a_s} et k_{d_s} : constantes de vitesse de transfert entre la fraction des ions libres et la fraction des complexes dans la solution.

Figure 2.4 Spéciation et partition d'un métal dans le sol. Adapté de (Degryse et al., 2009)

Le pH de la solution du sol est un des facteurs clés influençant la solubilité des métaux (Sauvé et al., 2000). Dans le cas du cuivre, celui-ci est plus mobile à des valeurs de pH inférieures à 6; ainsi, dans des vignobles de sols acides, le cuivre peut migrer à travers la matrice du sol et contaminer les eaux souterraines (Fernández-Calviño et al., 2008). Néanmoins, la mobilité du cuivre peut aussi augmenter à des valeurs de pH supérieures à 7,5 à cause de la solubilisation de la matière organique et de la formation de complexes Cu-MO dissous (Arias et al., 2006). Un autre facteur clé influençant la mobilité des métaux dans le sol est la teneur en matière organique; celle-ci génère des interactions complexes à cause de la dualité de ses effets (Sauvé et al., 2000). D'un côté, la matière organique associée à la phase solide (composés humiques) interagit avec les métaux et diminue leur mobilité. D'un autre côté, la matière organique dissoute permet la formation de complexes organiques solubles (Cu-MO) et augmente la biodisponibilité des métaux (Degryse et al., 2009).

La spéciation détermine l'impact écotoxique d'un métal, car ce phénomène influence sa mobilité, son devenir environnemental et sa disponibilité pour les organismes (Ardestani et al., 2014). Dans le cas du cuivre, par exemple, la forme libre Cu^{2+} est la plus toxique, suivie par les hydroxydes de cuivre : $Cu(OH)^+$/$Cu(OH)_2$; les carbonates de cuivre $CuCO_3$, $CuHCO_3^+$, $Cu(CO_3)_2^{2-}$; finalement, les espèces les moins toxiques sont les complexes chlorés ($CuCl_2$) (Komárek et al., 2010).

Il existe deux modèles principaux qui tiennent compte de la spéciation pour étudier le niveau de toxicité des métaux. Le modèle de l'activité de l'ion libre (FIAM pour « *free ion activity model* ») considère que la toxicité des métaux est déterminée par l'absorption d'espèces métalliques spécifiques à l'interface organisme-eau et que la concentration en ions libres dans la solution du sol est un meilleur indicateur de la toxicité par rapport à la concentration totale dissoute du métal (Ardestani et al., 2014). Pour sa part, le modèle du ligand biotique adapté aux sols (TBLM pour « *terrestrial biotic ligand model* ») suppose que le métal dans le sol est en équilibre avec le métal dans la solution de sol et donc que la toxicité est déterminée par les ions libres dans la solution du sol qui se fixent sur un ligand, qui est la surface exposée d'un organisme. De plus, le modèle TBLM tient compte de la compétition entre l'ion libre et des cations comme le Ca^{2+}, le Mg^{2+}, le Na^{+}, le K^{+} et le H^{+}, car cette compétition peut entraîner une réduction de la toxicité métallique (Di Toro et al., 2001; Thakali et al., 2006).

2.4.1.2 Des avancements récents dans l'évaluation de l'écotoxicité terrestre des métaux

Deux approches méthodologiques ont été développées pour tenir compte de la spéciation des métaux dans le calcul des facteurs de caractérisation pour l'écotoxicité terrestre. Une de ces approches a utilisé des régressions empiriques (Owsianiak et al., 2013), tandis que l'autre a employé un modèle géochimique de spéciation (Plouffe et al., 2015a, 2016). Les régressions empiriques permettent de calculer les différentes fractions de métal dans le sol, notamment les fractions dissoutes, réactives et libres en fonction des propriétés physicochimiques des sols. Les modèles géochimiques aussi appelés modèles multi-surfaces, combinent des modèles thermodynamiques de spéciation inorganique, avec des modèles de complexation de surface (c'est-à-dire d'interaction entre l'ion et des surfaces réactives), ainsi que des modèles de compétition cationique. Particulièrement, les modèles NICA-Donnan et WHAM sont les modèles géochimiques les plus utilisés dans l'étude de la spéciation des métaux dans l'eau et dans le sol (Jan E Groenenberg et al., 2012).

La méthode basée sur l'utilisation de régressions empiriques propose l'introduction d'un facteur de biodisponibilité (BF) et d'un facteur d'accessibilité (ACF) dans le calcul des facteurs de caractérisation afin d'y intégrer la spéciation du métal. Le facteur d'accessibilité correspond à la fraction réactive de la quantité totale de métal dans le sol, c'est-à-dire, la fraction accessible susceptible de provoquer des effets écotoxiques. Le facteur de biodisponibilité quant à lui représente la fraction d'ions libres de la quantité réactive. Les facteurs de devenir ont été calculés avec USEtox en utilisant les K_d des sols obtenus à partir des régressions linéaires. À leur tour, des facteurs d'effet ont été calculés en utilisant le modèle TBLM. Des facteurs de caractérisation pour le Cu et le Ni ont été obtenus pour un ensemble de 760 sols à travers le monde. Ces facteurs de caractérisation s'étendent sur 3,5 et 3 ordres de grandeur, respectivement (Owsianiak et al., 2013). Finalement, l'étude a permis de développer des régressions permettant de calculer des facteurs de caractérisation pour le Cu et le Ni en fonction du pH, de la teneur en carbone organique, de la concentration de cations Mg^{2+} dans la solution du sol et du pourcentage en argile du sol. Ces régressions, qui ont été développées notamment à l'aide des modèles TBLM, ont l'avantage d'être très faciles à mettre en œuvre avec peu d'information et avec un temps de calcul très rapide. Cependant, il faut souligner que leur validité se limite aux sols non calcaires puisque les modèles TBLM ne sont pas valides pour les sols calcaires.

Plouffe et al., (2015a) ont calculé des facteurs de biodisponibilité du Zn pour l'ensemble des sols de la planète en utilisant le modèle WHAM 6.0, les concentrations de fond du métal et les propriétés des sols répertoriées dans la *Harmonized World Soil Database* (HWSD). Les K_d obtenus avec le modèle WHAM 6.0 ont été utilisés dans le calcul des facteurs de devenir avec USEtox, ensuite la méthode AMI a été utilisée dans le calcul des facteurs d'effet. Les facteurs de caractérisation rapportés pour l'ensemble du monde s'étendent sur 1,76 ordre de grandeur lorsque la fraction soluble du Zn est considérée comme la fraction biodisponible. Néanmoins, lorsque la solution vraie du Zn est considérée comme la fraction biodisponible, les facteurs de caractérisation s'étalent sur 14 ordres de grandeur. Les facteurs de caractérisation agrégés en fonction de la fraction soluble et de la solution vraie du Zn sont respectivement 28 et 88 fois inférieurs au facteur de caractérisation de la méthode IMPACT 2002 (Plouffe et al., 2016).

Malgré les différences dans les choix méthodologiques des deux approches décrites précédemment pour intégrer la spéciation des métaux dans le calcul des facteurs de caractérisation pour l'écotoxicité terrestre (Owsianiak et al., 2013; Plouffe et al., 2016), ces travaux ont montré la pertinence d'inclure ce processus et ont montré à la fois l'influence de la variabilité spatiale des sols pour cette catégorie d'impact.

L'évaluation de l'écotoxicité terrestre devient plus compliquée si l'on considère que le sol est un milieu complexe et dynamique puisqu'il peut subir des variations d'humidité, de pH, des conditions redox ainsi que des changements des concentrations en matière organique, ou d'autres processus tels que l'érosion du sol. Ainsi, dans ce milieu, des métaux peuvent réagir de manière différente au cours du temps. Une étude a analysé l'influence du vieillissement sur la dynamique des métaux dans le sol et son intégration dans le calcul des facteurs de caractérisation dynamiques. Le phénomène de vieillissement a été modélisé en fonction de la fraction inerte dans la phase solide et celle dans la solution du sol (M_{inerte} et ML_{inerte}, respectivement dans la Figure 2.4). Ces fractions ont été intégrées aux facteurs d'accessibilité et de devenir à l'aide de régressions empiriques. Toutefois, les résultats de cette étude montrent que la prise en compte du vieillissement n'est pas concluante en raison de l'incertitude sur le moment d'émission des métaux (Owsianiak et al., 2015).

Comme il a été mentionné à la section 2.1, il est projeté que l'influence du changement climatique sur l'environnement modifiera l'ampleur des impacts écotoxiques des polluants. En conséquence, il devient nécessaire de considérer cet aspect temporel dans le calcul des facteurs de caractérisation pour l'évaluation des scénarios prospectifs. On qualifie de « futurs » ces facteurs de caractérisation parce qu'ils sont développés en fonction des conditions environnementales futures prédites par des scénarios de GES, par exemple, à un horizon de temps donné.

2.4.2 Influence du changement climatique sur l'impact écotoxique des polluants

Les êtres vivants sont exposés à plusieurs facteurs de stress chimiques, physiques et biologiques, lesquels sont issus principalement des activités anthropiques, bien que des sources naturelles puissent aussi y contribuer. Le changement climatique est un facteur de stress qui peut modifier simultanément le devenir environnemental et l'effet toxique des substances chimiques (Kimberly & Salice, 2014; Noyes & Lema, 2015). En effet, un groupe de travail de la SETAC s'est attaqué à la question suivante : « Comment le changement climatique et d'autres facteurs de stress influenceront-ils les impacts environnementaux des substances chimiques? » Ce groupe de travail a conclu que les activités humaines, incluant celles d'atténuation et d'adaptation au changement climatique, peuvent modifier l'occurrence et le devenir des polluants dans l'environnement. En outre, le changement climatique peut affecter la toxicité de ces substances, et à l'inverse, l'exposition à des polluants peut réduire la capacité d'acclimatation des organismes aux changements climatiques projetés (Stahl Jr. et al., 2013). En conséquence, dans le contexte du changement climatique, l'analyse environnementale des émissions de polluants devrait tenir compte de l'interaction de plusieurs facteurs de stress.

2.4.2.1 L'impact sur le devenir des polluants

Le changement climatique peut affecter le devenir des substances toxiques dans l'environnement à cause des modifications des propriétés physiques, chimiques et biologiques influençant la partition des polluants entre l'atmosphère, l'eau, les sols, les sédiments et le biote. De plus, des processus de transfert et de dégradation peuvent être affectés, tels que l'échange air-surface, la déposition humide et sèche, ainsi que les vitesses de réaction de photolyse, de biodégradation, et d'oxydation dans l'air (Gouin et al., 2013; Noyes & Lema, 2015). Des modifications de la température moyenne et des régimes de précipitation auront des impacts importants sur la distribution des polluants dans l'environnement (Gouin et al., 2013; Noyes & Lema, 2015; Stahl Jr. et al., 2013). Par exemple, l'augmentation de la température peut entrainer le relargage de composés organiques persistants (POPs) qui étaient préalablement stockés dans la glace, la neige et les sols (Gouin et al., 2013; Noyes & Lema, 2015).

Dans le contexte du changement climatique, le sort des métaux peut être aussi affecté à cause des modifications de la teneur en matière organique, du pH, de la salinité et de l'humidité des sols, car ces propriétés physico-chimiques déterminent la spéciation de ces substances, et en conséquence ces changements affectent la mobilité, la biotransformation, la biodisponibilité et la toxicité des métaux (Noyes & Lema, 2015). De plus, le ruissellement des pesticides, des engrais et d'autres produits utilisés en agriculture peut augmenter à cause d'un accroissement de la fréquence des précipitations ou de l'érosion du sol, entrainant la contamination des eaux de surface (Noyes et al., 2009). Par exemple, une étude simulant l'impact du changement climatique sur les propriétés du sol a montré que des modifications faibles de l'ordre de 5 à 20% des variables hydrologiques, telles que l'humidité du sol, la recharge des nappes phréatiques et la conductivité hydraulique, peuvent engendrer une augmentation de la mobilité du cadmium d'environ 27% (Augustsson et al., 2011).

Le sort de polluants est modélisé à l'aide des modèles multimédias. Ces modèles incluent des paramètres comme les dimensions des milieux environnementaux, la température moyenne, la configuration de la circulation atmosphérique, des mécanismes de transfert et de dégradation, ainsi que des propriétés physicochimiques des compartiments environnementaux. D'après Gouin et al., (2013), la structure de la plupart des modèles multimédia permet d'analyser l'influence du changement climatique sur le sort des polluants. L'approche consiste à définir d'abord une paramétrisation de référence et une deuxième correspondant à un scénario futur. La paramétrisation des scénarios futurs est basée sur des résultats des modèles de circulation générale ainsi que des projections d'autres propriétés de l'environnement altérées par le changement climatique.

2.4.2.2 L'impact combiné du changement climatique et des substances toxiques

Le changement climatique peut entrainer des impacts directs et indirects sur les êtres vivants. Des impacts directs correspondent par exemple à des changements des distributions et des migrations des espèces, ainsi que sur leur capacité d'acclimatation; tandis que des impacts indirects découlent de l'interaction du changement climatique avec d'autres facteurs de stress, tels que la présence de polluants dans l'environnement. Des variations des conditions environnementales peuvent altérer l'exposition à des polluants de façon directe, en augmentant l'utilisation de pesticides, par exemple (flèche 1, Figure 2.5), ou en modifiant le devenir des polluants (flèche 2, Figure 2.5). Deux hypothèses principales ont été posées pour expliquer l'impact toxique entrainé par l'interaction entre le changement climatique et les substances toxiques : des facteurs de stress associés à des conditions climatiques rendent les organismes plus vulnérables à l'exposition des substances toxiques (hypothèse CITS pour « *Climate-induced toxicant sensitivity* » , flèche 3, Figure 2.5), et l'exposition à des substances toxiques diminue la capacité des organismes à s'acclimater à des conditions climatiques modifiées (hypothèse TICS pour « *Toxicant-induced climate sensitivity* », Flèche 4, Figure 2.5) (Hooper et al., 2013; Moe et al., 2013).

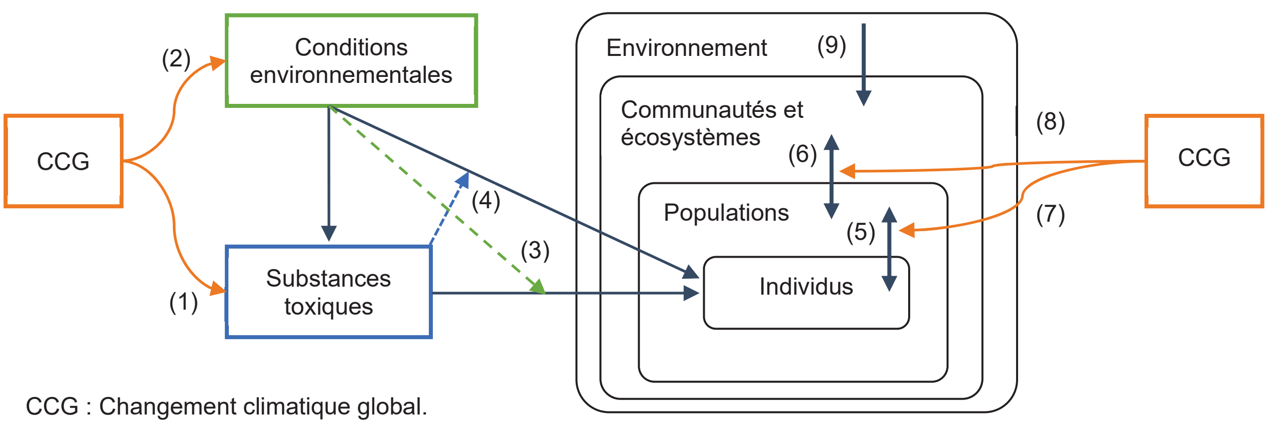


Figure 2.5 Effets combinés et interactifs du changement climatique et des substances toxiques sur différents niveaux d'organisation biologique. Adapté de Moe et al. (2013).

Une série de trois études a porté sur l'analyse de l'influence du changement climatique sur l'impact toxique de métaux chez l'invertébré terrestre *Enchytraeus crypticus*. Dans ces études, des conditions futures ont été simulées en augmentant la température ambiante de 20 °C à 25 °C et en diminuant l'humidité du sol de 50% à 30% de la capacité de rétention d'eau. Les résultats des expériences réalisées avec des sols contaminés par des déchets miniers ont montré que la survie du *Enchytraeus crypticus* n'a pas été affectée ; en revanche, sa reproduction a été négativement affectée. Ces résultats sont expliqués par le stress causé par la sécheresse et par l'augmentation de la biodisponibilité des métaux (cadmium, plomb, et zinc) (González-Alcaraz et al., 2015; González-Alcaraz & van Gestel, 2015). Concernant les expériences réalisées avec un sol agricole pollué, la salinité et la teneur élevée en argile du sol ont contribué aux effets négatifs observés chez le *Enchytraeus crypticus* et ces impacts ont été plus élevés sous des conditions de sécheresse. En effet, la EC_{10} (concentration entrainant un effet chez 10% de la population) et la EC_{50} ont montré une réduction d'environ 10% sous des conditions futures simulées par rapport à la situation de référence (González-Alcaraz & van Gestel, 2016).

Les expériences au laboratoire permettent d'étudier l'impact combiné du changement climatique et des polluants sur des individus. Cependant, il reste à évaluer la façon dans laquelle les impacts combinés du changement climatique et des substances toxiques sur les organismes pourraient se propager à des niveaux d'organisation biologique supérieurs comme les populations, les communautés et les écosystèmes. Ces impacts peuvent être modifiés à la hauteur de la population (Flèche 5, Figure 2.5) et de la communauté (Flèche 6, Figure 2.5). À leur tour, des processus agissant sur la population ou la communauté peuvent être influencés de manière directe et indirecte par le changement climatique (Flèches 7 et 8, Figure 2.5). De plus, l'environnement (Flèche 9, Figure 2.5) peut aussi jouer un rôle sur la réponse des populations et des communautés aux effets combinés du changement climatique et des substances toxiques (Moe et al., 2013). Ainsi, une étude qui a analysé l'impact combiné de plusieurs facteurs de stress sur un écosystème marin a conclu que le type de réponse observée, synergique, additive ou antagoniste dépend de la combinaison des facteurs de stress, ainsi que du niveau trophique et d'organisation biologique analysée (Crain et al., 2008).

2.5 Problématique générale issue de la revue de littérature

Les systèmes agricoles sont liés à des impacts environnementaux comme l'utilisation des terres, la rareté d'eau, la perte de biodiversité, des impacts écotoxiques et le réchauffement climatique. Grâce à sa nature holistique, l'ACV est considérée une méthodologie qui peut contribuer à la transition vers des systèmes agricoles plus durables (voir Section 2.2). Néanmoins, il existe quelques défis méthodologiques concernant des aspects spatiaux et temporels que cet outil doit surmonter (voir Sections 2.3.2 et 2.3.3). D'une part, des problèmes liés à l'aspect spatial surviennent quand l'influence de la variabilité spatiale de l'environnement est négligée lors de l'ÉICV. D'autre part, des défis d'ordre temporel surgissent lorsque l'ACV est utilisée pour supporter la prise des décisions dans la définition des stratégies à moyen et long terme en agriculture dans le contexte du changement climatique, car il influence à la fois l'inventaire du cycle de vie (voir Section 2.2.3) et interagit avec d'autres catégories d'impact (voir Section 2.4.2).

Un autre enjeu de la filière vitivinicole correspond à l'utilisation extensive de fongicides à base de cuivre pour pallier les impacts du mildiou. En effet, l'application de ces fongicides depuis longtemps a donné lieu à la pollution en cuivre des sols viticoles et a entraîné des impacts écotoxiques terrestres. Des plus des effets écotoxiques aquatiques sont aussi engendrés par des processus de transfert de polluants comme la lixiviation et le ruissellement (Section 2.2.1). Néanmoins, la contribution des métaux aux impacts écotoxiques était parfois négligée dans les études d'ACV portant sur l'empreinte environnementale de la viticulture ou plus largement de la production du vin, du fait que les premiers modèles d'ÉICV ne tenaient pas compte de la spéciation des métaux, ce qu'augmentais l'incertitude de ces modèles et causait une potentielle surestimation des impacts écotoxiques. Malgré les avancements récents d'amélioration dans l'évaluation de l'écotoxicité terrestre, de nos jours, il n'existe pas de facteurs de caractérisation qui ont été intégrés dans des méthodes d'ÉICV pour leur application dans des études d'ACV où des émissions de métaux ont lieu.

En outre, les scénarios de GES du GIEC prévoient des changements des conditions environnementales dans le futur, tels que l'augmentation de la température moyenne à la surface de la planète, des modifications des régimes de précipitations. De plus, les modifications des variables climatiques entraîneront des répercussions sur d'autres caractéristiques de l'environnement comme la teneur en matière organique des sols, ainsi que d'autres processus résultants de l'interaction des conditions climatiques et des activités humaines, comme l'érosion du sol. À leur tour, ces modifications de l'environnement affecteront le devenir, l'exposition et le niveau d'effet des polluants dans des conditions futures (Section 2.4.2). En conséquence, la validité des modèles de caractérisation pour l'évaluation des scénarios futurs est remise en question.

Il existe un intérêt grandissement dans l'application de l'ACV prospective, principalement dans l'évaluation de nouvelles technologies ou l'étude des impacts environnementaux des systèmes vulnérables aux changements climatiques, tels que le domaine agricole. Dans le cas des systèmes agricoles et en particulier de la viticulture, le changement climatique influe à la fois sur l'inventaire du cycle de vie et sur l'ampleur des impacts environnementaux dans des scénarios prospectifs. Pourtant, en ACV, l'interaction des catégories d'impact n'est pas considérée, car chaque catégorie d'impact est modélisée selon une chaine de cause à effet qu'y est associée (exemple de chaine de cause à effet pour l'écotoxicité terrestre, Section 2.4).

En résumé, le changement climatique peut entrainer des impacts à la fois sur les pratiques en viticulture, associées avec la mise en place des stratégies d'adaptation, et également sur l'ampleur de l'impact (éco)-toxique des polluants, ce qui donne lieu à des modifications de l'inventaire et de l'ÉICV des scénarios prospectifs de la viticulture. Bien que l'impact d'autres substances puisse aussi varier dans le futur, cette thèse se limitera à évaluer l'influence du changement climatique sur l'impact écotoxique terrestre du cuivre.

CHAPITRE 3 OBJECTIFS DU PROJET ET MÉTHODOLOGIE GÉNÉRALE

3.1 Hypothèse de recherche

À la lueur de l'état de connaissances, l'hypothèse de recherche suivante a été formulée :

Négliger l'influence du changement climatique sur les pratiques agricoles et sur l'évaluation de l'écotoxicité terrestre du cuivre dans l'analyse du cycle de vie prospective de la viticulture entraine une sous-estimation des impacts environnementaux potentiels.

3.2 Définition des objectifs de la thèse

L'objectif principal de cette thèse est de *« développer une approche d'analyse du cycle de vie prospective permettant d'inclure l'influence du changement climatique à la fois sur l'inventaire du cycle de vie et sur l'évaluation des impacts écotoxiques terrestres du cuivre ».* Plus particulièrement, cette approche méthodologique sera appliquée à l'ACV prospective de la viticulture et à l'évaluation de l'écotoxicité terrestre entrainée par l'application de fongicides à base de cuivre.

Pour atteindre cet objectif général, les objectifs spécifiques suivants ont été formulés :

1. Évaluer la variabilité spatiale de l'écotoxicité terrestre des fongicides à base de cuivre appliqués en viticulture.
2. Développer une approche d'analyse du cycle de vie prospective tenant compte de l'influence du changement climatique sur l'inventaire des pratiques en viticulture.
3. Développer des facteurs de caractérisation futurs pour l'écotoxicité terrestre du cuivre intégrant l'influence du changement climatique.

3.3 Méthodologie générale

Cette section présente la méthodologie développée afin de répondre aux objectifs spécifiques de cette thèse. La Figure 3.1 illustre les diverses étapes réalisées pour atteindre chaque objectif spécifique défini à la section précédente. L'atteinte des objectifs spécifiques a donné lieu à la rédaction de trois articles scientifiques qui sont présentés dans les chapitres suivants. Les diverses réalisations méthodologiques permettent donc d'atteindre l'objectif général d'inclure l'influence du changement climatique à la fois sur l'inventaire du cycle de vie et sur l'évaluation des impacts du cycle de vie des scénarios prospectifs en viticulture.

Afin d'éviter de la répétition, les sous-sections suivantes présentent une brève description des diverses réalisations méthodologiques pour l'atteinte de chaque objectif spécifique, car elles sont décrites en détail sous la forme d'article scientifique dans les chapitres suivants.

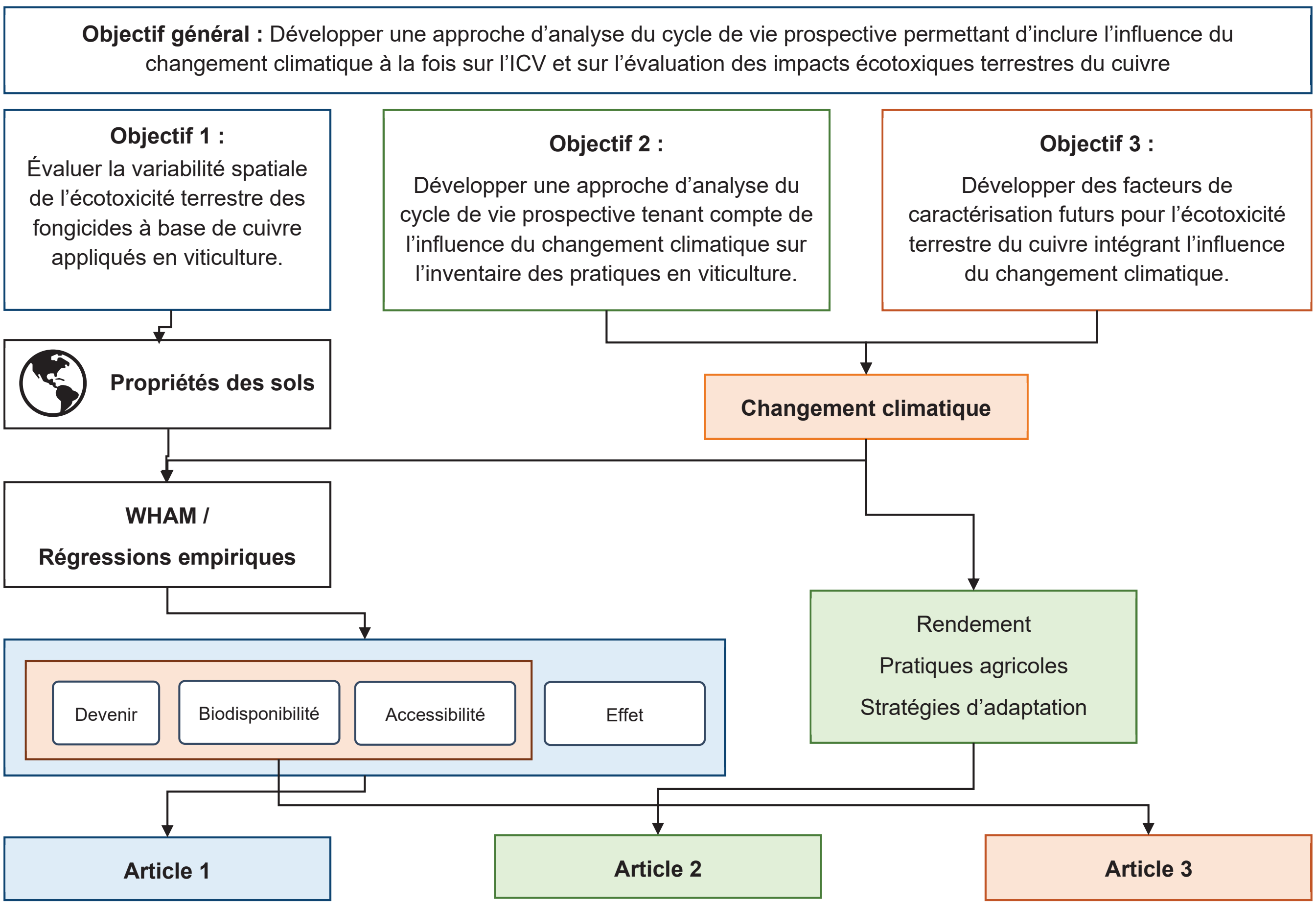


Figure 3.1 Étapes de la méthodologie générale scindées par objectif spécifique ayant mené à la rédaction d'un article scientifique

3.3.1 Évaluer la variabilité spatiale de l'écotoxicité terrestre des fongicides à base de cuivre appliqués en viticulture

Ce premier objectif spécifique correspond à la première étape de la méthodologie générale. Afin de déterminer la variabilité spatiale de l'écotoxicité terrestre entrainée par l'application de fongicides à base de cuivre, deux principales phases ont été mises en place : d'abord, le développement de facteurs de caractérisation pour l'écotoxicité terrestre du cuivre intégrant la spéciation du métal en fonction de la variabilité spatiale des sols; et deuxièmement, l'application de ces facteurs de caractérisation à un inventaire régionalisé tenant compte des différences dans les pratiques en viticulture, des propriétés des sols, ainsi que du climat de quelques régions viticoles européennes.

La première étape de cet objectif spécifique visait à déterminer la variabilité spatiale des facteurs de caractérisation pour l'écotoxicité terrestre du cuivre pour l'ensemble des sols non calcaires du monde. Ces facteurs de caractérisation ont été développés en mettant en œuvre deux approches méthodologiques : une approche basée sur l'utilisation du modèle géochimique de spéciation WHAM 6.0, et l'autre employant des régressions empiriques. Ensuite, les facteurs de caractérisations calculés ont été agrégés selon trois échelles spatiales : l'Europe, des pays, et des régions viticoles européens. De plus, les facteurs de caractérisation agrégés aux diverses échelles sont accompagnés de leur variabilité spatiale, laquelle serait un indicateur du niveau d'incertitude lorsqu'un facteur de caractérisation agrégé est utilisé au lieu d'un facteur de caractérisation spécifique à un vignoble donné.

Lors de la deuxième phase, un inventaire régionalisé de l'utilisation de fongicides à base de cuivre a été bâti sur la base d'une revue de littérature. Le choix des régions viticoles européennes s'est orienté selon les principaux pays d'origine du vin consommé au Québec : France, Italie, Espagne et Portugal selon les statistiques de 2017. Ensuite, le choix des régions viticoles à l'intérieur de chaque pays a été fait en fonction de la disponibilité des données sur l'utilisation des fongicides à base de cuivre.

Finalement, les facteurs de caractérisation régionalisés pour l'écotoxicité terrestre du cuivre ont été appliqués dans l'évaluation de l'inventaire régionalisé de l'utilisation de fongicides à base cuivre. D'abord, les facteurs de caractérisation spécifique à chaque région de l'étude de cas ont été employés, ensuite des facteurs de caractérisation agrégés ont été utilisés afin de tester l'influence de l'utilisation de ce qui serait l'équivalent d'un facteur de caractérisation générique en ACV traditionnelle sur le score d'impact. Le but de cette dernière phase était d'évaluer l'influence de la régionalisation à la fois sur l'inventaire et sur l'évaluation des impacts dans le cas spécifique de l'utilisation de fongicides à base de cuivre.

L'atteinte de cet objectif spécifique a abouti à la rédaction de l'article scientifique « *Regionalized Terrestrial Ecotoxicity Assessment of Copper-Based Fungicides Applied in Viticulture* » qui est présenté au Chapitre 4.

3.3.2 Développer une approche d'analyse du cycle de vie prospective tenant compte de l'influence du changement climatique sur l'inventaire des pratiques en viticulture

Le second objectif spécifique correspond au développement d'une méthodologie pour intégrer l'influence du changement climatique sur l'inventaire du cycle de vie de la viticulture dans des scénarios prospectifs. La méthodologie proposée inclut quatre phases principales : (1) évaluer les impacts projetés sur le rendement qui sont attribués au climat; (2) évaluer l'impact des extrêmes climatiques sur l'inventaire du cycle de vie en tenant compte du rendement et des options d'adaptation mises en place; (3) déterminer l'influence des impacts entrainés par le changement climatique sur les résultats de l'ACV prospective de la viticulture; et finalement (4) déterminer l'influence des extrêmes climatiques et des options d'adaptation sur les résultats de l'ACV prospective de la viticulture.

La première étape de ce deuxième objectif spécifique a mis en œuvre une technique de dissociation statistique afin d'évaluer les impacts des variables climatiques, plus particulièrement la température et la précipitation durant le cycle de croissance de la vigne, sur le rendement de raisins. Cette analyse statistique a été basée sur la collecte des données de la production des régions viticoles en France. De plus, deux scénarios de références ont été définis pour deux régions viticoles françaises contrastées par leurs climats et leurs pratiques agricoles. Ensuite, les impacts projetés du changement climatique sur le rendement des régions de l'étude de cas ont été calculés à partir des résultats de la technique statistique de dissociation et des changements de température et de précipitation prévus durant le cycle végétatif de la vigne selon deux scénarios d'émissions de GES et pour quatre périodes futures.

La deuxième étape vise à intégrer l'impact des extrêmes climatiques sur le rendement de la vigne et sur la mise en œuvre des options d'adaptation pour pallier les impacts des événements extrêmes. Plus spécifiquement, ces événements incluent la grêle, le gel printanier, la sécheresse, les vagues de chaleur, et l'augmentation de l'incidence des maladies de la vigne. L'impact des événements extrêmes est évalué en fonction de leur probabilité projetée, ainsi que sur le niveau de dommage associé à chacun de ces événements selon deux scénarios de GES et quatre périodes prospectives. Hormis la grêle, la probabilité des extrêmes est basée sur des indicateurs agroclimatiques et leur niveau de dommage est basé sur de valeurs rapportées dans la littérature scientifique.

Finalement, les étapes (3) et (4) correspondent à la réalisation de l'ACV prospective tenant compte de l'influence du changement climatique sur l'inventaire des pratiques en viticulture. Dans un premier temps (3), une condition c*eteris paribus* est considérée dans laquelle seulement le rendement est modifié en raison de l'impact entrainé par le changement climatique. Donc, à ce stade, les vignerons subissent ou bénéficient des changements projetés des variables climatiques durant la période de croissance de la vigne. Ensuite (4), l'impact des événements extrêmes est intégré dans l'évaluation des impacts du cycle de vie afin d'évaluer leur incidence à la fois sur le rendement et sur l'activation des mesures d'adaptation.

La réponse de ce deuxième objectif a donné lieu à l'article « *Prospective life cycle assessment of viticulture under climate change scenarios, application on two case studies in France* » qui fait l'objet du Chapitre 5.

3.3.3 Développer des facteurs de caractérisation futurs pour l'écotoxicité terrestre du cuivre intégrant l'influence du changement climatique

Le troisième objectif de cette thèse vise à montrer la pertinence d'inclure l'interaction entre des catégories d'impacts en ACV. En effet, la pratique courante en ÉICV est de modéliser les catégories d'impact de manière indépendante. Néanmoins, le changement climatique devient un facteur de stress qui modifie l'ampleur d'autres impacts environnementaux tels que l'écotoxicité terrestre des métaux. Ainsi, cet objectif se focalise sur la modélisation de l'influence du changement climatique sur les facteurs de caractérisation futurs pour l'écotoxicité terrestre du cuivre. L'approche méthodologique pour le développement des facteurs de caractérisation pour l'écotoxicité terrestre comporte quatre phases : (1) la définition de la zone d'étude, ainsi que des scénarios futurs; (2) le calcul des facteurs de caractérisations incluant des paramètres affectés par le changement climatique; (3) l'analyse d'attribution des changements projetés des facteurs de caractérisation; et (4) la différentiation spatiale des facteurs de caractérisation à différents niveaux de résolution spatiale.

Dans la première phase pour parvenir au troisième objectif spécifique, la zone d'étude a été limitée au vignoble européen en raison de l'utilisation fréquente des fongicides à base de cuivre. De plus, à ce stade, les scénarios prospectifs ont été définis à l'horizon 2050, correspondant à la période 2041-2060 établie par le GIEC pour évaluer les impacts du changement climatique à moyen terme. Ce choix a été fait en fonction de la disponibilité des projections des paramètres affectés par le changement climatique et qui font partie de la chaîne de cause à effet de l'écotoxicité terrestre, plus spécifiquement, des projections de carbone organique dans les sols, des projections d'érosion de sol, et des précipitations ont été collectés selon trois scénarios RCP, allant d'un scénario de faibles émissions de GES à un scénario à hautes émissions de GES, en passant par un scénario intermédiaire.

Lors de la deuxième étape, les facteurs de caractérisation futurs ont été calculés en considérant les projections de matière organique des sols, d'érosion de sol, et de précipitation à l'horizon 2050. Pour ce faire, des régressions empiriques ont été utilisées pour calculer d'abord la spéciation des métaux, c'est-à-dire, les différentes fractions de cuivre dans le sol. Ensuite, les facteurs intermédiaires pour le calcul des facteurs de caractérisation ont été calculés, parmi ces facteurs, notamment les facteurs de devenir qui intègrent les coefficients de partition (K_d), les projections d'érosion, ainsi que les projections de précipitation annuelle.

La troisième étape de cet objectif correspond à une analyse d'attribution des changements projetés des facteurs de caractérisation à partir de la différentielle de l'équation pour le calcul des facteurs de caractérisation, ce qui a permis ensuite d'évaluer l'influence des changements projetés dans la concentration en matière organique des sols, d'érosion du sol, et de précipitation dans les facteurs intermédiaires intégrant les facteurs de caractérisation. Finalement, la quatrième étape est portée sur l'agrégation des facteurs de caractérisation dans trois échelles spatiales : des vignobles européens, des régions administratives européennes et des pays européens.

L'atteinte du troisième objectif spécifique a amené à l'article scientifique « *Modelling the influence of climate change on characterization factors for copper terrestrial ecotoxicity* » qui est présenté au Chapitre 6.

CHAPITRE 4 ARTICLE 1 : REGIONALIZED TERRESTRIAL ECOTOXICITY ASSESSMENT OF COPPER-BASED FUNGICIDES APPLIED IN VITICULTURE

4.1 Présentation de l'article

Dans ce premier manuscrit, des facteurs de caractérisation pour l'écotoxicité terrestre du cuivre ont été calculés pour l'ensemble de sols non calcaires à l'échelle de la planète en utilisant deux approches existantes en ÉICV : l'une basée sur des régressions empiriques, et l'autre basé sur l'utilisation du modèle géochimique de spéciation WHAM 6.0. Ensuite, les facteurs de caractérisation régionalisés ont été appliqués dans l'inventaire régionalisé de quatre régions viticoles européennes.

Les auteurs de cet article sont Ivan Viveros Santos, Cécile Bulle, Annie Levasseur et Louise Deschênes. Il a été publié le 19 juillet 2018 dans *Sustainability.* Le matériel supplémentaire publié avec l'article est disponible en Annexe A et à l'adresse suivante : https://www.mdpi.com/2071-1050/10/7/2522.

4.2 Manuscrit

4.2.1 Abstract

Life cycle assessment has been recognized as an important decision-making tool to improve the environmental performance of agricultural systems. Still, there are certain modelling issues related to the assessment of their impacts. The first is linked to the assessment of the metal terrestrial ecotoxicity impact, for which metal speciation in soil is disregarded. In fact, emissions of metals in agricultural systems contribute significantly to the ecotoxic impact, as do copper-based fungicides applied in viticulture to combat downy mildew. Another issue is linked to the ways in which the intrinsic geographical variability of agriculture resulting from the variation of management practices, soil properties, and climate is addressed. The aim of this study is to assess the spatial variability of the terrestrial ecotoxicity impact of copper-based fungicides applied in European vineyards, accounting for both geographical variability in terms of agricultural practice and copper speciation in soil. This first entails the development of regionalized characterization

factors (CFs) for the copper used in viticulture and then the application of these CFs to a regionalized life-cycle inventory that considers different management practices, soil properties, and climates in different regions, namely Languedoc-Roussillon (France), Minho (Portugal), Tuscany (Italy), and Galicia (Spain). There are two modelling alternatives to determine metal speciation in terrestrial ecotoxicity: (a) empirical regression models; and (b) WHAM 6.0, the geochemical speciation model applied according to the soil properties of the Harmonized World Soil Database (HWSD). Both approaches were used to compute and compare regionalized CFs with each other and with current IMPACT 2002+ CF. The CFs were then aggregated at different spatial resolutions—global, Europe, country, and wine-growing region—to assess the uncertainty related to spatial variability at the different scales and applied in the regionalized case study. The global CF computed for copper terrestrial ecotoxicity is around 3.5 orders of magnitude lower than the one from IMPACT 2002+, demonstrating the impact of including metal speciation. For both methods, an increase in the spatial resolution of the CFs translated into a decrease in the spatial variability of the CFs. With the exception of the aggregated CF for Portugal (Minho) at the country level, all the aggregated CFs derived from empirical regression models are greater than the ones derived from the method based on WHAM 6.0 within a range of 0.2 to 1.2 orders of magnitude. Furthermore, CFs calculated with empirical regression models exhibited a greater spatial variability with respect to the CFs derived from WHAM 6.0. The ranking of the impact scores of the analyzed scenarios was mainly determined by the amount of copper applied in each wine-growing region. However, finer spatial resolutions led to an impact score with lower uncertainty.

4.2.2 Introduction

This study aims to conduct a regionalized life-cycle assessment of the terrestrial ecotoxicity of copper-based fungicides applied in viticulture in Europe while accounting for regionalization both in terms of inventory and impact assessment.

Agricultural systems satisfy basic, social, and cultural human needs. However, the environmental footprint of the agricultural sector is exacerbated by the growing population, which is currently over seven billion (Notarnicola et al., 2017). The environmental impacts of agricultural systems include resource depletion, global warming, biodiversity and soil fertility loss, water scarcity, nitrification, and human and ecological toxicity impacts (Notarnicola et al., 2017). Life-cycle assessment (LCA) is recognized as a key decision-making tool (Notarnicola et al., 2017; Sala et al., 2017) to improve the environmental performance of food production and consumption patterns. However, when LCA is applied to agricultural systems, certain methodological challenges must be addressed to increase the robustness of the results. It is essential to consider the intrinsic geographical variability of agricultural systems in LCA since this variability affects the inventory analysis, impact assessment, and interpretation phases of LCA (Notarnicola et al., 2017).

Metal emissions were shown to contribute significantly to the ecotoxicity impact of agricultural systems. Specifically, copper has been identified as the main contributor to the ecotoxicity impacts in the life cycle of wine bottles, resulting from the application of copper-based fungicides in viticulture (Falcone et al., 2016; Vázquez-Rowe, Villanueva-Rey, Moreira, et al., 2012). Copper-based fungicides are widely used in both conventional and organic viticulture systems to combat vine fungal diseases caused by *Plasmopara viticola* (Komárek et al., 2010). Copper applied to vines reaches the soil and ground and surface waters by different mechanisms, leading to impacts on terrestrial and aquatic ecosystems (Bart et al., 2017; Fernández et al., 2015; Frankart et al., 2002; Ruyters et al., 2013). However, the assessment of the terrestrial ecotoxicity impact of these emissions in LCA studies is not consistent. For instance, in a study comparing conventional and biodynamic viticulture techniques, even though the emissions of copper to soil are included in the inventory analysis, the authors chose to omit the impact because the pesticide dispersion model (PestLCI) could not compute the primary distribution of inorganic pesticides and the modelling of the copper terrestrial ecotoxicity impact was considered highly uncertain (Pedro Villanueva-Rey et al., 2014). However, the omission of metals from terrestrial ecotoxicity impact assessment could lead to biased conclusions and raise credibility issues (Plouffe et al., 2015b).

In LCA, the potential terrestrial ecotoxicity impact of a metal emission is calculated as the product of its magnitude (mass per functional unit) and a characterization factor (CF). Characterization modelling is traditionally based on the physicochemical properties of metals and average environmental conditions. CFs derived through this approach are called site-generic or global CFs because they are considered applicable to the entire world, regardless of the receiving environmental compartment's location. However, spatial differentiation is required to acknowledge the influence of the spatial variability of environmental conditions on the terrestrial ecotoxicity impact of metals. Two levels of spatial differentiation of CFs are defined in LCA: site-dependent and site-specific. Site-dependent CFs include some geographical differentiation, at the level of countries or states, for example. Site-specific CFs are defined at finer spatial resolutions, for instance, at the level of an agricultural field (Hauschild, 2006; Patouillard et al., 2016). In the case of acidification and eutrophication, LCA studies have shown that site-dependent impact assessment provides more realistic and accurate results, demonstrating the relevance of including regionalization in the impact assessment phase (Mutel & Hellweg, 2009; Roy et al., 2014).

USEtox is the scientific consensus multimedia model used to characterize human toxicity and ecotoxicity in LCA. It was developed under the leadership of the UNEP/SETAC Life Cycle Initiative and makes it possible to compute CFs according to the parameterization of 8 continental landscapes and 17 subcontinental landscapes (USEtox, 2017). However, for substances such as metals, CFs at a higher spatial resolution are required since properties influencing the fate and exposure of these substances, such as pH, redox potential, and cation exchange capacity, are highly variable (Notarnicola et al., 2017; USEtox, 2017). The latest version of USEtox makes it possible to calculate CFs accounting for metal speciation based on freshwater chemistry for the aquatic ecotoxicity of 14 metals using the WHAM 6.0 (Windermere Humic Aqueous Model) geochemical speciation model (Dong et al., 2014; Gandhi et al., 2010; USEtox, 2017). However, USEtox labels CFs for metals as indicative, acknowledging the high uncertainty in the modelling of the fate, exposure, and effects of these substances. Moreover, CFs for terrestrial ecotoxicity are still missing in USEtox, and this has been recognized as a methodological challenge to improve the robustness of LCA studies of agricultural systems (Notarnicola et al., 2017; USEtox, 2017).

Two methodological approaches were developed to compute CFs integrating metal speciation for terrestrial ecotoxicity assessment in LCA. Owsianiak, et al. (2013) developed a method based on empirical regression models to calculate metal speciation and ecotoxicity according to the spatial variability of soil properties. CFs for copper and nickel after a unit emission to air were calculated for a set of 760 non-calcareous soils of the ISRIC-WISE3 soil database. The CFs showed a spatial variability of 3.5 and 3 orders of magnitude for copper and nickel, respectively. A second methodological approach consisted in using WHAM 6.0 to calculate metal speciation according to soil properties reported in the Harmonized World Soil Database (HWSD) (Plouffe et al., 2015a, 2016). Plouffe, et al. (2015a) derived bioavailability factors (BFs) for zinc that correspond to the fraction of metal available for soil organisms of total metal concentration in soil. BFs for the soluble and true solution fraction extended over 6 and 18 orders of magnitude, respectively. In a following study, the CFs for zinc were derived and extended over 1.8 orders of magnitude when considering

BFs defined by the soluble fraction and 14 orders of magnitude when using BFs for the true solution fraction (Plouffe et al., 2016).

Heavy metals occur naturally in soils at varying concentrations as a result of geochemical fluxes between soils and their parent material. The magnitude of these geochemical fluxes is determined by both the parent material composition and the weathering processes (Cabral Pinto et al., 2016, 2017). Because airborne metal deposition is another significant source of metals in soils, a study addressed the integration of both atmospheric dispersion and speciation in soils in the computation of CFs for the airborne emissions of copper, nickel, and zinc. The derived CFs showed a spatial variability of around 3 orders of magnitude. Furthermore, the authors suggest reporting the uncertainty arising when using aggregated CFs in cases in which the location of the emissions is unknown (Aziz et al., 2018).

Very recently, Peña et al., (2018) assessed the ecotoxicity impact of organic pesticides and copper-based fungicides in European vineyards. In the study, CFs for aquatic ecotoxicity were calculated with USEtox and CFs for copper terrestrial ecotoxicity were derived from regression models developed by Owsianiak, et al. (2013). However, the chosen CFs were derived for an emission to continental rural air and not to a direct emission in soil, which is debatable since most of the copper-based fungicide will fall within the parcel while being applied or after precipitations and not be diluted in the continental air compartment. The aquatic ecotoxicity impacts showed a spatial variability over 3 orders of magnitude for the seven water archetypes analyzed, and the terrestrial ecotoxicity impacts extended over 2 orders of magnitude (Peña et al., 2018).

The objective of this study is to conduct a regionalized impact assessment of the terrestrial ecotoxicity of copper-based fungicides applied in viticulture. To do so, regionalized CFs for copper had to be (a) developed and (b) applied in a regionalized inventory of viticulture accounting for regionalized agricultural practices. The development of the CFs is threefold. First, the metal speciation is included in the calculation of the CFs for copper terrestrial ecotoxicity according to the spatial variability of soil properties. Second, the influence of the level of spatial resolution on the uncertainty of the regionalized impact assessment of copper-based fungicides in viticulture is evaluated. Finally, the CFs derived from the two available methodological approaches to account for metal speciation in terrestrial ecotoxicity in LCA are compared (a method based on WHAM

6.0 (Plouffe et al., 2016) and a method based on empirical regression models (Owsianiak et al., 2013).

4.2.3 Methods

4.2.3.1 Characterization Factors for Copper Terrestrial Ecotoxicity

In this study, the two available methodological approaches to integrate metal speciation and bioavailability in soil in LCA were employed and compared: a method based on WHAM 6.0 (Plouffe et al., 2016) and a method based on empirical regression models (Owsianiak et al., 2013).

The WHAM 6.0 geochemical speciation model is well-known and used to consider the complexation of metals with organic matter. This model is recommended by the Clearwater Consensus to derive CFs for freshwater ecotoxicity and applied in the USEtox model (Diamond et al., 2010; USEtox, 2017). Empirical regression models are equilibrium models that are used to derive different fractions of metal partitioning (e.g., soluble metal concentration or reactive fraction) from soil properties. However, the application of empirical regression models to compute metal speciation is recommended only for soils whose physicochemical properties are within the range of soil properties from which they were derived (J E Groenenberg et al., 2010). Both approaches are limited by the fact that they do not consider the kinetics of precipitation and dissolution (Dong et al., 2014; Plouffe et al., 2015a).

In the case of aquatic ecotoxicity, the Clearwater Consensus recommends using the truly dissolved fraction (free metal ions and ion pairs) to account for bioavailability in CFs for metals (Diamond et al., 2010; Gandhi et al., 2010). In this study, for both methodological approaches, BFs were calculated as a fraction of the free ion concentration, which is in line with the studies by Owsianiak, et al. (2013) and Plouffe, et al. (Plouffe et al., 2016). The reason for this modelling choice is that the parameters required to compute the truly dissolved fraction, such as the composition of major anions, are generally missing in soil databases (Owsianiak et al., 2013). Moreover, considering the free ion fraction as the bioavailable fraction is consistent with the application of terrestrial biotic ligand models (TBLMs) to derive effect factors (EFs).

CFs were calculated according to the Plouffe, et al. (Plouffe et al., 2016) model using Equation 4.1 and a second set of CFs was calculated using the Owsianiak, et al. (2013) method according to Equation 4.2, where *FF* (day) is the fate factor of copper for a direct emission in soil, *BF*

($kg_{free}{\cdot}kg_{total}^{-1}$ and $kg_{free}{\cdot}kg_{reactive}^{-1}$, respectively) is the bioavailability factor, *ACF* ($kg_{reactive}{\cdot}kg_{total}^{-1}$) corresponds to the fraction of reactive metal over total metal, and *EF* (potentially affected fraction (PAF)$\cdot m^3 \cdot kg_{free}^{-1}$) is the effect factor. These factors are described in detail in the following sections.

$$CF = FF \cdot BF \cdot EF \tag{4.1}$$

$$CF = FF \cdot ACF \cdot BF \cdot EF \tag{4.2}$$

The soil parameters required to estimate copper speciation, bioavailability, and effect factors were retrieved from the Harmonized World Soil Database (HWSD) version 1.2, which contains 48,148 soil mapping units (FAO/IIASA/ISRIC/ISS-CAS/JRC, 2012). The spatially dominant soil was selected (given by the SEQ field) from each mapping unit of the HWSD, leading to 16,327 unique soil mapping units. In a following step, only the mapping units corresponding to soils were selected (according to the field ISSOIL), which resulted in a set of 16,165 soil mapping units. For the application of the method based on empirical regression models, soil mapping units missing values of organic matter and clay content were excluded (28 cases), and 118 soil mapping units were omitted because the computed coefficient for divalent cations was greater than 1 in those units. For both methods, 8178 soil mapping units with calcium carbonate ($CaCO_3$) content superior to 0% were also discarded because TBLMs and WHAM 6.0 are not able to model calcareous soils. The parameterization of WHAM 6.0 was carried out as described in Plouffe, et al. (2015a), and 80 soil mapping units were discarded because of missing copper concentration values. In the end, two sets of 7841 and 7907 soil mapping units were used to calculate the CFs with empirical regression models and WHAM 6.0, respectively.

The application of WHAM 6.0 and empirical regression models is based on the assumption that topsoil properties are homogeneous in a given mapping unit of the HWSD according to the dominant soil. In consequence, copper speciation, bioavailability, toxicity level, and the corresponding CF are constant for a given soil mapping unit of the HWSD.

4.2.3.1.1 Fate Factors

The *FF* (day) corresponds to the residence time of the metal fraction in soil after a direct emission to this compartment (Equation 4.3); ΔC_{total} ($kg_{total}{\cdot}kg_{soil}^{-1}$) is the incremental change in concentration of metal in soil; ΔM ($kg_{total\ emitted}{\cdot}day^{-1}$) is the incremental change in the emission of metal to soil; V (m^3) is the volume of the soil compartment; and ρ_b ($kg_{soil} \cdot m^{-3}$) is the bulk density of the soil. The *FF* accounts for intermedia transport processes (advective and diffusive transport) as well as for removal processes, such as runoff and leaching to groundwater (Henderson et al., 2011).

$$FF = \frac{\Delta C_{total} \cdot V \cdot \rho_b}{\Delta M} \tag{4.3}$$

Fate factors of copper in agricultural and natural soil were calculated for a direct emission to these compartments by employing USEtox with partitioning coefficients (K_d, $L \cdot kg^{-1}$) specific to each soil mapping unit of the HWSD. K_d corresponds to the ratio of metal concentration in the solid phase over the concentration of solubilized metal. While Owsianiak, et al. (2013) considered a constant background concentration of 14 mg of copper per kg of soil, we estimated the copper background concentration for each soil mapping unit of the HWSD based on the soil's texture according to Kabata-Pendias and Mukherjee (2007) and Plouffe, et al. (Plouffe et al., 2016). Furthermore, instead of using the default landscape of USEtox to derive FFs, each soil mapping unit of the HWSD was related to one of the 17 subcontinental landscapes, which made it possible to consider the variation of precipitation and run-off rates.

4.2.3.1.2 Accessibility Factor

Owsianiak, et al. (2013) proposed to decouple an *ACF* ($kg_{reactive} \cdot kg_{total}^{-1}$) from the *BF* to recognize that the largest metal pool in soil is absorbed to soil particles and that only a fraction of total metal in soil is reactive. ACFs were calculated according to Equation 4.4, where $\Delta C_{reactive}$ ($kg_{reactive} \cdot kg_{soil}^{-1}$) is the incremental change of the concentration of reactive metal in soil, and ΔC_{total} ($kg_{total} \cdot kg_{soil}^{-1}$) is the incremental change in total metal in soil. In line with Owsianiak, et al. (2013), the reactive metal pool was calculated by applying a regression model reported by Römkens, et al. (2004).

$$ACF = \frac{\Delta C_{reactive}}{\Delta C_{total}} \tag{4.4}$$

4.2.3.1.3 Bioavailability Factor

BFs ($kgf_{ree} \cdot kg_{total}^{-1}$) for the approach using the geochemical speciation method were calculated according to Equation 4.5, where ΔC_{free} ($kg_{free} \cdot m^{-3}$) is the change of concentration of the free ion fraction of metal, and Θ_w ($m^3 \cdot m^{-3}$) is the volumetric soil water content. BFs ($kg_{free} \cdot kg_{reactive}^{-1}$) for the approach applying empirical regression models were calculated with Equation 4.6, and the free ion fraction was calculated with a regression reported by Groenenberg, et al. (2010) as in the study by Owsianiak, et al. (2013).

$$BF = \frac{\Delta C_{free} \cdot \Theta_w}{\Delta C_{total} \cdot \rho_b} \tag{4.5}$$

$$BF = \frac{\Delta C_{free} \cdot \Theta_w}{\Delta C_{reactive} \cdot \rho_b} \tag{4.6}$$

4.2.3.1.4 Effect Factor

The EFs ($PAF \cdot m^3 \cdot kg_{free}^{-1}$) were calculated according to Equation 4.7, which is applied in USEtox (Rosenbaum et al., 2008) and was used in the studies addressing the calculation of CFs for metal terrestrial ecotoxicity (Owsianiak et al., 2013; Plouffe et al., 2016; Tromson et al., 2017). In Equation 4.7, PAF is the potentially affected fraction of terrestrial species, and $HC50_{EC50}$ is the geometric mean of individual EC_{50} values (Owsianiak et al., 2013). EC50 values were calculated with TBLMs reported by Thakali, et al. (2006) for the following biological endpoints: *barley root elongation (BRE), tomato shoot yield (TSY), Folsomia candida juvenile production (FJP), Eisenia fetida cocoon production (ECP), glucose induced respiration (GIR)*, and *potential nitrification rate (PNR)*. TBLMs consider that the toxicity of a metal in soil is related to the fraction of metal bound to the biotic ligands, which are receptors of soil organisms. TBLMs also account for metal competition of the free ion metal and other major cations found in soil, such as Ca^{2+}, Na^+, Mg^{2+}, and H^+. In the case of copper, the parameters for the TBLMs are the activities of hydrogen and magnesium but these activities are not reported in the HWSD. Therefore, cation exchange was modelled according to Owsianiak, et al. (2013) and to the parameterization of WHAM 6.0 carried out by Plouffe, et al. (2015a).

One limit of using TBLMs in LCA is the fact that these models are only available for copper and nickel, which precludes the systematic assessment of the terrestrial ecotoxicity of metals. Another option to derive EFs for metal terrestrial ecotoxicity is the equilibrium partitioning method (EqP), which extrapolates terrestrial ecotoxicity from aquatic ecotoxicity data. The EqP method considers that the soluble fraction of metal is related to the level of toxicity and the sensitivity of terrestrial organisms is similar to that of aquatic organisms. However, the study by Tromson, et al. (2017) showed that the EqP method fails to assess the terrestrial ecotoxicity of metals. The authors suggest applying TBLMs and highlight the need to extend these models to a broader set of metals and terrestrial organisms. In keeping with the results of Tromson, et al. (2017) and the method of Owsianiak, et al. (2013), EFs were derived from TBLMs.

$$EF_S = \frac{\Delta PAF}{\Delta C_{free}} = \frac{0.5}{HC50_{EC50}} \tag{4.7}$$

4.2.3.2 Spatial Differentiation of Characterization Factors

Overall, 7841 CFs at the native spatial resolution of soil mapping units of the HWSD were derived with the method using empirical regression models, and 7907 CFs were computed with the method applying the WHAM 6.0 geochemical speciation model. Then, to assess the influence of the level of regionalization on the impact score, a global CF (site-generic CF) and site-dependent CFs were calculated. For site-dependent CFs, three spatial resolutions were considered: Europe, country, and wine-growing region levels of the case study (Section 4.2.3.3). The spatial calculations were carried out with QGIS version 2.14.15 open-source software. Aggregated CFs at different levels of spatial resolution were calculated by applying an area-weighted average, as shown in Equation 4.8, where CF_i is the characterization factor of the intersected soil mapping unit i, and A_i corresponds to the surface of vineyards of a given region used as a proxy of the probability of copper emission in viticulture. This means that the aggregated CFs at the country or continental scale in this study are sector-specific. The European vineyard surface was retrieved from the CORINE land cover project (EEA, 2017), and the Mollweide projection was used to calculate the area of soil mapping units, countries, and wine-growing regions of the case study (Section 4.2.3.3). In the case of the global CF, all the soil mapping units were considered in the computation, which is equivalent to applying a generic CF when impact regionalization is not considered in LCA. However, for site-dependent CFs, only soils corresponding to vineyards were used in the computation of area-weighted CFs.

$$CF_{area-weighted\ average} = \frac{\sum_{i=1}^{n}(CF_i \cdot A_i)}{\sum_{i=1}^{n} A_i} \tag{4.8}$$

In this study, the finest spatial differentiation of CFs for copper terrestrial ecotoxicity was made at the wine-growing region level because of two reasons. First, a mapping unit of the CORINE land cover shapefile, from which the European vineyard coverage was extracted, does not necessarily correspond to an individual farm (EEA, 2017). Secondly, the highest spatial resolution of the inventory analysis is also at the wine-growing region level. Nevertheless, the calculated CFs at the native resolution of the HWSD allow for the extraction of a site-specific CF if the exact location of a farm is known or to compute CFs at other spatial resolutions than those considered in this study (3.1).

4.2.3.3 Case Study

This case study is driven by the assessment of the terrestrial ecotoxicity impacts generated by the application of copper-based fungicides in viticulture. For comparative purposes, the functional unit corresponds to 1 kg of grapes for wine making in a European vineyard (Table 4.1). The system boundaries were set at the vineyard level, from gate-to-gate. However, the only elementary flow considered in the inventory is copper emitted to soil resulting from the spraying of copper-based fungicides. While we recognize that this is a very simplified assessment, full LCA studies on grape and wine production have already been published (Neto et al., 2013; Vázquez-Rowe et al., 2013; Vázquez-Rowe, Villanueva-Rey, Moreira, et al., 2012).

Table 4.1 Inventory of copper applied in the analyzed scenarios (data per functional unit (FU): 1 kg of grapes).

Region	**Type (Appellation)**	**Copper (kg)**	**Year of Inventory Collection**	**Data Source**
Languedoc-Roussillon (France)	-	1.51×10^{-3}	2008	Bellon-Maurel et al. (2015)
Tuscany (Italy)	Red (*Chianti Colli Senesi*)	8.58×10^{-5}	2007	Vázquez-Rowe et al. (2013)

Region	Type (Appellation)	Copper (kg)	Year of Inventory Collection	Data Source
Minho (Portugal)	White (Vinho verde)	1.72×10^{-3}	2008	Neto et al. (2013)
Galicia (Spain)	White (Ribeiro)	8.19×10^{-4}	2008	Vázquez-Rowe et al. (2012)

In this case study, four scenarios were defined according to the principal countries of origin of wine consumed in Québec, Canada: France (30% market share), Italy (23%), Spain (8%), and Portugal (4%) (SAQ, 2015). The coverage of European vineyards was obtained from the CORINE program of the European Environment Agency (EEA, 2017) and the QGIS v2.14.15 software was used to relate vineyard area and geopolitical information. Based on scientific literature on the environmental assessment of grapes and wine production, one wine-growing region by country was selected according to the following criteria: (1) the wine region is specified in the study, (2) the year of the inventory collection is reported, and (3) the amount of copper-based fungicide applied is reported (Figure 4.1). Organic vineyards were not distinguished from conventional vineyards because copper-based fungicides are applied to combat downy mildew in both farming practices (Komárek et al., 2010). Furthermore, the proportion of organic vineyards at country level is relatively low: 9% in France, 13% in Italy, 10% in Spain, and 1% in Portugal (Eurostat, 2015).

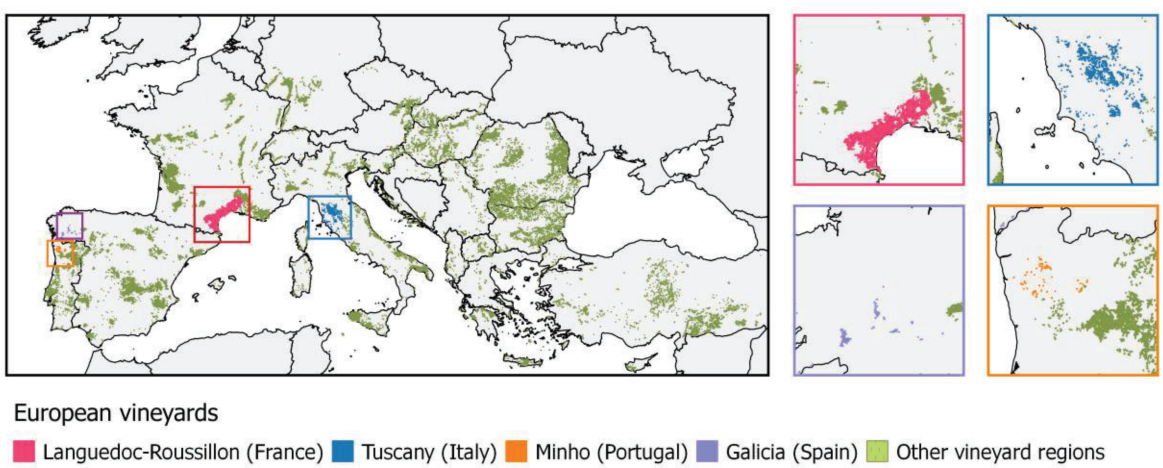


Figure 4.1 Location of wine regions analyzed in this case study.

The inventory of each scenario (wine-growing regions) was built with data from the literature. In cases in which the inventory is reported by a functional unit defined as a 750-mL wine bottle, the yield of grapes (mass of harvested grapes per surface of soil) and wine yield (volume of wine per mass of grapes) were used to obtain an inventory per kilogram of grapes (Table 4.1). We acknowledge that the difference in the year of inventory collection affects the amount of copper-based fungicides applied in each region (Table 4.1). This is recognized as a limitation of the comparison between the selected scenarios. The year-to-year variability of inventory in viticulture is mainly driven by varying meteorological conditions. This was shown in a study analyzing 4 years of production in Galician wineries, where, for instance, the ecotoxicity impact of a wine bottle produced in 2010 was 94% higher as compared to production in 2007 (Vázquez-Rowe, Villanueva-Rey, Moreira, et al., 2012). Additionally, wine type and appellations are reported in Table 4.1 where available.

Because the aim of this case study is to assess the terrestrial ecotoxicity impact of copper-based fungicides, the agricultural field was allocated to the ecosphere. This is in line with the recommendation by the Glasgow Consensus to define the boundary between the technosphere and the ecosphere according to the goal and scope of the LCA study (Rosenbaum et al., 2015). Regarding the inventory modelling of copper emissions, a pesticide model, such as PestLCI, was not used for two reasons: it allocates the agricultural field to the technosphere and even the viticulture-customized version of PestLCI 2.0 is not able to model copper emissions (Christel Renaud-Gentié et al., 2015). In order to avoid potential overlaps between inventory modelling and impact assessment, we assumed that the amount of copper applied is emitted 100% to agricultural soil in accordance with the approach of the widely used ecoinvent life-cycle inventory database (Nemecek et al., 2007). Furthermore, this avoids underestimating the potential ecotoxic effects at the end of life of vines, which is coherent with the precautionary principle and the egalitarian perspective in LCA (Hellweg et al., 2003).

The impact score of each scenario was obtained according to Equation 4.9, where I_w is the terrestrial ecotoxicity impact score ($\mathrm{PAF \cdot m^3 \cdot day \cdot kg_{grapes}^{-1}}$) of wine-growing region w, m_w is the amount of copper emitted to soil per kg of grapes ($\mathrm{kg\ Cu \cdot kg_{grapes}^{-1}}$), and CF_w is the characterization factor for copper terrestrial ecotoxicity ($\mathrm{PAF \cdot m^3 \cdot day \cdot kg_{Cu}^{-1}}$) of wine-growing region w at a given spatial resolution.

$$I_w = m_w \cdot CF_w \tag{4.9}$$

4.2.4 Results and Discussion

4.2.4.1 Characterization Factors for Copper Terrestrial Ecotoxicity

CFs for copper terrestrial ecotoxicity were obtained at the spatial resolution of soil mapping units of the HWSD. CFs within 95% spatial variability are represented in Figures 4.2 and 4.3 for the method based on empirical regression models and for the method based on WHAM 6.0, respectively. The geographical variability of CFs obtained with empirical regression models is around 7.3 orders of magnitude between the lowest and highest value, with a median value of 1.88×10^4 PAF·m^3·day·kg^{-1}. To put these results into perspective, Owsianiak, et al. (2013) found a spatial variability of 3.5 orders of magnitude with a median value of 1.4×10^3 PAF·m^3·day·kg^{-1}, and Peña, et al. (2018) reported a spatial variability of CFs over 1.5 orders of magnitude with a mean value of 2.3×10^3 PAF·m^3·day·kg^{-1}. The differences in the extent of the CFs calculated in this study and those obtained by Owsianiak, et al. (2013) are attributed to the soil sample size, soil properties, and the soil database used (Table A.1). Moreover, in this study, FFs for a direct emission of metal in agricultural soil were integrated into the computation of CFs, whereas Owsianiak, et al. (2013) and Peña, et al. (2018) applied FFs of metal in natural soil for an emission to continental rural air, which may explain an impact lower by around 1 order of magnitude, as all that is emitted to air does not reach the agricultural soil but rather is distributed among all the deposition compartments at the continental scale in USEtox or advected with air outside of the continental box. Most of the undefined CFs occurred in soil mapping units where the $CaCO_3$ content is greater than 0% (Figure 4.2 and Figure 4.3 and Figure A.1).

A spatial variability over 6.6 orders of magnitude was found for CFs obtained with the method based on WHAM 6.0, which is mainly caused by the geographical variability of BFs (Table A.1). Nevertheless, when considering a 95% spatial variability interval (2.5th and 97.5th percentiles), CFs for copper terrestrial ecotoxicity span 5.5 orders of magnitude (Figure 4.3), with a median CF of 1.71×10^3 PAF·m^3·day·kg^{-1}. In comparison, Plouffe, et al. (2016) estimated zinc speciation with WHAM 6.0 and derived CFs, for which the BF was based on the true solution fraction (free ions and ions pairs). The authors found that true solution CFs for zinc terrestrial ecotoxicity span over 14.3 orders of magnitude.

CFs derived from both methods show a similar pattern with respect to soil organic matter content and pH (Figures A.7 and A.8). Higher values of CFs occurred where organic matter content is low, but the influence of pH was found to be less significant with respect to organic matter content. This trend is explained by the fact that soil pH has less impact on copper partitioning between the solid and aqueous phases because copper has a high affinity for organic matter (Ivezić et al., 2012). The differences in the orders of magnitude of CFs derived from both methods were mainly determined by the extent of FFs (Figure A.3). Regardless of the method, higher FFs resulted from higher K_d values, which is explained by a lower contribution of removal processes (runoff and leaching) when the concentration of copper in the aqueous phase is lower.

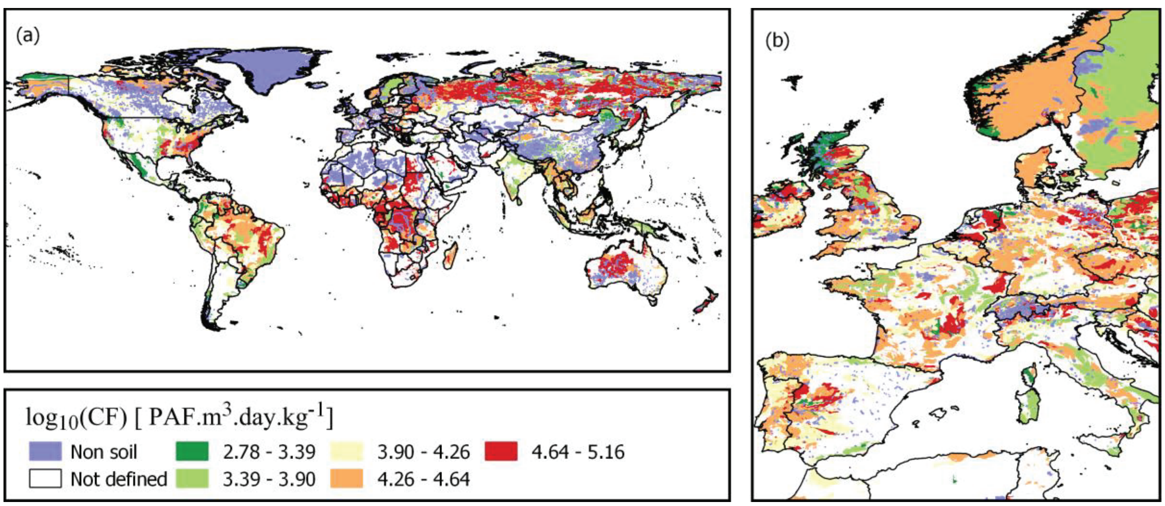


Figure 4.2 Regionalized characterization factors (CFs) for copper terrestrial ecotoxicity at Harmonized World Soil Database (HWSD) soil mapping unit resolution obtained with empirical regression models. (a) Map of the world indicating the calculated CFs. (b) Zoom of European wine-growing countries. PAF, potentially affected fraction.

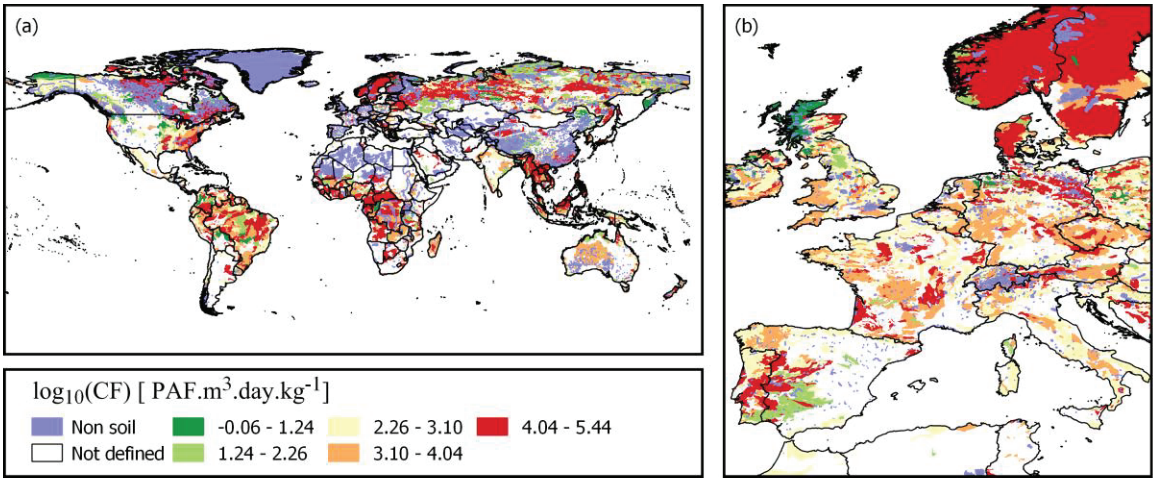


Figure 4.3 Regionalized CFs for copper terrestrial ecotoxicity at HWSD soil mapping unit resolution obtained with WHAM 6.0. (a) Map of the world indicating the calculated CFs. (b) Zoom of European wine-growing countries.

The CFs computed in this study are in shapefile format as attributes of the soil mapping units of the HWSD. These geographical information system (GIS) data can be imported, for instance, with openLCA open-source software, which includes an implementation approach to conduct GIS-based regionalized LCA. This approach enables the LCA practitioner to define geographic regions, and openLCA then calculates the corresponding area-weighted CF according to Equation 4.8 for the geographical zone defined by the user (Rodríguez et al., 2014). However, openLCA executes spatial calculations in WGS84 projection, which can potentially bias the actual area. Alternatively, the shapefiles may be loaded in a GIS software, such as QGIS, and it is possible to set an equal area projection, such as Mollweide, to compute surfaces and derive CFs at spatial resolutions other than those presented in Sections 4.2.3.3 and 4.2.4.2. Consequently, the application of the derived CFs is not limited to this case study (4.2.4.2). Besides, the LCA practitioner can establish the most appropriate proxy to aggregate CFs for the case study or use the default area weighting of openLCA. For instance, alternative proxies may be based on regionalized inventories of pollutants or on the location of the activities of the analyzed case study (e.g., mining sector). The latter proxy may be calculated by geospatial analysis, which makes it possible to correlate CFs at native resolution with a map of the analyzed economic sector.

Another potential application of the CFs derived in this study is in territorial LCA. Nitschelm, et al. (2016) developed a spatialized territorial LCA (STLCA) method for territories in which the main economic activity is agriculture. This method aims to consider the spatial variability of both the inventory and the potential environmental impacts of agriculture within a territory. Territorial LCA can serve to evaluate the current situation, to analyze different scenarios, or to assess future situations (Nitschelm et al., 2016). The latter application could be relevant for future planning scenarios in viticulture because climate change will affect the distribution of current European wine-growing regions by 2050 (M Moriondo et al., 2013). Furthermore, there is a growing interest to couple Environmental Management Systems with LCA in the evaluation and management of environmental issues at the territorial level (Loiseau et al., 2018; Mazzi et al., 2017).

4.2.4.2 Characterization Factors at Different Spatial Resolutions

The global aggregated CFs for copper terrestrial ecotoxicity calculated with both methods are approximately equivalent to 3.27×10^4 PAF·m^3·day·kg^{-1}. For both methods, the CF aggregated at the global scale is greater than the corresponding CF aggregated at the continental scale derived from European vineyards' soils. The aggregated CF for European vineyards derived from the method based on WHAM 6.0 is 1.42×10^4 PAF·m^3·day·kg^{-1}, and the aggregated CF obtained with empirical regressions is 2.47×10^4 PAF·m^3·day·kg^{-1}. These aggregated CFs are approximately 3.8 and 3.6 orders of magnitude lower than the CF for the copper terrestrial ecotoxicity of IMPACT 2002+ (9.9×10^7 PAF·m^3·day·kg^{-1}) (Jolliet et al., 2003), respectively. These results demonstrate the relevance of including metal speciation in the calculation of CFs for the assessment of metal terrestrial ecotoxicity.

The aggregated CFs at the country and wine-growing region levels are shown in Figure 4.4 with their respective spatial variability. In this figure, the width of boxes is proportional to the number of CFs of a scenario at a given spatial resolution (country or wine-growing region). Given that life-cycle inventory databases, such as ecoinvent, have a spatial resolution at country level, it would be appropriate to aggregate CFs at country-level spatial resolution. However, the higher spatial variability of CF factors at country level is approximately 2.5 orders of magnitude for the method applying empirical regression models and 7.2 orders of magnitude for the method based on WHAM 6.0, respectively. Figure 4.4 shows that CFs obtained with both methods follow a similar pattern. CFs at a spatial resolution of wine-growing regions have a lower geographical variability than CFs at country level. By applying the empirical regression models, the higher spatial variability of CFs at the wine-growing region level is 1.7 orders of magnitude, while that for CFs obtained with WHAM 6.0 is approximately 5.9 orders of magnitude.

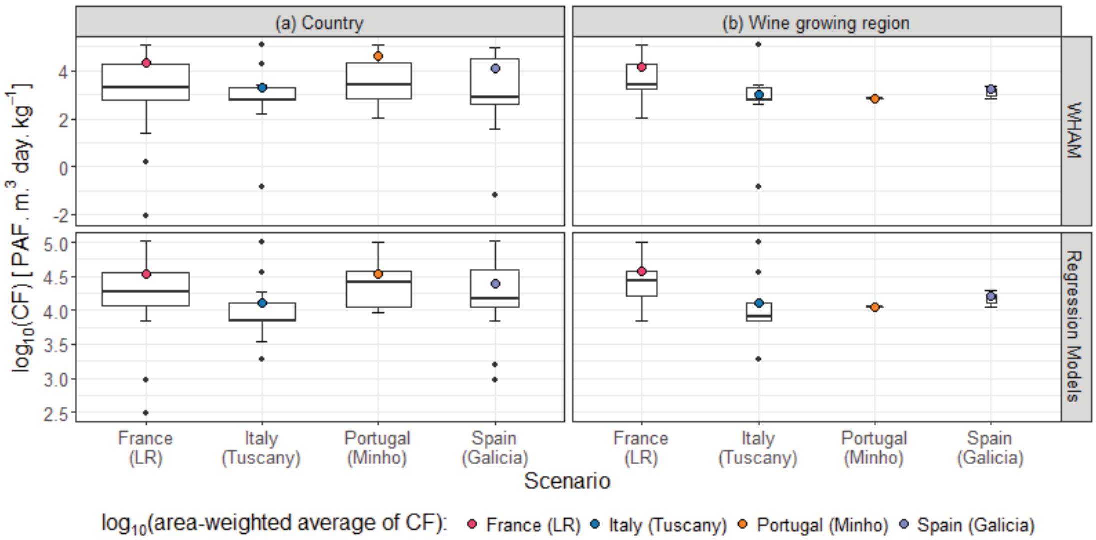


Figure 4.4 Site-dependent CFs for copper terrestrial ecotoxicity at the (a) country level and (b) wine-growing region level.

The aggregated CFs at the country level obtained with empirical regressions extend over 2.5 orders of magnitude for France, 1.7 for Italy, 1 for Portugal, and 2 for Spain (Figure 4.4), which are roughly in the range of the common uncertainty for ecotoxicity assessment (2 orders of magnitude). The aggregated CFs for France, Italy, Portugal, and Spain are in the same order of magnitude and respectively 3.51×10^4, 1.29×10^4, 3.47×10^4, and 2.48×10^4 PAF·m^3·day·kg^{-1}. Except for France and Portugal, these CFs are lower than the global CF (3.27×10^4 PAF·m^3·day·kg^{-1}). The spatial variability of the CFs at the wine-growing region level is in the following increasing order, which is related to the surface of vineyards: Minho (36.9 km^2), Galicia (59.6 km^2), Languedoc-Roussillon (4470 km^2), and Tuscany (447 km^2). In the case of Minho (Portugal), only one soil type is present and, as a result, the CF takes a unique value of 1.13×10^4 PAF·m^3·day·kg^{-1}. The spatial variability of CFs for Galicia (Spain) is negligible, with minimum and maximum values of 1.13×10^4 and 1.94×10^4 PAF·m^3·day·kg^{-1}, respectively. CFs for Tuscany (Italy) and Languedoc-Roussillon (France) span over 1.7 and 1.1 orders of magnitude, respectively. However, the aggregated CF for Tuscany is lower than that for Languedoc-Roussillon (Figure 4.4). The aggregated CFs for Tuscany and Languedoc-Roussillon at the wine-growing region level are 1.27×10^4 and 3.69×10^4 PAF·m^3·day·kg^{-1}, respectively.

When applying the method based on WHAM 6.0, CFs at the country level that correspond to France showed higher spatial variability (approximately 7.2 orders of magnitude), while CFs for Portugal exhibited the lowest spatial variability, around 3 orders of magnitude. CFs for Italy and Spain span over 5.9 and 6.2 orders of magnitude, respectively (Figure 4.4). Aggregated CFs for France, Italy, Portugal, and Spain are 2.04×10^4, 1.88×10^3, 4.16×10^4, and 1.36×10^4 $PAF \cdot m^3 \cdot day \cdot kg^{-1}$, respectively. With the exception of Portugal, aggregated CFs at country level are lower than the global CF (3.27×10^4 $PAF \cdot m^3 \cdot day \cdot kg^{-1}$). The aggregated CF for Italy obtained by applying empirical regression models is around 6.9 times the value calculated with WHAM 6.0, over 1.7 times for France and Spain, and 0.8 times for Portugal. The aggregated CFs at the wine-growing region level for Galicia, Languedoc-Roussillon, Minho, and Tuscany are 1.65×10^3, 1.53×10^4, 6.82×10^2, and 1.08×10^3 $PAF \cdot m^3 \cdot day \cdot kg^{-1}$, respectively, which are lower than the global CF obtained with WHAM 6.0 (3.27×10^4 $PAF \cdot m^3 \cdot day \cdot kg^{-1}$). CFs for Tuscany show higher geographical variability, which is around 5.9 orders of magnitude. CFs for Languedoc-Roussillon span over 3 orders of magnitude, while the spatial variability of CFs for Galicia is around 0.5 orders of magnitude and zero in the case of Minho.

4.2.4.3 Terrestrial Ecotoxicity Impact Score

Figure 4.5 shows the impact score calculated for each scenario by applying CFs for copper terrestrial ecotoxicity at different spatial resolutions and calculated with empirical regression models and WHAM 6.0. Additionally, vertical lines indicate the extent of the impact score that results from applying the minimum and maximum CFs. For both methods, an increase in the spatial resolution of the CFs translates into a reduction in the geographical variability of impacts (Figure 4.5), following the tendency of CF spatial variability (Figure 4.4). Regardless of the method used to compute CFs, when using the aggregated CF for European vineyards, the rank of an impact score is determined by the amount of copper applied in each wine-growing region. This is equivalent to using a generic CF, which is a current practice in LCA. The impact score ranking therefore follows the decreasing order defined by the amount of copper applied in the wine-growing regions that were analyzed: Minho (Portugal), Languedoc-Roussillon (France), Galicia (Spain), and Tuscany (Italy) (Table 4.1 and Figure 4.5). Nevertheless, the impact scores obtained with the global CF derived with empirical regressions are approximately 1.7 times the impact scores calculated with WHAM 6.0 (Figure 4.5).

When using CFs calculated with empirical regression models, the ranking of the country-level impact scores calculated with CFs is the same as the one obtained with CFs for European vineyards (Figure 4.5 a,b, upper panel). This tendency is determined by the amount of copper-based fungicides applied in each wine-growing region, since the ranking of the aggregated CFs is the same regardless of the European vineyard or country resolution. However, the impact score ranking changes when using CFs at the wine-growing region level with respect to that obtained at the European and country resolutions. The ranking of impact scores obtained with CFs at the wine-growing region level in descending order is Languedoc-Roussillon (France), Minho (Portugal), Galicia (Spain), and Tuscany (Italy) (Figure 4.5c, upper panel). There is an inversion of the ranking of impact scores between Languedoc-Roussillon (France) and Minho (Portugal) because the aggregated CFs at the wine-growing region level for Minho (Portugal) is one third of the corresponding aggregated CF at country level.

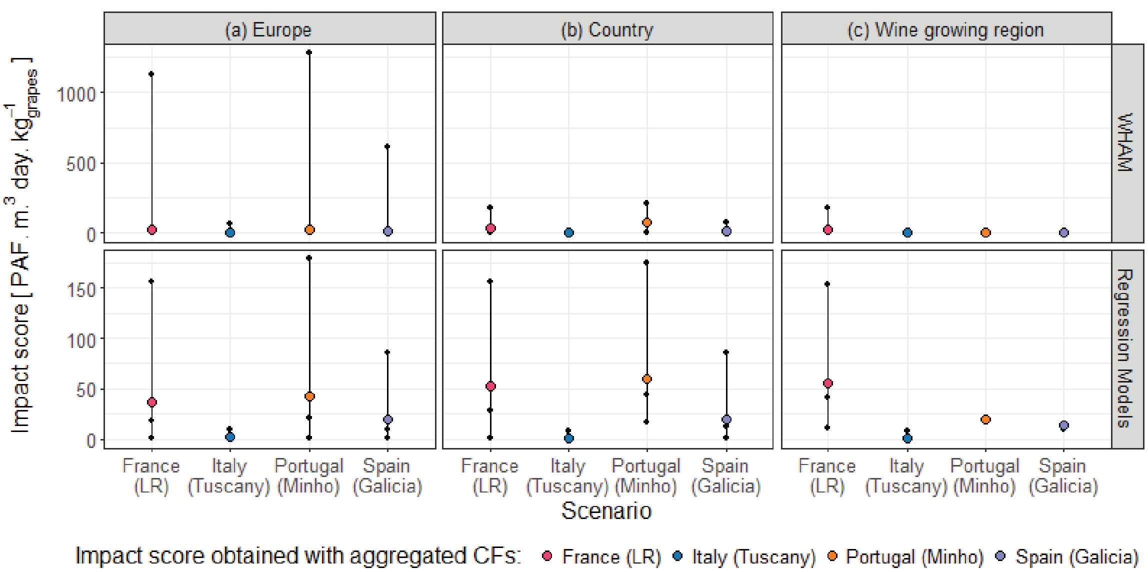


Figure 4.5 Terrestrial ecotoxicity impact score calculated with CFs at different spatial resolutions: (a) Europe, (b) country, and (c) wine-growing regions. Vertical lines indicate the range of impact scores obtained with the minimum and maximum CFs.

A similar pattern is obtained when applying CFs calculated with the method based on the WHAM 6.0 model. The ranking of impact scores obtained with aggregated CFs for European vineyards is equal to the ranking obtained when applying CFs at the country level: Portugal (Minho), France (Languedoc-Roussillon), Spain (Galicia), and Tuscany (Italy). However, the impact score for Minho (Portugal) and Languedoc-Roussillon (France) at the country level is 2.9 and 1.43 times the impact obtained with the aggregated CF for European vineyards, respectively. With regard to the country-level impact scores, Portugal (Minho) ranks first, France (Languedoc-Roussillon) ranks second, and Spain (Galicia) ranks third. However, these positions change when applying aggregated CFs at the wine-growing level. The impact score calculated with CFs at European vineyards is 60 and 8 times the corresponding score obtained with CFs aggregated at wine-growing regions and over 1.3 times for France and Italy.

The ranking of the impact scores of the selected scenarios is mainly determined by the amount of copper-based fungicides applied in each wine-growing region, thus demonstrating the relevance of regionalizing the inventory of agricultural systems since they are highly dependent on spatial conditions, such as climate, soil properties, and agricultural practices. For the analyzed scenarios, the higher amount of copper per kilogram of grapes is approximately 20 times the lower copper application rate (for Tuscany and Minho, respectively). However, the difference in the years of production (2007 for Tuscany and 2008 for the remaining wine-growing regions) limits the comparison and highlights the need to consider the temporal scale in the inventory of viticulture phase (Vázquez-Rowe, Villanueva-Rey, Moreira, et al., 2012). Even so, for the same production year, differences of more or less 50% in copper application rates are observed. Given these results, it would have been acceptable to compute the terrestrial ecotoxicity impact with a CF aggregated at the European vineyard level. However, the advantage of using finer spatial resolutions is the decrease in the uncertainty of the calculated impact score.

4.2.5 Conclusions

In this study, the two methods used to compute CFs make it possible to consider the influence of the geographical variability of soil and agricultural practices on the terrestrial ecotoxicity impact of copper-based fungicides applied in European vineyards. For both methods applied to develop regionalized CFs, an increase in the spatial resolution of CFs translated into a decrease in the spatial variability of the CFs. With the exception of the aggregated CF for Portugal (Minho) at the country level, all the aggregated CFs derived from empirical regression models are greater than the ones derived from the method based on WHAM 6.0 in a range of 0.2 to 1.2 orders of magnitude. Furthermore, CFs calculated with WHAM 6.0 exhibited greater spatial variability with respect to the CFs derived from empirical regression models. One limitation is common to both methods: the fact that CFs are only defined for non-calcareous soils because the TBLMs employed to derive EFs are not applicable to calcareous soils. Further research on how to assess the ecotoxicity of metals in calcareous soils is therefore required. Another limitation of this study is the fact that the essentiality of metal was not considered. This would require more information on the sensitivity of a broader set of terrestrial organisms and the mapping of their spatial distribution.

This study also shows the relevance of including metal speciation and bioavailability in the computation of CFs for copper terrestrial ecotoxicity since neglecting copper speciation in soil potentially overestimates the impact of copper by almost 4 orders of magnitude. Furthermore, this study illustrates the feasibility of including regionalization in both the inventory and impact assessment phases. The spatial variability of the inventory of copper emissions mainly determined the ranking of the impact scores among the wine-growing regions in the case study, highlighting the need to regionalize the inventory of agricultural systems. The regionalized CFs facilitated the computation of aggregated CFs at different spatial levels and, in all the cases, finer spatial resolutions translated into a lower uncertainty, corresponding to the spatial variability.

CHAPITRE 5 ARTICLE 2 : PROSPECTIVE LIFE CYCLE ASSESSMENT OF VITICULTURE UNDER CLIMATE CHANGE SCENARIOS, APPLICATION ON TWO CASE STUDIES IN FRANCE

5.1 Présentation de l'article

Ce deuxième article présente les résultats portant sur l'objectif de développer une approche d'analyse du cycle de vie prospective tenant compte de l'influence du changement climatique sur l'inventaire des pratiques en viticulture et sur les stratégies d'adaptation. La méthodologie est illustrée avec deux vignobles en Val de Loire et en Languedoc-Roussillon, France.

Les auteurs de cet article sont Ivan Viveros Santos, Christel Renaud-Gentié, Philippe Roux, Annie Levasseur, Cécile Bulle, Louise Deschênes et Anne-Marie Boulay. Il a été soumis à *Science of the Total Environment* le 3 juin 2022. Le matériel supplémentaire soumis avec l'article est disponible dans l'annexe B.

5.2 Manuscrit

5.2.1 Abstract

Viticulture needs to satisfy consumers' demands of environmentally sound grape and wine production, while envisaging adaptation options to diminish the impacts of projected climate change on future productivity. However, the impact of climate change and the adoption of adaptation levers on the environmental impacts of future viticulture has not been assessed. This study evaluates the environmental performance of grape production in two French vineyards, one located in the Loire Valley and another in Languedoc-Roussillon, under two climate change scenarios. First, based on grape yield and climate data sets, we assessed the effect of climate-induced yield change on the environmental impacts of future viticulture. Second, besides the climate-induced yield change, we accounted for the impacts of extreme weather events on grape yield and on the implementation of adaptation levers based on the future probability and potential yield loss due to extreme events. The life cycle assessment (LCA) results associated with climate-induced yield change led to opposite conclusions for the two vineyards of the case study. While the carbon footprint of the vineyard from Languedoc-Roussillon is projected to increase by 29%

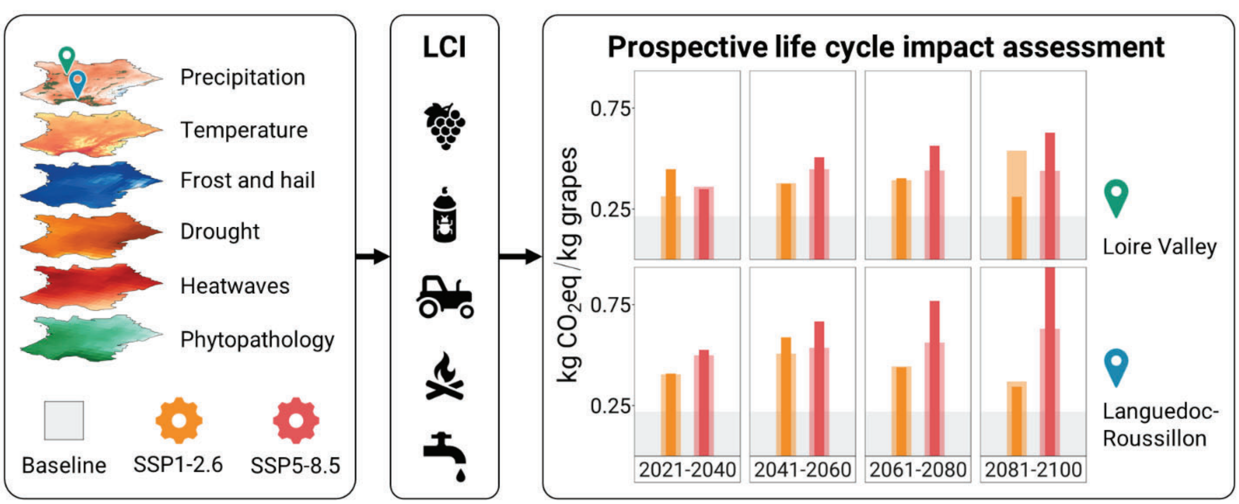
by the end of the century under the high emissions scenario (SSP5-8.5), the corresponding footprint is projected to decrease in the vineyard from the Loire Valley by approximately 10%. However, when including the effect of extreme events and adaptation options, the life cycle environmental impacts of grape production are projected to drastically increase for both vineyards. For instance, under the SSP5-8.5 scenario, the carbon footprint for the vineyard of Languedoc-Roussillon is projected to increase fourfold compared to the current footprint, while it will rise threefold for the vineyard from the Loire Valley. The obtained LCA results emphasized the need to account for the impact of both climate change and extreme events on grape production under future climate change scenarios.

Keywords: Climate change, Life cycle assessment, Environmental impacts, Viticulture, Prospective LCA

Highlights:

- Proposed approach for assessing environmental impacts of viticulture under climate change.
- Climate-induced yield changes point in opposite directions in the case studies.
- Future life cycle impacts of grape production are projected to increase.
- Potential adaptation levers are assessed.
- Extreme events are determinant on the environmental impacts of future grape production.

Graphical abstract



5.2.2 Introduction

5.2.2.1 The issue of the effects of climate change on agriculture and its environmental consequences

Agricultural systems are an integral part of the 2030 Agenda for Sustainable Development, a global engagement to eliminate hunger and poverty, and at the same time guaranteeing a decrease of socio-economic and environmental impacts (Sala et al., 2017). However, this goal constitutes a prominent challenge since agriculture causes considerable environmental impacts, being a key contributor to land-use change, biodiversity loss, pollution of terrestrial and aquatic ecosystems, depletion of freshwater resources, and climate change (Springmann et al., 2018). Greenhouse gas (GHG) emissions from Agriculture, Forestry and Other Land Use (AFOLU) activities represent 23% of total anthropogenic GHG (IPCC, 2021), and recently it has been estimated that climate change has caused a loss of agricultural productivity of around 21% since 1961 (Ortiz-Bobea et al., 2021). Hence, agriculture is a major contributor to climate change, while concurrently being affected by it. Climate change affects agricultural systems in direct and indirect ways. Direct impacts correspond to changes in abiotic factors, namely the effects of increased atmospheric CO_2 concentration and temperature, and changes in precipitation regimes, as well as the impact of extreme weather events such as hail, heatwaves, and flooding on crop yield. Indirect impacts are related to changes in environmental conditions such as the timing of weeds, pests and diseases, irrigation needs and changes in soil organic matter (Niero et al., 2015; Ortiz-Bobea et al., 2021). Besides ambitious mitigation policies to limit global warming to 1.5 °C above the pre-industrial level, adaptation strategies will be required in the agricultural sector due to substantial shifts in the local climate for the coming decades (IPCC, 2018). For this reason, evaluating the environmental impacts of agriculture under future climate conditions constitutes a pressing need to devise adequate mitigation and adaptation strategies for the forthcoming years (E. K. Lee et al., 2020; Sala et al., 2017).

Life cycle assessment (LCA) is a holistic method that quantifies the potential environmental impacts of a product or service throughout its entire life cycle, from raw material extraction to end of life (ISO, 2006a, 2006b). LCA has been recognized as a crucial decision-making tool to enhance the environmental performance of agricultural systems (Sala et al., 2017). Furthermore, combining LCA and future-scenarios thinking has been recently advocated in change management and decision-making to tackle global environmental challenges, since LCA facilitates anticipating the potential environmental impacts of novel technologies and products (Bisinella et al., 2021) or assessing them under future environmental conditions (Sala et al., 2017). Three future-oriented LCA studies have explored the environmental impacts of agricultural systems under projected climate change scenarios (Garba et al., 2014; E. K. Lee et al., 2020; Niero et al., 2015). Garba et al. (2014) coupled a crop system model and LCA to investigate the future GHG emissions from corn and soybean production in Gainesville, Florida under projected changes in temperature, precipitation, and CO_2 concentrations. Niero et al. (2015) evaluated the future environmental impact of spring barley based on the results of experiments simulating future Danish climate. A recent study applied machine learning to investigate county-level global warming and eutrophication impacts of corn production for the period 2022-2100 across U.S. Midwest states according to four climate change scenarios ranging from “high emissions or business-as-usual” to “low emissions or strong mitigation” pathways (E. K. Lee et al., 2020).

5.2.2.2 Grapevine context and specificities

Grapevine (*Vitis vinifera*) is a widely grown perennial crop: as of 2019, it covered an area of 7.3 million hectares and produced 85 million tons of grapes (OIV, 2022). Moreover, viticulture and winemaking are a very important cultural legacy both economically and socially in many parts of the world (David Santillán et al., 2019). Nevertheless, like any economic activity, viticulture and winemaking have an impact on the environment. For instance, in France in 2011, vineyards were responsible for 20% of the pesticides used, while they accounted for only 3.7% of cultivated land (Ugalde et al., 2021). Pesticides lead to multiple environmental concerns such as surface and groundwater pollution, as well as to potential ecotoxicological and toxicological impacts resulting from their fate into the air, water, and soil compartments (Beauchet et al., 2019; Christel Renaud-Gentié et al., 2015; Ivan Viveros Santos et al., 2018). These environmental concerns have raised the need for an efficient and environmentally-sound control of diseases and pests despite difficult climatic conditions (Beauchet et al., 2019; Marín et al., 2021). Adaptation to climate change is another major challenge facing the viticulture sector, since climate change has already caused the advancement of harvest dates, a rise of disease in wetter regions, recurrent water stress in Mediterranean vineyards, wines deficient in freshness and aromatic composition and an excessive increase in alcohol content, loss of production due to more frequent extreme events, and an increased concern for spring frost damage in some winegrowing regions (Leolini et al., 2018; Marín et al., 2021; David Santillán et al., 2019; Santos et al., 2020; van Leeuwen et al., 2019). However, the projections of European grapevine productivity point in both directions. Increased dryness across southern regions will favour decreased yields, while increased CO_2 may partially compensate dryness effects, leading to yield increases in central and northern Europe (Helder Fraga et al., 2016; van Leeuwen et al., 2019). Additional concerns are related to repercussions on profits and production costs through the logistics network (Sacchelli et al., 2017) and to potential changes in the suitability of land for viticulture (M Moriondo et al., 2013).

Because of the potential negative impacts of climate change on grape yields, as well as on major crops namely wheat, rice, maize and soybean, several studies have addressed this question by means of statistical models and process-based crop models (Helder Fraga et al., 2016; Kukal & Irmak, 2018; Lobell & Burke, 2010; Marco Moriondo et al., 2015; Zhao et al., 2017). Statistical regressions have been used to relate time series of crop yield to changes in selected climate variables. More specifically, detrending methods have been employed in regression models to eliminate the influence of non-climatic factors such as genetic improvements, crop and soil management advancements, and technological improvements (Kukal & Irmak, 2018; Zhao et al., 2017). It has been shown that statistical models reproduce well the process-based model responses to changes in temperature. Besides, regression models seem to be more suitable for wider geographical scales of analysis, which is in line with the scales at which climate projections are more available and reliable (Lobell & Burke, 2010). In the case of grapevines, various process-based models have been developed to simulate the entire phenological cycle, or to study particular processes such as water dynamics in the soil and plant gas exchange (Marco Moriondo et al., 2015). For instance, a grapevine growth simulator was used to calculate current and future projections of wine production in Chianti Classico region (Sacchelli et al., 2017). The STICS model was used to assess the impact of climate change on viticultural yield and phenology for European vineyards under two climate change scenarios. While some model deviations were identified, the model outputs showed a good agreement with records of phenology, leaf area index, yield, and water/nitrogen stress conditions. However, running STICS for the whole European vineyard for a period of 26 years is computationally very expensive (around two million iterations), consequently some computational strategies must be implemented to decrease the number of required iterations (Helder Fraga et al., 2016).

Mitigation and adaptation are complementary actions for decreasing and controlling the impacts of climate change in agriculture. For instance, a lever of GHG mitigation is decreasing the use of tractor for viticultural operations (Beauchet et al., 2019). However, defining adaptation options for viticulture is more challenging than for other crops because the grapevine is a perennial crop, which implies that investments in adaptation must persist for many decades (D Santillán et al., 2020). Naulleau et al. (2021) classified technical adaptation options according to their long- or short-term characteristics. The former options relate to perennial practices, while the latter correspond to annual practices. Long-term options include site selection, farm strategy, and selection of plant material (Gutiérrez-Gamboa et al., 2021; Naulleau et al., 2021; Neethling et al., 2017; Wolkovich et al., 2018). Among the short-term adaptation options for viticulture to climate change are irrigation, new vine management practices, and protection against extreme weather events (anti-hail nets, fans, heaters) (Gutiérrez-Gamboa et al., 2021; Naulleau et al., 2021; David Santillán et al., 2019; Wolkovich et al., 2018). However, the distinction between short- and long-term adaptation levers is site-dependent. For instance, in the Anjou-Saumur winegrowing sub-region, France, irrigation was identified as a long-term option rather than a short-term one (Neethling et al., 2017).

Previous studies have used different approaches to evaluate adaptation strategies in viticulture, namely suitability mapping, empirical models, agent-based models, and process-based models (Naulleau et al., 2021). Even though suitability maps derived from biocliomatic indices point to a remarkable change in the landscape for European viticulture under future climate change, it has been acknowledged that adaptation strategies can attenuate these potential shifts in viticulture suitability (M Moriondo et al., 2013). Santillán et al. (2019) developed a framework based on agro-climatic indices to assess the relative magnitude of adaptation effort for the major winegrowing regions. Subsequently, the required level of adaptation has to be refined at finer scales, accounting for local conditions, winegrowers' perceptions of climate change, and available resources. An agent-based model was developed to investigate grapevine phenology and grape ripening under future environmental conditions driven by climate change. The model accounts for viticultural practices and adaptation strategies, as well as economic and technical constrains at plot scale. Even though the model provides relevant output for analysis at local level, the main constraint to deploy the model corresponds to the huge amount of required data (Tissot et al., 2017). Sacchelli et al. (2017) developed a mix-method approach combining an economic model, a grapevine growth simulator, and a cognitive mapping technique to assess the economic performance of a viticultural farm when it implements adaptation measures to climate change.

5.2.2.3 Goal and scope of the study

The aim of this study is to conduct a prospective life cycle assessment of viticulture under climate change scenarios, according to Shared Socioeconomic Pathways (SSPs) developed by the Intergovernmental Panel on Climate Change (IPCC). SSPs are emissions scenarios driven by diverse socioeconomic assumptions (IPCC, 2021). More specifically, we considered two SSPs (SSP1-2.6 and SSP5-8.5) corresponding to trajectories of low and high GHG emissions, according to four periods from 2021 to 2100. In the first level of analysis, we considered a *ceteris paribus* situation in which only projected changes in temperature and total precipitation drive changes in future grape yield. The underlying research questions addressed at this level of analysis were: (1) What is the potential role of climate-induced yield change on the environmental performance of viticulture? (2) How does this projected environmental performance compare to reported inter-annual variability? In the second level of analysis, we accounted for the impact of extreme weather events (hail, drought, heatwaves, frost, and phytopathology) on grape yield and on the implementation of adaptation options based on future probability and potential level of damage of these extreme weather events. At this level of analysis, the research question was: (3) What is the influence of adaptation options on the environmental performance of viticulture under climate change?

The goal and scope of the study, as described above, were deployed on two contrasting vineyards located in the Loire Valley (temperate oceanic climate), and Languedoc-Roussillon (dry and warm temperate Mediterranean climate), France. The effects on pollutant emissions and natural resource consumption as well as on yield were modelled for these two examples in order to calculate the resulting environmental impacts using LCA framework.

5.2.3 Methods

5.2.3.1 Case studies description

Two vineyards located in two French winegrowing regions associated with different production characteristics and climates were analyzed in this study, one from the Loire Valley region in western France and another from Languedoc-Roussillon in southern France (Figure 5.1, Table 5.1). The pathway of technical operations (PTO), that is the set of viticulture practices, implemented annually for a given production objective on each plot are examples chosen within the range of technical options employed in each winegrowing region. The PTO applied in the analyzed vineyard from Loire Valley is representative of low input conventional vineyard management. It is characterized by minimum application rates of phytosanitary products, and by a low number of interventions on the field (Christel Renaud-Gentié et al., 2014). However, in the Loire Valley region, the temperate oceanic climate could promote pest and disease development, as well as vine vigour, which subsequently could lead to a higher number of viticulture operations, namely treatments for disease and pest control, and weeding (Christel Renaud-Gentié et al., 2020). In contrast, the PTO implemented in the studied vineyard from Languedoc-Roussillon is representative of high input conventional vineyard management. It is characterized by a high application rate of phytosanitary products, as well as the use of herbicides for weed control (Table B.1). The Languedoc-Roussillon winegrowing region is characterized by Mediterranean climate and maritime effects limit summer temperature extremes or winter frost during key grapevine phenological growth stages (Lereboullet et al., 2014).

Figure 5.1 Location of the analyzed vineyards in this study.

Table 5.1 Climatic characteristics of the analyzed vineyards in this study (Data Météo-France).

Case study	**Loire Valley**	**Languedoc-Roussillon**
Annual T_{mean} (°C)	12.4	15.1
Annual T_{min} (°C)	7.8	10.3

Case study	Loire Valley	Languedoc-Roussillon
Annual T_{max} (°C)	17.1	19.8
Growing season T_{mean} (°C)	16.2	18.6
Annual precipitation (mm)	556	629
Summer precipitation (JJA) (mm)	123	79
Average solar irradiation (kWh m^{-2} day^{-1})	3.5	5.1

5.2.3.2 Life Cycle Inventory (LCI)

The life cycle inventory (LCI) consists in quantifying all pollutants emitted to air, water and soil, and all natural resources used during the entire process of winegrowing (cradle to grave). The LCI relied on data obtained from winegrowers. The collected data includes the characteristics of the farm, the operations (fertilization, crop protection, harvesting) and their frequency, as well as the amount of agrochemical products applied, the labour times, the materials used, as well as agricultural machinery and equipment used. In the case of the vineyard from Languedoc-Roussillon, data was gathered directly from Agreo, a traceability system used in that vineyard domain (Bellon-Maurel et al., 2015).

Direct field emissions resulting from the application of fertilizers, soil amendments, and metal-based fungicides were calculated with the models shown in Table 5.2. These emission models were programmed in AGEC-LCI, a Visual Basic for Applications (VBA) Excel tool that generates inventory reports compatible with LCA software such as openLCA and SimaPro (I Viveros Santos et al., 2020). Apart from copper, active-substance emissions from pesticides were computed with PestLCI 2.0, using a version customized to viticulture (Christel Renaud-Gentié et al., 2015).

Table 5.2 Emissions models for computing direct environmental emissions with AGEC-LCI (I Viveros Santos et al., 2020) (https://iviveros.github.io/agec-lci-tutorial).

Modelled phenomenon	Unit	Model or method	Reference
Soil erosion	[t soil/ha/year]	RUSLE2	Foster (2005)
Ammonia emissions	[kg NH_3/ha]	Tier 2 of EMEP/EEA	EMEP (2009)

Modelled phenomenon	Unit	Model or method	Reference
Nitrogen oxides emissions (NOx: NO and NO_2)	[kg NO_2/ha][1]	Tier 1 of EMEP/EEA	EMEP (2009)
Nitrate emissions	[kg NO_3^-/ha]	SQCB	Faist Emmenegger et al. (2009)
Nitrous oxide emissions	[kg N_2O/ha]	Tier 1 of IPCC (2006)	Eggleston et al. (2006)
Phosphorus emissions	[kg P/ha] and [kg PO_4^{3-}/ha]	SALCA-P	Nemecek and Schnetzer (2011)
Trace metals (TM) emissions {x|x= Cd, Cr, Cu, Hg, Ni, Pb, Zn}	[kg TMx/ha]	SALCA-ETM	Freiermuth (2006)

The foreground inventory data was modelled in SimaPro v8.5.2.2 software according to the product systems shown in Figure B.1 and B.2. The inventory of background processes relied on ecoinvent v3.5 database and Agribalyse® (Colomb et al., 2015) for some agricultural machinery contextualized to France. The functional unit was one kilogram of grapes produced, in order to account for yield differences of baseline and future scenarios.

5.2.3.3 Data sources

5.2.3.3.1 Grape yield and vineyard surface

Data sets of viticultural yield at the level of French Department for the period 2000-2020 were retrieved from the French Ministry of Agriculture and Food Statistics Service (Agreste, 2021). The beginning of the study period corresponds to the earliest year with grape yield records. In total, Agreste database of viticultural yield holds 36450 records according to 21 reported years, 121 geographic identifiers, 3 categories of vines, and 5 indicators (Table B.2). Even though this study

[1] A conversion factor of 46/14 was applied to nitrogen emissions (kg NOx-N/Kg N applied) to translate these emissions in terms of NO_2, which is in line with ecoinvent database.

focuses on vineyard plots within only two French Departments, to take advantage of the extent of data availability, we extracted a set of 1260 records from Agreste database of grape yield, according to 21 years and 60 Departments of Metropolitan France (filter criteria shown in Table B.3). Grape yields obtained from Agreste are reported in quintals per hectare, consequently, a conversion was made to metric tonnes per hectare to make the data suitable for further computations.

The vineyard area over Europe was obtained from the CORINE program of the European Environment Agency (EEA, 2017b) and Python was used to extract the coverage of French vineyards by means of the clip algorithm from GeoPandas module (Jordahl et al., 2020) with a polygon shapefile of Metropolitan France as mask layer. Subsequently, since the spatial resolution of available viticultural yield is at French Department level, GeoPandas (Jordahl et al., 2020) was employed to relate vineyard surface and administrative divisions defined by a polygon shapefile of French Departments (Table B.4 and Figure B.3).

5.2.3.3.2 Climate data

One specific objective of this study is to investigate the impacts of recent climate on grape yield to subsequently estimate the impacts of projected change in temperature and precipitation on future grape yield. The former analysis was performed based on a climate data set retrieved from the Agri4Cast Resource Portal, established and maintained by the Joint Research Centre of the European Commission (Biavetti et al., 2014). The Agri4Cast data set comprises eight daily meteorological variables from 1979 to the last calendar year completed, interpolated to a 25 x 25 km regular grid over Europe (Table B.5). The Agri4Cast data set has been extensively employed in agricultural studies (Bradshaw et al., 2019; Charalampopoulos et al., 2021; Korycinska & Baker, 2017) because of its consistency and high accuracy in temporal and spatial resolution, which makes it a convenient resource for agroclimatic and agrometeorological analyses (Mavromatis & Voulanas, 2021). From the Agri4Cast data set, we collected mean air temperature (°C) and the sum of precipitation (mm) over France for the period 2000-2020 (so the temporal scale is coherent with that of Agreste data set), which generated a subset of 7,694,013 rows across 1003 grid points (Fig S4). Then, the data set was filtered according to the growing season of wine grapes (March to September), which reduced the data set to 4,507,482 rows.

To calculate the difference between future and present climate during the growing season of wine grapes, monthly values of average temperature (°C) (Figure B.5) and total precipitation (mm)

(Figure B.6) were retrieved from WorldClim 2.0 for five periods at 2.5 min resolution (Fick & Hijmans, 2017). The first period extends from 1970 to 2000 and corresponds to present climate or baseline scenario. The other four periods are 20-year periods of projected climate for the near future (2021-2040), mid-century (2041-2060, 2061-2080), and the end of the century (2081-2100) derived by the CMIP6 (Coupled Model Intercomparison Project Phase 6) (Fick & Hijmans, 2017; Petrie et al., 2021). The climate change scenarios included in this study are derived from the Global Circulation Model (GCM) IPSL-CM6A-LR and two Shared Socioeconomic Pathways (SSPs) (SSP1-2.6 and SSP5-8.5) corresponding respectively to trajectories of “low GHG emissions or strong mitigation” and “high GHG emissions or business-as-usual” (Table B.6). In line with Wolkovich (2018), we considered only one GCM to calculate projected changes in temperature and precipitation, since the purpose of our study is to compute the relative impact of climate change on grape production. Moreover, projections of climate change derived from IPSL-CM5A-LR have been used to compute robust climatic indices for the Mediterranean region (D Santillán et al., 2020).

5.2.3.3.3 Agroclimatic indicators

Apart from hail, to project the potential impacts of extreme events on future grape yield, we used agroclimatic indicators data sets obtained from the Climate Data Store (CDS) of the Copernicus Climate Change Service (C3S) (Copernicus, 2021), namely consecutive frost days (CFD, Figure B.7), consecutive dry days (CDD, Figure B.8), warm-spell duration index (WSDI, Figure B.9), and warm and wet days (WW, Figure B.10).

5.2.3.4 Climate-induced yield impacts

To assess the relationship between the time series for grape yield and climate, it is required that both grape yield and climate data sets are at the same temporal and spatial scales. We selected a department as an adequate scale for inter-comparison since grape yields from Agreste (2021) are reported at this spatial resolution. Moreover, calculating the impacts of climate on grape yields on a department scale entails a potential increase of the application of these results on future planning scenarios in viticulture. Surface data sets of average temperature and total precipitation corresponding to the growing season of grapes were calculated according to 1003 grid points from the Agri4Cast data set (Figure B.4) (Biavetti et al., 2014), using the inverse distance weighting (IDW) interpolation technique implemented in the R package “gstat” (Gräler et al., 2016; Pebesma,

2004) (Figure B.11 and Figure B.12). After interpolating climate variables, zonal statistics at department resolution were calculated using the R package "exactextractr" (Baston, 2020). Overall, we obtained data sets of department-averaged growing season total precipitation and average temperature for 60 French Departments for the period of 2000-2020, which corresponds to the temporal and spatial scales of the Agreste data set (Table B.2).

With both climate and grape yield data sets at the same spatial and temporal scales, to evaluate the sensitivity of grape yields to climate, grape yields and climate variables were detrended by means of linear regression. Detrending is a common method in agricultural research based on the first-difference time series for yield and climate (namely the difference in values from one year to the next) as the dependent variable and time as the independent variable (Kukal & Irmak, 2018; Lobell & Field, 2007). The purpose of this operation is to minimize the effects of other non-climatic variables, such as genetic improvement, technological advancement, and soil and crop management progress (Lobell & Field, 2007). Detrending produced residuals of yield ($\Delta Yield$) (kg ha^{-1} yr^{-1}), average temperature (ΔT_{avg}) (°C yr^{-1}) and total precipitation ($\Delta \mathrm{ppt}$) (mm yr^{-1}). Successively, we performed multiple linear regressions with yield residuals ($\Delta Yield$) as the dependent variable and residuals of average temperature (ΔT_{avg}) and total precipitation ($\Delta \mathrm{ppt}$) as explanatory variables (see Figure B.13 and Figure B.14 along with a brief description of detrending). The slope coefficients of these multiple linear regressions correspond to yield change per unit change in mean temperature (kg ha^{-1} °C^{-1}) and total precipitation (kg ha^{-1} mm^{-1}).

Next, we generated maps of change in average temperature (Figure B.15) and total precipitation (Figure B.17) during the growing season of wine grapes by calculating the difference between current and future climate, according to four future periods and two SSPs (SSP1-2.6 and SSP5-8.5) derived by the GCM IPSL-CM6A-LR (Table B.6). Successively, zonal statistics of these maps were computed in R v4.1.2 by means of the package "exactextractr" (Baston, 2020), resulting in department-level values of projected climate change.

According to Equation 5.1, the expected change in grape yield ($E(YC)_{t,s}$), in kg ha^{-1}, due to projected climate change for each period (t) and SSP (s) was computed by adding the product of the sensitivity of grape yield β_c to each climate variable (c) (i.e., the slope coefficients of multiple linear regressions, kg ha^{-1} °C^{-1} and kg ha^{-1} mm^{-1} for average temperature and total precipitation, respectively) and the projected change in average temperature (°C) and total precipitation (mm)

($\Delta C_{c,t,s}$) (Figure B.16 and Figure B.18). This approach is consistent with other LCA studies that have used historic trends to forecast the environmental performance of product systems under future conditions, whether technical (Beloin-Saint-Pierre et al., 2020) or environmental (E. K. Lee et al., 2020). Finally, according to Equation 5.2, the expected future grape yield ($E(Y)_{t,s}$), in kg ha^{-1}, for period (t) and SSP (s) was obtained by the algebraic addition of expected change in grape yield ($E(YC)_{t,s}$) to the grape yield of baseline scenarios ($Y_{baseline}$), in kg ha^{-1}.

$$\mathrm{E}(YC)_{t,s} = \sum_{c} \beta_{\mathrm{c}} \cdot \Delta \mathrm{C}_{c,t,s} \tag{5.1}$$

$$E(Y)_{t,s} = Y_{baseline} + E(YC)_{t,s} \tag{5.2}$$

5.2.3.5 Effects of extreme events and adaptation strategies

In this study, we adapted the methodological approach developed by Sacchelli et al. (2017) to account for the impact of extreme events on future grape yield. According to Equation 5.3, the expected yield loss rate ($E(YLR)_{t,s}$) (%) for each period (t) and SSP (s) due to an extreme event – e – was calculated as a function of the future probability of the extreme events – $P(e)_{t,s}$ (%) – and the potential level of damage - d_e. In line with Sacchelli et al. (2017), the probability of extreme events was treated as an independent variable, consequently, the probabilities of change in grape yield resulting from extreme events were aggregated (Equation 5.3). However, this approach does not allow to account for potential synergistic effects of drought and heatwaves that have been reported in the literature (H Fraga et al., 2018).

$$E(YLR)_{t,s} = \sum_{e=1}^{m} P(e)_{t,s} \cdot d_e \tag{5.3}$$

As described in Table 5.3, for frost, drought, heatwaves, and phytopathology, we computed the future probability $P(e)_{t,s}$ (%) of extreme event e during the growing season of wine grapes based on agroclimatic indicators for future periods (2021-2040, 2041-2060, 2061-2080, and 2081-2100) derived from the GCM IPSL-CM5A-LR and two SSPs. Whereas for hail, we used data on projected change in summer mean temperature (Fick & Hijmans, 2017). Therefore, a total of eight combinations of periods and SSPs were considered. Regarding the potential level of damage (d_e) (%), we considered reported values in the literature (Table 5.3).

Table 5.3 Definition of extreme events.

Extreme events	**Note**
Hail	The current probability of hail was based on literature review (Berthet et al., 2011; Raupach et al., 2021). Likewise, the current hail damage on grape yield was extrapolated from an empirical study on table grapes that reported a yield reduction of 39% after a hail event (Petoumenou et al., 2019). For future hail damage, we considered a correlation between summer mean temperature and hail damage in France, according to which, an increase of one degree leads to a 40% rise in hail damage (Berthet et al., 2011; Raupach et al., 2021).
Frost	Based on the maximum number of consecutive frost days (CFD) for the season March-April-May (MAM). For a frost day, the minimum daily air temperature is < 0°C (Copernicus, 2021) (Figure B.7), which is in line with the temperature threshold defined in other studies assessing spring frost damage (Molitor et al., 2014). The probability of frost (p_f) was approximated by $p_f = CFD/n$ where n is the number of days during the season MAM and CFD is the mean value of this parameter for a given period (historical or future). The underlying assumption was that spring frost risk increases with CFD. This does not allow to account for the duration of sub-zero temperatures on the severity of frosts; however, the number of CFD are projected to decrease. Regarding the level of yield damage caused by spring frost, we considered a historical value in European vineyards of 39% by frost episode (Molitor et al., 2014).
Drought	Based on the maximum number of consecutive dry days (CDD) during the grape-growing season (approximated by the period March-August) (Copernicus, 2021) (Figure B.8). A threshold of drought (D) was defined according to the reported CDD for most wine regions in France over the period 1986-2015 (C. Yang et al., 2022). Then, the probability of drought was calculated as $p_{drought} = CDD/D$. Moreover, we considered the mean

Extreme events	**Note**
	potential yield loss rate due to seasonal drought of 30% for France over the period 1986-2015 (C. Yang et al., 2022).
Heatwaves	Following Fischer and Schär (2010), multi-day heatwaves were calculated based on the warm-spell duration index (WSDI) (Copernicus, 2021) (Figure B.9). However, instead of a spell of six days, we considered the definition of a heatwave as a spell of at least nine consecutive days (Helder Fraga et al., 2020). Furthermore, we considered the highest level of damage (35%) by heatwave on grape yield simulated for France over the period 1986-2015 (Helder Fraga et al., 2020).
Phytopathology	Warmer conditions combined with precipitation may trigger pests and disease pressure in viticulture, particularly downy mildew (H Fraga et al., 2013; Santos et al., 2020). However, there are still uncertainties on the correlation between disease occurrence and climatic data (Ollat et al., 2018). For this reason, in line with Sacchelli et al. (2017), we considered a low-level increase in pesticide application with respect to the application rate of baseline scenarios. We hypothesized a 5% increase in pesticide application per warm and wet days (WW) during the growing season (Copernicus, 2021) (Figure B.10).

According to the literature review, we identified some adaptation levers that correspond to incremental changes in viticulture to attenuate the impacts of extreme weather events (Table 5.4). For the implementation of the adaptation levers, we considered that farmers have a high level of risk aversion and activate the corresponding adaptation option when the expected yield loss rate is greater than or equal to 1% (the term $P(e)_{t,s} \cdot d_e$ of Equation 5.3) for a given extreme event (e). Besides, we considered that activating the corresponding adaptation option completely mitigates the associated yield loss rate. Consequently, the expected future grape yield $E(Y*)_{t,s}$ (kg ha^{-1}) for period t and SSP s when adaptation options are activated was calculated according to the second case of Equation 5.4, reflecting the fact that no adaptation was considered for heatwaves (Table

5.4) and that adaptation levers are activated if their potential yield reduction is greater or equal than one. Finally, the first case of Equation 5.4 corresponds to the situation in which no adaptation is activated.

$$E(Y*)_{t,s} = \begin{cases} E(Y)_{t,s} \cdot \left(1 - \sum_{e=1}^{m} P(e)_{t,s} \cdot d_e\right) \\ E(Y)_{t,s} \cdot \left(1 - \sum_{e=1}^{m} P(e)_{t,s} \cdot d_e\right) \; if \; \left(P(e)_{t,s} \cdot d_e < 1 \; and \; e = heatwave\right) \end{cases} \tag{5.4}$$

The LCIA results of adaptation levers were computed according to the description presented in Table 5.4 and with the IMPACT World+ methodology as described in section 2.1. Furthermore, the LCIA results account for the projected increased in phytosanitary treatments as described in Table 5.4.

Table 5.4 Adaptation strategies

Extreme events	**Adaptation lever**	**Note**
Hail	Anti-hail net	The LCI of anti-hail nets was modelled according to an LCA study on HDPE nets (Dassisti et al., 2016).
Frost	Among the adaptation options against frost are fans, heaters, candles (Sacchelli et al., 2017), and wood-burning (Frota de Albuquerque Landi et al., 2021).	In this study, we considered oak wood-burning as the selected anti-frost lever. The wood-burning system was modelled according to a study that estimated that 5000 kg of wood could supply 315 W/m^2 to protect one hectare of a vineyard for 8 hours (Frota de Albuquerque Landi et al., 2021).
Drought	Irrigation	Irrigation demands were estimated based on a study that computed irrigation requirements under future climatic conditions in a French vineyard watershed (Naulleau et al., 2022). It was calculated that under RCP8.5 scenario, the future base

Extreme events	Adaptation lever	Note
		irrigation requirement is 25.2 mm (intercept), which increases by 1.01 mm per each decreased mm in precipitation (slope). While irrigation demands depend on other factors such as soil types, research has shown that irrigation requirements follow the patterns of precipitation during the growing season of grapes (H Fraga et al., 2018), which justifies using this proxy for computing the irrigation requirements.
Heatwaves	Modification of pruning (David Santillán et al., 2019) and heat-tolerant varieties (Wolkovich et al., 2018) are adaptations levers against heatwaves.	In this study, we did not consider adaptation options against heatwaves since they imply transformative changes. While changing variety is a foreseen adaptation lever, it will require new laws and regulations that currently restrict what varieties can be cultivated in a given region (Wolkovich et al., 2018). Moreover, it is difficult to estimate the effect of pruning modification and new grape varieties on grape yield under future climate change scenarios.
Phytopathology	Increase of phytosanitary treatments	We considered the estimated increase in pesticide application described in Table 5.3.

5.2.3.6 Life Cycle Impact Assessment indicators (LCIA)

The life cycle impact assessment (LCIA) was calculated using IMPACT World+ methodology, which is the update of the IMPACT 2002+, LUCAS and EDIP methods (Bulle et al., 2019). In order to facilitate the interpretation of the LCIA results of both present and future scenarios, their environmental performance was assessed according to one midpoint (v1.28) and two endpoint indicators (v1.49) from IMPACT World+. Regarding midpoint indicators, we selected the category climate change in the short term (over the first 100 years after the emission) for the carbon footprint (kg CO_2eq). Concerning the endpoint indicators, we considered the impacts on ecosystem quality (EQ) measured in terms of the potentially disappeared fraction of species over a surface area of one square meter over one year (PDF·m^2·yr), and the impact on human health (HH) measured in disability-adjusted life years (DALY). The EQ endpoint encompasses freshwater ecotoxicity, freshwater acidification, marine acidification, terrestrial acidification, freshwater eutrophication, marine eutrophication, ionizing radiation, land transformation and land occupation, and water availability. The HH endpoint comprises human toxicity cancer, human toxicity non-cancer, particulate matter formation, photochemical oxidant formation, ozone layer depletion, ionizing radiation, and water availability (Bulle et al., 2019). The contribution of climate change to EQ and HH endpoints was excluded to prevent double counting.

5.2.4 Results and discussion

5.2.4.1 Projected climate-induced yield impacts

Figure 5.2 shows the spatial variability of grape yield sensitivity to climate variables at the scale of French Departments, that is, the change in grape yield (kg/ha) per 1 °C rise in temperature (Figure 5.2 a) and per 10 mm rise in precipitation (Figure 5.2 b). Negative values indicate that the grape yields decrease with temperature or precipitation increase, whereas positive values indicate that they increase with temperature or precipitation rise. With respect to grape yield sensitivity to temperature, it was found that 38 out of 60 Departments exhibited negative values, accounting for approximately 54% of the French vineyard surface. A similar figure was found regarding grape yield sensitivity to precipitation, for which 37 out of 60 Departments showed negative values, corresponding to around 57% of the French vineyard surface.

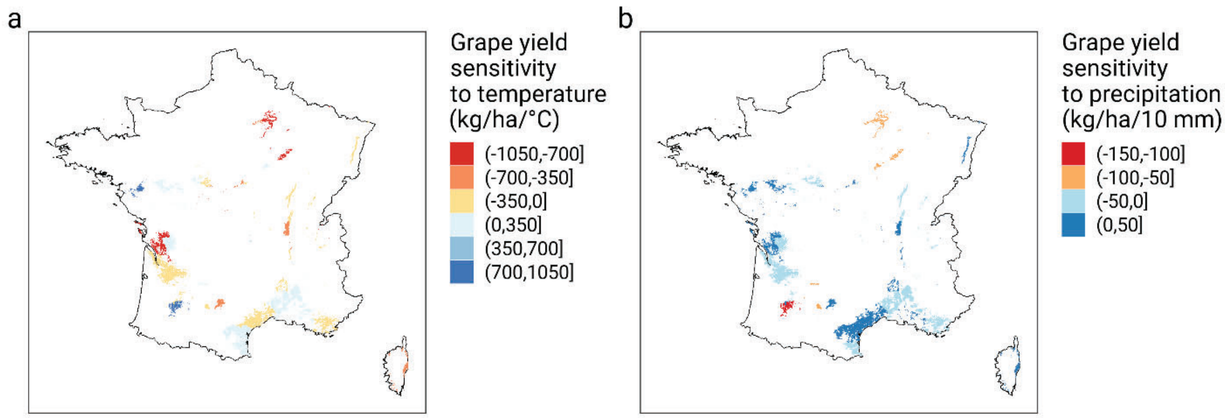


Figure 5.2 Grape yield sensitivity to (a) temperature (kg ha^{-1} $°C^{-1}$) and (b) precipitation (kg ha^{-1} 10 mm^{-1}) over the period 2000-2020

The computed grape yield sensitivities to climate variables at the level of Department (Figure 5.2) were used as a proxy of the grape yield sensitivity to climate of each vineyard of the case study (Figure 5.1), and were combined with the projected changes in mean temperature and total precipitation (Table 5.5) to compute climate-induced yield impacts. We acknowledge that doing so introduces spatial uncertainty at the level of the plot vineyard, however, this scale issue is not exclusive to our study, since climate projections at coarser scales do not perfectly replicate local growing conditions (Naulleau et al., 2022). Regarding the vineyards of the case study, temperature increase was found to adversely affect grape yield in Languedoc-Roussillon, with a value of -233.7 (kg ha^{-1} $°C^{-1}$). On the other hand, in the Loire Valley, the sensitivity of grape yield to temperature was estimated at 151.9 (kg ha^{-1} $°C^{-1}$), which implies that temperature rise in this vineyard is favourable to grape yield. With respect to grape yield sensitivity to precipitation, both vineyards of the case study have positive values of 13.4 (kg ha^{-1} 10 mm^{-1}) in Languedoc-Roussillon and 38.8 (kg ha^{-1} 10 mm^{-1}) in the Loire Valley (Figure 5.1 and Figure 5.2).

Table 5.5 Projected changes in mean temperature (°C) and total precipitation during the grape-growing season in the vineyards of the case study by SSP and period (See also Figures. B.16 and B.18).

Vineyard	**SSP1-2.6**				**SSP5-8.5**			
	2021-2040	**2041-2060**	**2061-2080**	**2081-2100**	**2021-2040**	**2041-2060**	**2061-2080**	**2081-2100**
Temperature change (°C)								
Loire Valley	1.3	1.9	2.2	2.1	1.5	3.1	4.9	6.7
Languedoc-Roussillon	1.4	2.2	2.4	2.3	1.6	3.4	5	7.2
Precipitation change (mm)								
Loire Valley	-16.1	-18.5	-11.8	-3	-7	-11.2	-31.7	-57.5
Languedoc-Roussillon	-9.1	-19.5	-20.5	-14.1	-16.2	-35.5	-27.4	-63.8

Figure 5.3 shows the average projected climate-induced yield impacts (kg/ha) in both vineyards of the case study according to both emission scenarios and the four periods included in the analysis. Under the low emission scenario (SSP1-2.6), the higher reduction in grape yield for Languedoc-Roussillon is projected to approximately -594 kg/ha for 2061-2080, the rise in mean temperature being responsible for 95% of the predicted yield decrease. Likewise, the high emission scenario (SSP5-8.5) projects significant reductions in grape yield in Languedoc-Roussillon, reaching a maximum decrease of -1777 kg/ha by 2081-2100. In contrast, in the Loire Valley, grape yield is expected to increase regardless of the emission scenario because of the positive grape yield sensitivity to mean temperature in this wine-growing region. Under SSP1-2.6, grape yield is projected to increase, ranging from 127 kg/ha for 2021-2040 to 302 kg/ha for 2081-2100. Similarly, the high emission scenario (SSP5-8.5) predicts higher increases in grape yield in Loire Valley, ranging from 203 kg/ha for 2021-2040 to 800 kg/ha for 2081-2100. However, we acknowledge that the rules of Protected Denominations of Origin (PDOs) set the maximum grape yield according to the target quality of wine. Hence, even if our results indicate potential increases in grape yield in Loire Valley, PDOs rules may limit them, where more than 80% of the vineyards are included

in one of the 51 different wine PDOs of the region (InterLoire, 2022). As depicted in Figure 5.3, the projected net climate-induced yield impacts are mainly driven by the projected change in mean temperature; accordingly their direction (increase or decrease) is determined by the direction of grape yield sensitivity to temperature (positive or negative) since mean temperature during the wine grape growing season is projected to increase in both vineyards of the case study (Table 5.5).

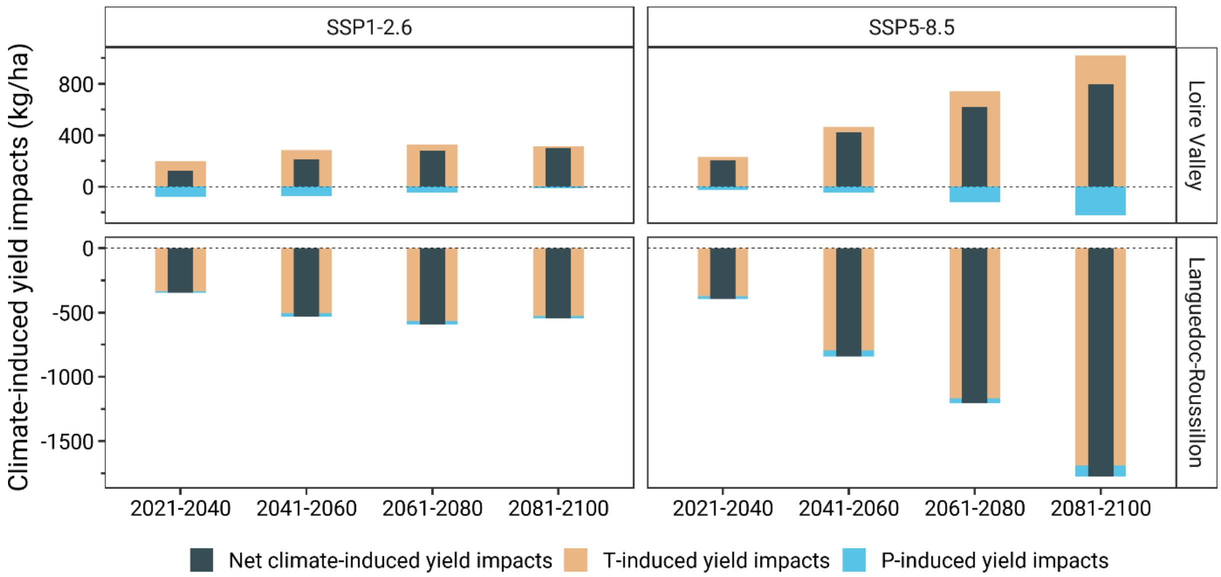


Figure 5.3 Climate-induced yield impacts (kg/ha) in the vineyards of the case study by SSP and period

5.2.4.2 Influence of projected climate-induced yield impacts on LCIA results

In this section, we present the LCIA results obtained at the first level of our analysis, in which a *ceteris paribus* situation was considered, being the projected changes in average temperature and total precipitation the drivers of grape yield variation. Accordingly, the pathways of technical operations (POTs) for future scenarios remained the same as those of the baseline scenario of each vineyard of the case study.

The spatial and temporal variability of the projected climate-induced yield impacts reflected on the life cycle impacts for producing 1 kg of grapes in the two analyzed French vineyards, for which an increase in mean temperature during the grape-growing season led to opposite outcomes in grape yield. Because of the spatial uncertainty resulting from computing zonal statistics of projected climate change, we present the LCIA results, for both midpoint and endpoint impacts, in the form of bullet graphs showing the minimum, median and maximum impact scores calculated according to the same statistics for mean temperature and total precipitation. The LCIA results follow the main diagonal (from the top left corner to the bottom right corner) of the heat map of projected changes in grape yield (Figure B.19).

Figure 5.4 shows a moderate decrease in the carbon footprint for the vineyard from the Loire Valley under the two emission scenarios and during the four periods included in this study. The current carbon footprint of the vineyard from Loire Valley is higher (0.215 kg CO_2eq kg grapes^{-1}) than the median values under the low (SSP1-2.6) and high (SSP5-8.5) emissions scenarios. The highest decrease in the carbon footprint for the vineyard from Loire Valley is projected under SSP5-8.5 for the period 2081-2100 (0.195 kg CO_2eq kg grapes^{-1}), which corresponds to a decrease of around 10% with respect to the baseline scenario. In contrast, the median values of carbon footprint under both emissions scenarios of the vineyard from Languedoc-Roussillon are higher than the current value (0.220 kg CO_2eq kg grapes^{-1}). For the period 2021-2041, the carbon footprint of the vineyard from Languedoc-Roussillon increases by 4.9 and 4.4% with respect to the baseline scenario under the SSP1-2.6 and SSP5-8.5 scenarios, respectively. Under the lower emissions scenario (SSP1-2.6), the higher increase in carbon footprint in Languedoc Roussillon is projected for the period 2061-2080 (0.238 kg CO_2eq kg grapes^{-1}), which corresponds to an increase of 8.2%. The SSP5-8.5 scenario projects higher increases in the carbon footprint of the vineyard from Languedoc-Roussillon, of approximately 18% and 29% for 2061-2080 and 2081-2100, respectively.

Figure 5.4 Carbon footprint (kg CO_2eq·kg grapes^{-1}) of grape production for future periods under two emissions scenarios in two French vineyards. The height of the lighter shaded envelope behind the bars represents the impact per kg of grapes of baseline scenarios. The bullet graph shows the spatial variability arising from the computation of zonal statistics of projected climate change.

Figure 5.5a shows a similar trend for the ecosystem quality impacts resulting from grape production as the carbon footprint shown in Figure 5.4. Nevertheless, there is an important difference in the magnitude of ecosystem quality impacts for the analyzed vineyards. The ecosystem quality impact associated with the baseline scenario of grape production in Languedoc-Roussillon (2.31 PDF·m^2·yr·kg grapes^{-1}) is approximately 6-fold the value computed for the vineyard in the Loire Valley (0.37 PDF·m^2·yr·kg grapes^{-1}). These results are driven by the sharp difference between the number and amount of pesticides applications in each vineyard (Table B.1). Regarding the vineyard from Languedoc-Roussillon, pesticide protection treatments account for around 67% of ecosystem quality impacts, while the contribution of this operation accounts for approximately 22% in the vineyard from the Loire Valley. Regarding future scenarios, under the high emissions scenario (SSP5-8.5) the greatest increase in ecosystem quality impacts for the vineyard from Languedoc-Roussillon is projected for the period 2081-2100, being around 33% greater with respect to the baseline scenario. Conversely, under the same emission scenario (SSP5-8.5), the highest reduction in the ecosystem quality impacts for the vineyard from the Loire Valley is projected for 2081-2100, which is around 9% with respect to the baseline scenario.

Similar trends were found for the human health impacts (Figure 5.5b) to those shown in Figure 5.4 for the carbon footprint and in Figure 5.5a for ecosystem quality impacts. However, to put our LCIA results derived from climate-induced yield impact into perspective, an LCA of organic viticulture in France showed that the influence of interannual variability in the environmental profile of grape production, can reach up to 52% per impact category because of changes in management practices according to different climatic conditions and pests and disease pressure (Christel Renaud-Gentié et al., 2020), compared to around 33% of maximal variability shown here. However organic PTOs were mentioned being more subject to interannual variations by Beauchet et al. (2019), especially due to treatment frequency, than conventional PTOs because of the wash off of the pesticides used in organic viticulture by the rain.

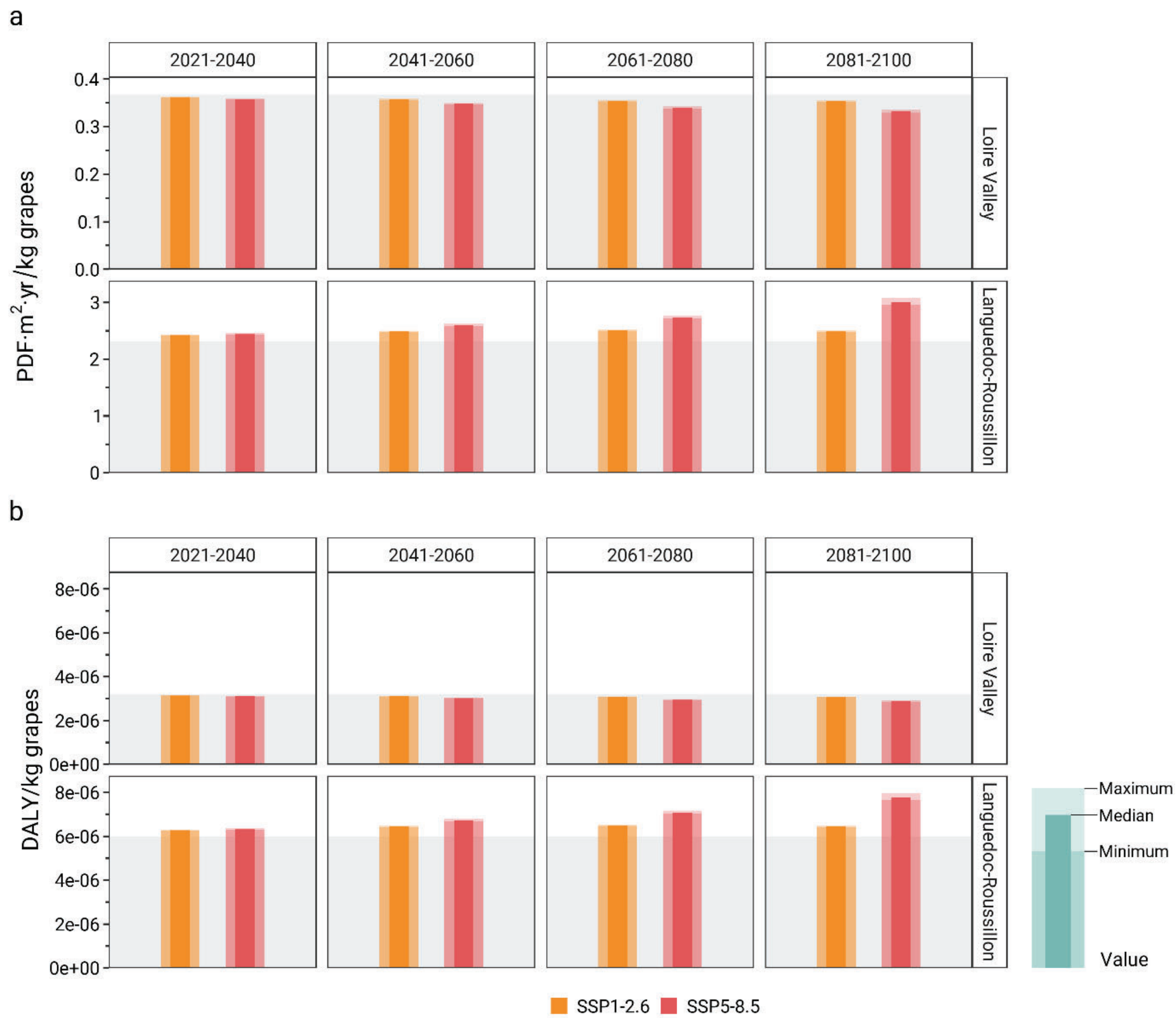


Figure 5.5 Ecosystem quality (EQ) impacts (PDF·m^2·yr·kg grapes^{-1}) (a) and human health (HH) impacts (DALY·kg grapes^{-1}) (b) of grape production for future periods under two emissions scenarios in two French vineyards. The height of the lighter shaded envelope behind the bars represents the impact per kg of grapes of baseline scenarios. The bullet graph shows the spatial variability arising from the computation of zonal statistics of projected climate change. Please note that in the panel (a) the y-axis is not at the same scale

With regard to ecosystem quality and human health potential impacts resulting from pesticide emissions, we observe a large variation in the number and amount of pesticides applied in each analyzed vineyard, and the number of active ingredients varies in the same way. Overall, 17 active ingredients are applied in the vineyard from Languedoc-Roussillon (Table B.7), while 9 different active ingredients are applied in Loire Valley (Table B.8). For the vineyard from Languedoc-

Roussillon, 7 out of 17 active ingredients were not characterized due to the lack of characterization factors, whereas for the vineyard from Loire Valley, 4 out of 9 active ingredients were not characterized. We acknowledge that the potential environmental impact of pesticides emissions is underestimated due to the lack of characterization factors for some active ingredients, still the vineyard from Languedoc-Roussillon showed larger impacts related to pesticides emissions due to the high application rate. Besides, the fact that no impact assessment method covers characterization factors for all pesticides is a current challenge when modelling agricultural systems in LCA (Nemecek et al., 2022).

5.2.4.3 Influence of extreme events and adaptation strategies on LCIA results

Figure 5.6 reports the potential yield loss rate due to extreme events (Equation 5.3) for grape production under future climate change scenarios without the implementation of adaptation levers. For both vineyards, regardless of the climate change scenario and period, spring frost is expected to produce the lowest yield loss rate. In fact, the greatest impact of spring frost amounts to 1.1% in Loire Valley by the end of the century. However, the assessment of climate change impact on spring frost risk has led to a contentious debate in the scientific literature. Spring frost arises when budburst precedes the occurrence of the last frost event in the spring of a grape-growing season. According to the projected impacts of climate change on viticulture, these events will occur earlier (Santos et al., 2020), leading to lessen spring frost risk in the future (Molitor et al., 2014; Santos et al., 2020), while other studies found conflicting results among models or projected increased risks for spring frost damage in viticulture (Leolini et al., 2018; Mosedale et al., 2015).

With respect to yield loss rates due to hail (Figure 5.6), the values for both vineyards are under 3% for the near- and mid-term future, whereas the highest impacts were found for both vineyards under the high emissions scenario (SSP5-8.5), which correspond to 4.7 and 5% for Loire Valley and Languedoc-Roussillon, respectively. Because hailstorms are normally site-specific events, there are still uncertainties on predicting the potential impact of hail on agriculture (Petoumenou et al., 2019). However, it is likely that hailstorms will increase under climate change because of the rise of low-level moisture and convective instability driven by anthropogenic warming (Raupach et al., 2021). The low potential yield loss rates due to frost and hail are associated with their low probability; consequently, they represent the average impact of a given period and not a precise point in time (Tables S9, S10, and S10).

Regarding the potential yield loss rate due to drought, the same level of damage was computed for both vineyards under the low emissions scenario (SSP1-2.6), which is around 20% (Figure 5.6). On the other hand, the impact of drought under the high emissions scenario (SSP5-8.5) is expected to increase steadily, and by the end of the century the potential yield loss rate is estimated at 28.9 and 29.4% for Loire Valley and Languedoc-Roussillon, respectively. Figure 5.6 shows that the highest potential yield loss rates due to heatwaves are projected under the high emissions scenario (SSP5-8.5) for both vineyards. In fact, under the latter scenario, in the case of Loire Valley, the potential yield loss rate doubles from the near-future period to the subsequent periods, while it is always at the highest level in Languedoc-Roussillon (35%) for future periods. Nonetheless, these estimates are potentially conservative since they do not account for the occurrence of multiple heatwaves. For example, under the high emissions scenario (SSP5-8.5), six and seven heatwaves (spell of nine consecutive days) are projected for Loire Valley and Languedoc-Roussillon by the end of the century, respectively (Table B.15). In fact, Fraga et al. (2020) have stressed the need to account for multiple heatwaves when modelling the impact of climate change on European viticulture.

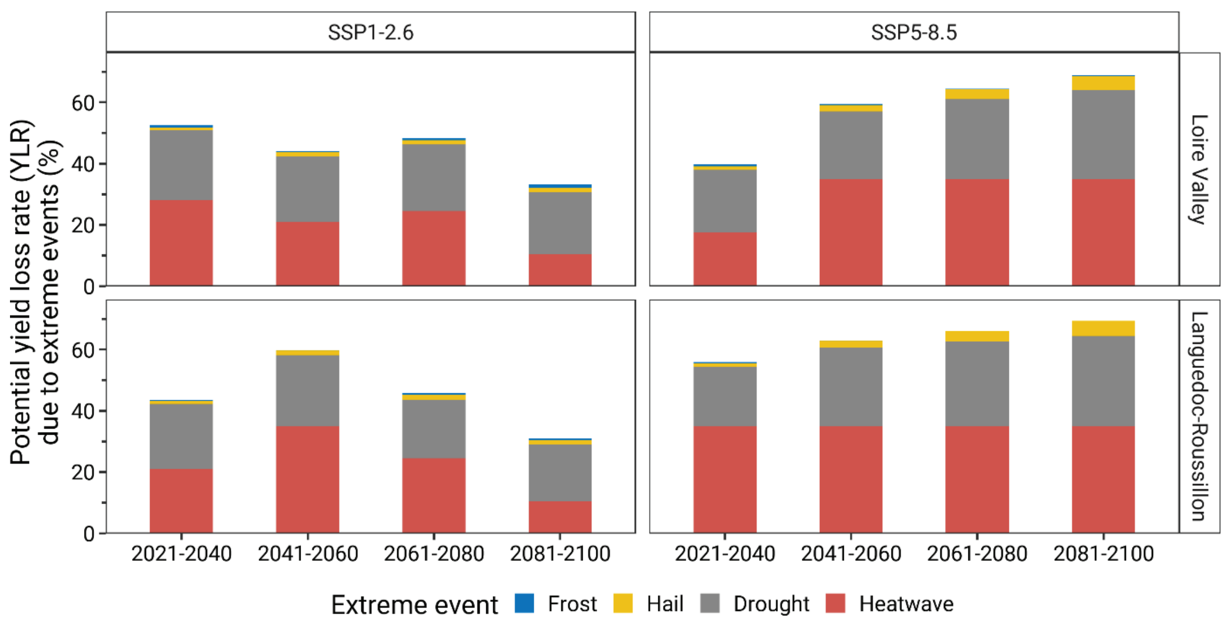


Figure 5.6 Potential yield loss rate (YLR - %) due to extreme events on future grape yields for the vineyards of the case study

While accounting only for the projected climate-induced yields resulted in opposite conclusions about the LCIA results of the vineyard of the case study: a potential decrease of LCIA results in

Loire Valley and an increase in Languedoc-Roussillon under future scenarios (Figure 5.4 and Figure 5.5), introducing the impact of extreme events translated into an increase in the life cycle impacts associated with grape production for both vineyards compared to baseline scenarios.

To communicate the impact of extreme events on the LCIA results for both vineyards, the corresponding figures show the LCIA of the baseline scenarios, the LCIA when no adaptation lever is implemented (grape yield calculated according to the first case of Equation 5.4), and the LCIA results when a combination of adaptation options are activated (grape yield calculated according to the second case of Equation 5.4 and inclusion of LCIA impacts of adaptation options according to Figure B20). We recall that a combination of adaptation levers for each vineyard may include, besides increasing the number of phytosanitary treatments, the inclusion of irrigation, the installation of anti-hail nets, and oak wood-burning against spring frost according to the expected impact of extreme events for each period and SSP (if their expected yield loss rate is higher than 1%). Figure B.20 shows the combination of implemented adaptation strategies in each vineyard for a given period and SSP. Since our study does not account for adaptation options to mitigate the expected yield loss due to heatwaves, both scenarios of “activated” and “non activated” adaptations account for the yield loss rate due to heatwaves (Equation 5.4).

Figure 5.7 shows a sharp increase in carbon footprint for both vineyards due to the impact of extreme events on grape yield. Besides, for this impact category, the implementation of adaptation levers in both vineyards entails lower scores (26% on average) with respect to a situation in which adaptation strategies are not implemented. That is, the avoidance of yield loss by activating adaptation strategies, in most situations, compensates for their related carbon footprint. In fact, other studies have stressed the key influence of yield on the environmental score per kilogram of grapes in both interannual (Christel Renaud-Gentié et al., 2020) and interregional dimensions (Vázquez-Rowe, Villanueva-Rey, Iribarren, et al., 2012).

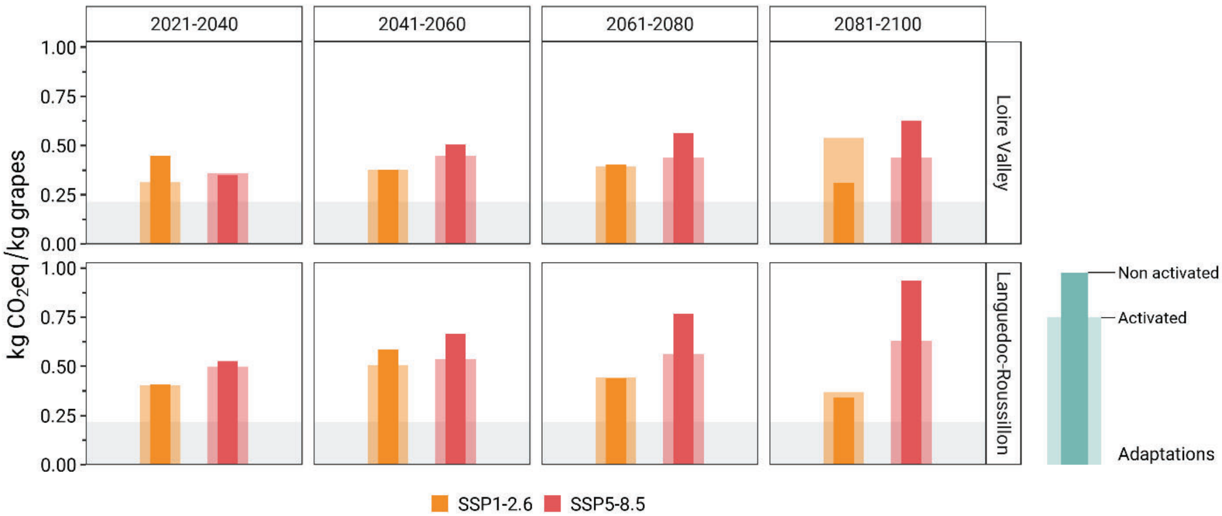


Figure 5.7 Carbon footprint (kg CO_2eq·kg grapes^{-1}) of grape production for future periods under two emissions scenarios in two French vineyards. The height of the lighter shaded envelope behind the bars represents the impact per kg of grapes of baseline scenarios. The lighter bars indicate the impact score when adaptation levers are activated, while darker bars indicate that adaptation levers are not activated.

The highest increase in carbon footprint for the vineyard from Loire Valley is projected under the high emissions scenario (SSP5-8.5) by the end of the century, when adaptation levers are not implemented (0.628 kg CO_2eq kg grapes^{-1}), which corresponds to an increase of approximately 191% with respect to the baseline scenario. In contrast, the lowest increase in carbon footprint for Loire Valley is projected under SSP1-2.6 by 2081-2100 when no adaptation strategies are implemented, corresponding to an increase of 44% with respect to the baseline scenario. The reason for the latter trend reversal is that the installation of the anti-hail net system and the anti-frost lever increase the related carbon footprint by 73% with respect to a situation of inaction (Figure B.20, Figure 5.7, see Figure B.21 for the contribution of adaptation levers), whereas the avoidance of the associated yield loss is 2.5%. Because of the uncertainty associated with spring frost damage and the high carbon footprint of wood-burning (0.21 kg CO_2eq kg grapes^{-1}, Figure B.21), future research may address other adaptation strategies for these events, such as the use of fans, heaters, and changes in the canopy. Regarding the vineyard from Languedoc-Roussillon, the increase in carbon footprint is even more exacerbated under SSP5-8.5 by the end of the century, with an increase of around 326% when adaptation strategies are not implemented (0.935 kg CO_2eq kg grapes^{-1}), and of 187% with adaptation levers (0.630 kg grapes^{-1}) with respect to the baseline scenario (Figure 5.7). Under SSP5-8.5, for the vineyard of Languedoc-Roussillon, the related carbon footprint of implementing adaptation strategies is counterbalanced by the prevention of yield loss that these adaptation levers would entail. However, this conclusion is reversed under SSP1-2.6 by 2081-2100 since the expected yield loss rates due to extreme events are lower with respect to other periods (Figure 5.6), and particularly that related to heatwaves damage (Table B.16).

Figure 5.8a shows an opposite trend for the ecosystem quality impacts of grape production in the vineyard from Loire Valley with respect to the carbon footprint shown in Figure 5.7, as including adaptation levers leads to higher impacts, 190% greater on average, with respect to a situation of inaction. The latter trend is explained by the high contribution of the anti-hail system and irrigation to ecosystem quality impacts (Figure B.22a). In fact, under the SSP1-2.6, irrigation increases the ecosystem quality impacts by 75% on average, whereas the anti-hail system leads to an increase of around 170% with respect to not adopting adaptation levers, while the potential loss rate due to extreme weather is at most 53% (for the period 2021-2040). Consequently, the related ecosystem quality impacts of implementing adaptation levers are not offset by the avoidance of yield loss due to extreme events. However, in the case of the vineyard from Languedoc-Roussillon under the high emissions scenario SSP5-8.5, the activation of adaptation levers, starting from the period 2041-2060 to the end of the century, leads to lower ecosystem quality impacts with respect to a situation of inaction. The reason of this decrease in ecosystem quality impacts is that including irrigation increases these impacts by 15% on average, and the anti-hail system raises the impacts by 20% with respect to not activating adaptation strategies, whereas the potential loss rate due to extreme events is 64% on average. Therefore, the associated ecosystem quality impacts of the adaptation strategies are outweighed by the avoided yield loss due to extreme events. In fact, for this vineyard, the increase in ecosystem quality impacts by the end of the century with activated adaptation levers is 255%, whereas the increase is 327% when no adaptation levers are applied, with respect to the baseline scenario.

When considering the human health impacts of grape production (Figure 5.8b), the conclusion on the influence of activating adaptation levers to mitigate extreme weather events is similar to the trends of the carbon footprint (Figure 5.7), and in some cases for ecosystem quality (Figure 5.8). The reason for these trends is that human health impacts increase by approximately 7% when adopting adaptation levers in Languedoc-Roussillon (Figure B.22b), avoiding a potential yield loss of around 25%. Likewise, in the case of the vineyard from Loire Valley, the adoption of adaptation options increases the impact by approximately 13%, preventing a potential yield loss of around 25%. However, the impact increases by 34% when including the anti-frost systems, besides the anti-hail system, and irrigation (Figure B.22b). Under these situations, the adoption of adaptation levers counterbalances the influence of yield loss due to extreme events on the environmental profile of grape production. For the vineyard from Languedoc-Roussillon, the human health impacts are approximately 31% lower when adaptation options are applied than in a situation of inaction. Regarding the vineyard from Loire Valley, the same trend is observed, with a decrease of around 27%. In general, the increase of human health impacts accounting for extreme events and adaptation options are lower with respect to carbon footprint and ecosystem quality impacts. For instance, for the vineyard of Languedoc-Roussillon, the higher expected increase is 116% under SSP5-8.5 by 2081-2100, whereas the highest increase in these impacts for Loire Valley is 69% by the period 2041-2060.

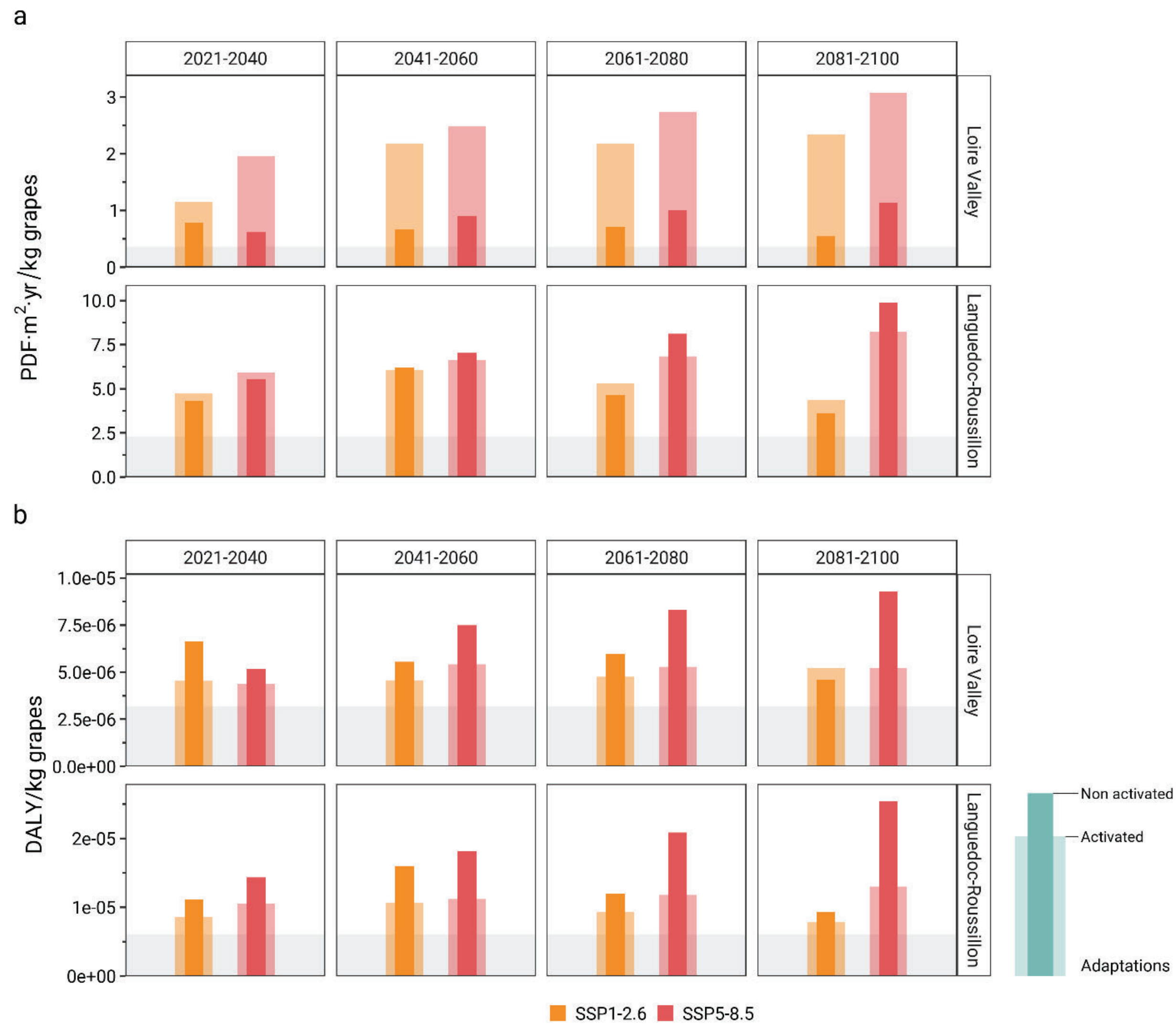


Figure 5.8 Ecosystem quality (EQ) impacts (PDF·m^2·yr·kg grapes^{-1}) (a) and human health (HH) impacts (DALY·kg grapes^{-1}) (b) of grape production for future periods under two emissions scenarios in two French vineyards. The height of the lighter shaded envelope behind the bars represents the impact per kg of grapes of baseline scenarios. The lighter bars indicate the impact score when adaptation levers are activated, while darker bars indicate that adaptation levers are not activated. Please note that in this figure the y-axis is not at the same scale.

Given the exacerbated LCIA results of grape production for the vineyards of the case study when accounting for the impact of both changes in climate variables and extreme events, other adaptation strategies may be required to improve the environmental performance under projected tough climate conditions, and to guarantee grape yield productivity. In this study, we did not consider introducing grape varieties tolerant to heat and water stress, which is also an alternative to cope with the effect of extreme events, without considering the constraints imposed by current regulations on the allowed varieties in a given region (Wolkovich et al., 2018). Furthermore, since the analyzed vineyards are examples within the range of technical options implemented in each winegrowing region, future research may address the variability of LCIA results according to the diversity of practices in each territory.

The projected irrigation demands for the vineyards of the case study are relatively low (Table B.19), the highest value is 896 m^3/ha under SSP5-8.5 by the end of the century, with respect to more water-demanding vineyards where water consumption can reach more than 2600 m^3/ha (Russo et al., 2021). However, regions facing water scarcity, which is the case of the Mediterranean region, should advocate for improved water use efficiency (Romero et al., 2022), or to implement low-density systems to cope with projected water scarcity (Naulleau et al., 2021). Although outside the scope of this study, another aspect to consider when envisaging irrigation as an adaptation lever is its impact on wine composition. In the case of high-quality wine, most winemakers are opposed to irrigation, intending to preserve wine typicity (H Fraga et al., 2018). While the functional unit of the analyzed product systems is expressed in kilogram of grapes, future research may address the influence of grape quality as a driver of environmental performance.

In this study we considered that the impacts of extreme weather events on grape yield are additive, however, the synergistic effect of projected extreme water and heat stresses may produce lower yields (H Fraga et al., 2018). Consequently, the computed LCIA results potentially underestimated the interaction of drought and heatwaves events. Furthermore, this study does not account for changes in vine training systems, which are able to modify the vine microclimate. Still, there is no consensus on the optimal choice of training system to cope with drought (Naulleau et al., 2021).

5.2.4.4 Model and data uncertainty and future perspectives

One key objective of models is to help understand the future and to support planning or adaptation. This is particularly important for agriculture, which faces the challenge to mitigate and adapt to the impacts of projected climate change. For this reason, combining the use of future scenarios and LCA is a promising tool to anticipate the impacts and act before the environment is damaged. Still, modelling future scenarios is intrinsically uncertain, without considering the practical challenges associated with their definition. Consequently, in this study, we aimed at projecting plausible future life-cycle environmental impacts of grape production according to the range established by a low and a high GHG emissions scenario and four future periods. While the main goal of this study was to contribute to the scientific discussion on the impacts of climate change on the environmental performance of future viticulture, future research could include participatory approaches to integrate local knowledge, as well as social and economic aspects in the analysis (Naulleau et al., 2021; Rouault et al., 2020).

Several approaches have been used to evaluate the impact of change in temperature and precipitation on grape yields. We used statistical modelling based on reported grape yields (Agreste, 2021) and climate records (Mavromatis & Voulanas, 2021) to fit linear regressions and to subsequently project grape yield responses. However, we acknowledge some limitations to the application of linear regression to predict future grape yields. Besides precipitation and temperature, there are other aspects that can influence grape yield such as solar radiation, extreme precipitation and temperature, canopy management, training systems, as well as extreme weather events (though, these were included in the second part of the study). Furthermore, this study does not account for the effects of rising atmospheric CO_2 concentration that potentially stimulates grape yields when nutrients are not limited (Helder Fraga et al., 2016). Still, more multiple-year tests are required to conclude on the effects of CO_2 fertilization on perennial crops (Biavetti et al., 2014). Moreover, the projected changes in grape yields of our study are likely to be conservative since our study does not consider nonlinear responses of grape yield to temperature, as has been shown for major crops (wheat, rice, maize, and soybean) (Zhao et al., 2017). In addition, this study does not address covariability of climate variables, which has been found to play a key role in computing accurate crop-yield sensitivity to climate factors (Leng et al., 2016). Nevertheless, our study is consistent with other studies that have analyzed yield sensitivity to climate variables independently (Kukal & Irmak, 2018; Zhao et al., 2017).

We acknowledge some limitations resulting from the quality and availability of input data. Despite the high spatial resolution of the data set of grape yield (Agreste, 2021), the temporal resolution is limited to the period 2000-2020. Even so, the 21-year period for computing grape yield sensitivity to climate of this study is broader with respect to the 9-year period considered in a novel study implementing machine learning techniques for projecting life-cycle environmental impacts of corn production in the U.S. Midwest (E. K. Lee et al., 2020). Furthermore, the aggregation of data hindered the possibility to account for the variability of yield sensitivity to climate according to grape variety and agricultural system (conventional, organic). However, this limitation is shared with other studies that have considered a standard grape variety for modelling the impacts of climate change on grape yield at the watershed scale (Naulleau et al., 2022) and at European scale (Helder Fraga et al., 2016).

As climate change brings more frequent and longer heat and drought extremes, there is an increasing concern that these events lead to a decrease in grape yield and quality under future regimes. Nonetheless, it is challenging to quantify the impact of extreme events on agricultural systems, given that it is difficult to adequately set up experiments and calibrate models to estimate the impacts of these events. For instance, the authors of a study assessing the impact of climate change on grape yield at the European scale highlighted the need that future research addresses the influence of extreme weather events on grapevine yield and quality, since it may offer a better understanding of climate change impacts on viticulture (Helder Fraga et al., 2016). In this study, we took a risk approach to account for the impact of extreme events on grape yield, calculated according to the product of their probability occurrence and level of damage. We assumed that farmers have full capacity to respond to these events and that they will make the effort to keep grape yield close to current levels. However, future research could incorporate an economic model to account for different levels of risk aversion among winegrowers (Sacchelli et al., 2017). Even though this study does not account for the interactions of extreme climate change risks, it aimed at providing projections of the impacts of these events on the environmental profile of viticulture under future conditions. Besides, climate change risk assessment is an evolving field that deserves special attention to better account for interactions among several drivers of climate change risks and how these may lead to cascade effects (Simpson et al., 2021).

In this study, we took an attributional approach to estimate future environmental impacts of grape production, which is consistent with studies that have analyzed small-scale systems (Bisinella et al., 2021). As such, we implicitly extrapolate current system processes to model the background and foreground of viticultural systems under future conditions. As a result, this study does not account for potential changes in the types of pesticides or other agricultural inputs in the future. Hence, this aspect remains part of the “known unknowns” related to climate change, and broadly to future scenario modelling. However, future studies may consider a consequential approach if the aim is to support regulations at a larger scale. For instance, while irrigation is a technical option to mitigate grape yield losses due to drought in the Mediterranean basin, it is expected that the future water demand will not be met for all future vineyards (van Leeuwen et al., 2019). Furthermore, if the aim is to keep future grape production at present levels, this would lead to a snowball effect because of the projected rise of the carbon footprint of grape production, which consequently would lead to higher impacts of climate change on this agricultural system. Other potential impacts of keeping current grape annual production under future less favourable environmental conditions would be competition for land and an increased impact of agriculture due to this feedback loop, which merits further research according to a consequential approach.

Our study addressed the impacts of climate change on future grape production in two French vineyards at the life-cycle inventory level. Nevertheless, projected environmental conditions under climate change, such as increased mean and extreme temperatures, prolonged periods of wetter and drier conditions, changes in soil characteristics, and modified salinity dynamics in estuaries will also alter the fate and effect of pollutants in the environment (Noyes & Lema, 2015). This aspect has not been considered in this paper but merits equal consideration. Therefore, when pertinent, LCIA developers may work on the computation of future characterization factors accounting for projected environmental conditions in the characterization modelling. Some studies have already addressed the development of characterization factors under future scenarios. For instance, Núñez et al. (2015) derived characterization factors for evaluating water use-related impacts of future technologies projected in Spain for a mid-term future scenario of 2030, while Cosme and Niero (2017) computed future characterization factors for marine eutrophication under a high GHG emissions scenario by introducing the impact of future climate-driven changes in the fate, exposure and effect factors.

5.2.5 Conclusions

Feasibility of implementing a prospective LCA was demonstrated by using the principle of parsimony (application of relatively simple models with accessible data sets) on two winegrowing case studies. As is often the case in LCA, the lessons learned can be partially counter-intuitive, which is why the approach is so interesting in terms of decision support.

The findings of this exploratory research show that projected changes in mean temperature and total precipitation during the wine grape growing season play a key role in influencing grape yield, and consequently in the life cycle impacts of the two analyzed French vineyards. Furthermore, it was shown that the projected increase of extreme weather events, particularly heatwaves and drought, will likely lead to heightened environmental impacts per kilogram of grapes. Finally, including projected climate change when defining agricultural systems appears to be a key aspect to increase the relevance of LCA for supporting decision-making in prospective analysis.

CHAPITRE 6 ARTICLE 3 : MODELLING THE INFLUENCE OF CLIMATE CHANGE ON CHARACTERIZATION FACTORS FOR COPPER TERRESTRIAL ECOTOXICITY

6.1 Présentation de l’article

Dans ce troisième article, des facteurs de caractérisation futurs pour l’écotoxicité terrestre du cuivre incluant l’influence du changement climatique ont été calculés pour les sols non calcaires des vignobles d’Europe selon trois scénarios prospectifs à l’horizon 2050, issus de trois scénarios RCP.

Les auteurs de cet article sont Ivan Viveros Santos, Annie Levasseur, Cécile Bulle, Louise Deschênes et Anne-Marie Boulay. Il a été soumis à *Journal of Cleaner Production* le 19 juin 2022. Le matériel supplémentaire soumis avec l’article est disponible dans l’annexe C.

Les cartes des facteurs de caractérisation en format GIS sont disponibles à cette adresse :

https://github.com/iviveros/viveros-santos_et_al_2022_jclp/tree/main/cf_spatial_res

Les cartes interactives des facteurs de caractérisation accompagnant l’article sont disponibles à cette adresse :

https://iviveros.github.io/viveros-santos_et_al_2022_jclp/

6.2 Manuscrit

6.2.1 Abstract

The use of copper-based preparations is a long-standing practice in viticulture to control vine fungal diseases, which has led to high copper concentrations in vineyard soils and impacts on off-target terrestrial organisms. Under projected climate change, some of the mechanisms and properties of soils that influence the extent of metals ecotoxicity impacts are projected to be altered, namely soil erosion, rainfall, temperature, and organic carbon content. In this context, and in the framework of life cycle assessment, this study aims to simulate the influence of projected changes in soil organic carbon, soil erosion, and rainfall on characterization factors (CFs) for copper terrestrial ecotoxicity across non-calcareous European vineyard soils. We employed empirical regression models to account for metal speciation and bioavailability as determined by soil characteristics. CFs were computed for a current scenario and mid-term future scenarios by 2050 determined across three Representative Concentration Pathways (RCP2.6, RCP4.5, and RCP8.5). Although future scenarios suggest that CFs may either increase or decrease, CFs are projected to increase for a larger share of European vineyard surface, which may lead to higher terrestrial ecotoxicity impacts. The RCP4.5 scenario projects the highest increases in CFs, with a 27% rise in the median CFs in comparison with the current scenario. Whereas the RCP8.5 and RCP2.6 scenarios project a 19% and 14% increase in median CFs, respectively. The changes in CFs were determined principally by the changes in copper bioavailability driven by projected changes in soil organic carbon. However, the spatial variability of CFs was larger than the temporal variation, with a variability of roughly 2 orders of magnitude across the analyzed scenarios. This study highlights the relevance of integrating the spatial differentiation and the influence of projected climate change in the characterization modelling of copper terrestrial ecotoxicity.

Keywords: Life cycle impact assessment, terrestrial ecotoxicity, speciation, climate change, regionalization, interactions.

Highlights:

- Method accounting for the interaction of climate change and terrestrial ecotoxicity.
- Neglecting the interaction of impact categories potentially underestimates terrestrial ecotoxicity impacts.
- Changes in future soil organic matter content drive changes in CFs values.

6.2.2 Introduction

Copper-based fungicides are widely used in organic and conventional viticulture to control fungal diseases, namely downy mildew (*Plasmopara viticola*) (Droz et al., 2021). The use of copper-based fungicides is a long-standing practice in viticulture, initiated in the late 19th century, because of the high effectiveness of copper under rainy conditions and its relatively low market cost (Tamm et al., 2022). The application rate and timing of copper-based fungicides are closely related to hydroclimatic conditions, agricultural practices, vine variety, and policies (Droz et al., 2021). An average fungicide application rate of 8.1 $kg \cdot ha^{-1}$ has been estimated by Panagos et al., (2018) for permanent crops (among which viticulture), which is higher than that for arable land (0.54 $kg \cdot ha^{-1}$) (Panagos et al., 2018) and greater than the highest application rate established by the European

Union regulations (4 kg·ha^{-1}) (Droz et al., 2021). The continuous application of copper-based fungicides has led to high copper content in vineyard soils in comparison with other land uses (Ballabio et al., 2018; Droz et al., 2021), leading to impacts on terrestrial and aquatic organisms (Bart et al., 2017; Fernández et al., 2015; Ruyters et al., 2013).

Life Cycle Assessment (LCA) is a holistic method to evaluate the potential environmental impacts related to a product or service across its entire life cycle from resource acquisition to its end of life (ISO, 2006a, 2006b). In the context of LCA, the impacts on terrestrial and aquatic organisms resulting from the emissions of chemicals are evaluated under the ecotoxicity impact category (P Fantke et al., 2017; Peter Fantke et al., 2018). LCA studies on wine production have reported copper as the dominant contributor to the ecotoxicity impacts due to the use of copper-based fungicides in viticulture (Falcone et al., 2016; Vázquez-Rowe, Villanueva-Rey, Moreira, et al., 2012). Nevertheless, the high contribution of metals to ecotoxicity impacts is not restricted to viticulture. A study addressing the question as to whether the inclusion of speciation (i.e., the different pools of metal in soil) changes the rank contribution of metals to terrestrial ecotoxicity assessed roughly 13000 unit processes of the ecoinvent database employing three life cycle impact assessment methods. The authors concluded that even when metal speciation is integrated into the characterization of terrestrial ecotoxicity, metals remain the main contributors to this impact category and that the difference in results among different impact methods is explained by variations in the coverage of chemicals (Sydow et al., 2020).

Recent efforts in LCA were devoted to enhancing the assessment of metals ecotoxicity-related impacts by including metal speciation in the computation of characterization factors (CFs) (Dong et al., 2014; Gandhi et al., 2010; Owsianiak et al., 2013; Plouffe et al., 2016). Within LCA, CFs are applied to convert and aggregate life cycle inventory interventions (i.e., emissions or resource consumption) into scores of potential impacts. Regarding terrestrial ecotoxicity, two main methodological frameworks have been developed to derive CFs including metal speciation driven by soil properties. One method employed empirical regression models to compute the different pools of metal in soil (Owsianiak et al., 2013), whereas a second approach used the geochemical speciation model WHAM 6.0 for the same purpose (Plouffe et al., 2016). Both methodological approaches break down the CF into a fate factor (FF) describing the fate of the substance in the environment, a bioavailability factor (BF) corresponding to the fraction of the total metal that is available for uptake by organisms, and an effect factor (EF) representing the potentially affected fraction of species (PAF) by the available metal pool (Owsianiak et al., 2013; Plouffe et al., 2016). Owsianiak et al., (2013) also introduced an accessibility factor (ACF) in the computation of CFs, which represents the fraction of reactive metal over total metal. In consequence, the BF is expressed by the fraction of free ions of the reactive metal in soil. Moreover, Owsianiak et al., (2013) derived a multiple linear regression (MLR) for the calculation of CFs for copper, with soil organic carbon being the controlling factor due to its influence on metal fate. Besides, the proposed MLR was shown to be improved by integrating soil pH, which influences metal bioavailability. In addition, it has been reported that the key factors explaining 45% of the copper content in European vineyards are precipitation, aridity, and soil organic carbon (Droz et al., 2021).

In the context of copper-based fungicides use in viticulture, subsequent studies derived CFs for copper terrestrial ecotoxicity including metal speciation (Peña et al., 2018; P Villanueva-Rey et al., 2019; Ivan Viveros Santos et al., 2018). Peña et al. (2018) and Viveros Santos et al. (2018) computed site-dependent CFs for non-calcareous soils, that is, CFs at a relatively low spatial resolution inherited from the Harmonized World Soil Database, which is a 30 arc-second raster database. However, both studies aimed at computing CFs for large geographical areas. Peña et al. (2018) concentrated on European vineyards and found a spatial variability of CFs over 1.5 orders of magnitude. Viveros Santos et al. (2018) focused on non-calcareous soils of the world and reported a spatial variability of CFs derived with WHAM 6.0 of 5.5 orders of magnitude. On the other hand, Villanueva-Rey et al. (2019) calculated spatially differentiated CFs for wine-growing regions in Northern Spain and Portugal and found a spatial variability of 1.6 orders of magnitude between the lowest and highest value of CFs.

There is a growing interest in assessing future-oriented scenarios employing LCA to foresee the potential environmental impacts of new technologies and products (Bisinella et al., 2021; Sacchi et al., 2022), or to evaluate them under projected environmental conditions altered by climate change (Sala et al., 2017). Particularly, since agriculture is highly dependent on climate, some LCA studies have addressed the potential impact of projected climate change on the environmental performance of some crops, namely corn, soybean, spring barley, and wine grapes (Cosme & Niero, 2017; Garba et al., 2014; E. K. Lee et al., 2020; Ivan Viveros Santos et al., 2022). These studies simulated the impact of climate change at the life cycle inventory level, considering the variation of crop yield due to climate and extreme events, as well as changes in agricultural practices such as the application rate of pesticides and fertilizers. However, it is expected that the fate, exposure, and effect of pollutants will be altered under future environmental conditions (Noyes & Lema, 2015; Stahl Jr. et al., 2013). In this regard, research has been conducted to develop CFs under future scenarios. For instance, CFs for assessing water use-related impacts were developed to integrate the impact of climate change on water availability in the near- and mid-term future in Spain. It was found that future decreases in water withdrawals compared to the current situation would lead to lower impacts related to freshwater resources accessibility (Núñez et al., 2015). Moreover, CFs for marine eutrophication were parameterized to simulate the influence of projected environmental conditions on the fate, exposure, and effect of nitrogen emissions. While the effect factors are predicted to increase by around 7% for marine eutrophication, the decreases in fate and exposition factors will result in a decrease in CFs of around 22% by 2050 for the North Sea and Baltic Sea (Cosme & Niero, 2017).

The terrestrial ecotoxicity of metals is site-dependent owing to the influence of soil properties and climate conditions on metal speciation (Owsianiak et al., 2013; Plouffe et al., 2016). Besides, some soil properties and mechanisms that affect the ecotoxicity impact of metals such as organic matter, moisture, rainfall, microbial activity, and soil erosion are susceptible to climate change (Biswas et al., 2018; Fu et al., 2018; Noyes & Lema, 2015; Pham et al., 2021). Projected soil erosion rates indicate a potential increase of 13% to 22.5% by 2050 compared to the current situation in Europe (Panagos et al., 2021), which might increase the runoff of contaminants (Biswas et al., 2018), and of particular concern in sloping vineyards (Pham et al., 2021). Moreover, projected changes in the frequency and intensity of rainfall will also impact the runoff and leaching of pollutants (Biswas et al., 2018). In addition, the projected changes in soil organic carbon (SOC) may modify the mobility of copper in vineyard soils, which will vary spatially given that projections indicate increases in SOC stocks in most parts of Europe by 2050, but also point to decreases principally in southern Europe (Droz et al., 2021; Yigini & Panagos, 2016). Some studies have also reported adverse effects of increasing air temperature and drier conditions on the performance of soil invertebrates in metal-polluted soils. Still, the authors stressed the need to conduct more research to disentangle the effect of changing environmental conditions on species sensitivity from induced changes in metal speciation (Fu et al., 2018; González-Alcaraz & van Gestel, 2016).

Given that some soil properties and mechanisms affecting the fate, mobility, and bioavailability of metals in soils are projected to change under future environmental conditions, as derived from climate change scenarios, the purpose of this exploratory study is to simulate the influence of expected changes in soil organic carbon, soil erosion rates, and precipitation on future CFs for copper terrestrial ecotoxicity. Sets of CFs were calculated for baseline and mid-term future scenarios by 2050. The latter scenarios were based on projections of the above-mentioned parameters derived according to the Global Climate Model (GCM) IPSL-CM5A-LR across three Representative Concentration Pathways (RCP2.6, RCP4.5, and RCP8.5). This study addresses both the spatial and temporal dimensions of the characterization modelling of copper terrestrial ecotoxicity. Hence, on the one side, this study contributes to the ongoing efforts in LCA to account for spatially differentiated impacts. On the other side, it provides CFs for studies aimed at assessing the environmental impacts of product systems under future scenarios, which is particularly relevant to agricultural systems. Furthermore, this study proposes an approach to account for the interaction of impact categories, which are normally characterized independently in the framework of LCA.

6.2.3 Methods

6.2.3.1 Study area and temporal scenarios

The study area corresponds to the European vineyard surface (Figure C1b), representing approximately 26% of land dedicated to permanent crops and approximately 1.7% of the total agricultural land of the European Union. The European vineyard area is defined by a layer retrieved from the CORINE Land Cover project of the European Environment Agency (2017b) (Figure C1b). Despite the low share of agricultural land under vineyards, a large amount of copper-based fungicides sold in the EU are used extensively in permanent crops (vineyards, olive groves, and fruit trees). Furthermore, in the EU, vineyard soils have the highest median copper concentration (26.09 $mg \cdot kg^{-1}$) in comparison to other land uses (11.58 $mg \cdot kg^{-1}$) (Figure C1) (Ballabio et al., 2018).

To assess the effect of projected changes in soil organic carbon, soil erosion rates, and precipitation on the characterization modelling of copper terrestrial ecotoxicity, CFs were computed for two temporal scenarios:

- Baseline scenario: A scenario based on historical or recent datasets of climate and soil properties to determine CFs for the current state of the environment. This is a common practice in environmental modelling, namely in LCA (P. Fantke et al., 2018; Kounina et al., 2014) and in environmental risk assessment (Stahl Jr. et al., 2013) to evaluate the potential environmental impacts of emissions.

- Midterm future scenario: A prospective scenario by 2050, according to data availability on projections of soil organic carbon, soil erosion rates, and precipitation. Moreover, the selected time horizon is aligned with the 20-year midterm period (2041-2060) defined by the Intergovernmental Panel on Climate Change (IPCC) to project the impact of climate change (IPCC, 2021).

In line with the available data, we considered projections of soil organic carbon, soil erosion rates, and precipitation according to three Representative Concentration Pathways (RCP) spanning from the trajectory of ambitious greenhouse gas (GHG) emissions mitigation (RCP2.6) to the less aggressive one (RCP8.5) and including an intermediate trajectory of GHG emissions mitigation (RCP4.5).

6.2.3.2 Characterization factors for copper terrestrial ecotoxicity

The characterization model for metals terrestrial ecotoxicity utilized in this study is based on a methodological framework that employs multiple linear regressions to compute metal speciation and bioavailability in soil (Owsianiak et al., 2013). According to Equation 6.1, the characterization factor $CF_{i,s}$ ($PAF \cdot m^3 \cdot day \cdot kg_{total\ emitted}^{-1}$) of total metal *s* emitted to the environmental compartment *i* is composed of a fate factor $FF_{i,s}$ (day) for total metal *s* in soil, an accessibility factor ACF_s ($kg_{reactive} \cdot kg_{total}^{-1}$) that expresses the fraction of reactive metal over total metal *s* in soil, a bioavailability factor BF_s ($kg_{free} \cdot kg_{reactive}^{-1}$) that quantifies the free ion fraction of the reactive metal *s* in soil, and an effect factor EF_s ($PAF \cdot m^3 \cdot kg_{free}^{-1}$) that represents the potentially affected fraction (PAF) of soil organisms by the free ion metal in soil.

$$CF_{i,s} = FF_{i,s} \cdot ACF_s \cdot BF_s \cdot EF_s \tag{6.1}$$

In section C2 of the supplementary materials, we present the empirical regression models used for calculating the different pools of copper in soil (metal speciation), as well as the equations for computing the different intermediate factors of Equation 6.1. To integrate the effect of expected changes in soil properties and climatic conditions altered by climate change on future CFs for copper terrestrial ecotoxicity, we used projections of soil organic carbon, soil erosion rates, and precipitation. The selection of these parameters is justified by their influence on the fate and bioavailability of metals in soil (Droz et al., 2021; Owsianiak et al., 2013). The projected changes in parameters were integrated into the computation of CFs for copper terrestrial ecotoxicity employing the empirical regression models. The influence of the parameters affected by climate change on the intermediate factors of CFs (as per Equation 6.1) are summarized in Table 6.1.

Table 6.1 Changes that were introduced in the characterization modelling to simulate the influence of projected environmental conditions on CFs for copper terrestrial ecotoxicity.

Parameter influenced by projected climate change (source)	**Affected parameter**	**Equation (Model)**	**Affected factor of Equation 6.1**
Projections of soil erosion (Panagos et al., 2021)	Transfer rate from soil to freshwater	Equation C1 (USEtox)	$FF_{i,s}$
Projections of precipitation (Fick & Hijmans, 2017; Petrie et al., 2021)	Transfer rate from soil by leaching	Equation C1 Equation C2 (USEtox)	$FF_{i,s}$
Projections of soil organic matter (Yigini & Panagos, 2016)	$Cu_{total\ dissolved}$ K_d	Equation C7 Equation (C10)	$FF_{i,s}$
	$Cu_{reactive}$	Equation C6	ACFs, BFs
	Cu_{free}	Equation C8	BFs

For the computation of fate factors of copper in agricultural soil ($FF_{i,s}$), we employed USEtox, the UNEP/SETAC scientific consensus multimedia model for characterizing the (eco)toxicological impacts of chemicals in LCA (P Fantke et al., 2017). The landscape parameters of USEtox were modified to account for current and projected values of soil erosion rates and average annual precipitation. Moreover, we considered partitioning coefficients (K_d) computed for each mapping unit according to Equation C10. In section C1.2 of the supplementary materials, we present a preliminary sensitivity analysis of fate factors to partitioning coefficients, average annual precipitation, and soil erosion, as computed with USEtox. This brief sensitivity analysis aimed to confirm that USEtox allows accounting for changes in average annual precipitation, soil erosion, and partitioning coefficients in fate factors for copper emissions.

In the computation of future CFs for copper terrestrial ecotoxicity, soil organic carbon, soil erosion rates, and precipitation were the only parameters that we considered as influenced by climate change in the future, whereas other soil properties were considered constant. The latter assumption is in line with the studies that generated maps of projected SOC stocks and soil erosion rates (Panagos et al., 2021; Yigini & Panagos, 2016). Soil pH influences metal speciation and this soil property is also vulnerable to changing environmental conditions driven by climate change. Nonetheless, the dynamic of pH in the soil is slow in comparison to other environmental compartments such as freshwater and the ocean, because of the buffer effect of soil minerals. Yet, in some circumstances such as those of increased rainfall, soil pH may be altered due to the leaching of basic cations, leading to acidic soils (Biswas et al., 2018). However, to the best of our knowledge, there are no projections on future soil pH estimated according to environmental conditions influenced by climate change. Furthermore, the temporal resolution of USEtox hinders the modelling of time-specific events such as heavy rainfall.

The EFs ($PAF \cdot m^3 \cdot kg_{free}^{-1}$) were computed following the USEtox approach, as per Equation 6.2, where ΔPAF is the incremental change in the potentially affected fraction of species per free ion concentration in soil (ΔC_{free}), $HC50_{EC50}$ is the geometric mean of distinct median effective concentration (EC_{50}) values, that is the concentration that leads to an observed effect in 50% of organisms (P. Fantke et al., 2018). In keeping with previous studies addressing the calculation of CFs for metals terrestrial ecotoxicity (Owsianiak et al., 2013; Ivan Viveros Santos et al., 2018), terrestrial biotic ligand models (TBLMs) were employed to compute EC_{50} of six different endpoints (Table C2) (Thakali et al., 2006). TBLMs have been considered an adequate method to compute site-dependent EFs including metal speciation, but their range of applicability is limited to non-calcareous soils. The TBLMs for computing EC_{50} values for copper take as parameters the activities of hydrogen and magnesium. The latter parameter was computed following the modelling of cation exchange described in Owsianiak et al., (2013). Hydrogen and magnesium activities were considered constant; therefore, for a given mapping unit, the EF is the same for both current and future scenarios. The use of TBLMs allowed integrating metal speciation and bioavailability but precluded including the effect of changing parameters such as temperature on the extent of effect factors. However, it is still challenging to disentangle the influence of changing environmental conditions on species sensitivity from the changes caused by modifications of metal speciation (Fu et al., 2018; González-Alcaraz and van Gestel, 2016). The inclusion of ACFs and BFs in the computation of future CFs permitted to consider the influence of changes in organic matter content on bioavailability, but the potential impact of changing temperature on the sensitivity of terrestrial organisms was not included.

$$EF = \frac{\Delta PAF}{\Delta C_{free}} = \frac{0.5}{HC50_{EC50}} \tag{6.2}$$

6.2.3.3 Analysis and attribution of the changes in characterization factors

Projected changes in CFs ($\Delta\mathrm{CF}_{\mathrm{i,s}}$) can be associated with the changes in FFs, ACFs, and BFs according to the differential of Equation 6.1:

$$\begin{aligned}\Delta\mathrm{CF}_{\mathrm{i,s}} &= \frac{\mathrm{dCF}_{\mathrm{i,s}}}{\mathrm{dFF}_{\mathrm{i,s}}}\Delta\mathrm{FF}_{\mathrm{i,s}} + \frac{\mathrm{dCF}_{\mathrm{i,s}}}{\mathrm{dACF}_{\mathrm{s}}}\Delta\mathrm{ACF}_{\mathrm{s}} + \frac{\mathrm{dCF}_{\mathrm{i,s}}}{\mathrm{dBF}_{\mathrm{s}}}\Delta B\mathrm{F}_{\mathrm{s}} + \frac{\mathrm{dCF}_{\mathrm{i,s}}}{\mathrm{dEF}_{\mathrm{s}}}\Delta E\mathrm{F}_{\mathrm{s}} \\ &= \mathrm{ACF}_{\mathrm{s}} \cdot \mathrm{BF}_{\mathrm{s}} \cdot \mathrm{EF}_{\mathrm{s}} \cdot \Delta\mathrm{FF}_{\mathrm{i,s}} + \mathrm{FF}_{\mathrm{i,s}} \cdot \mathrm{BF}_{\mathrm{s}} \cdot \mathrm{EF}_{\mathrm{s}} \cdot \Delta\mathrm{ACF}_{\mathrm{s}} + \mathrm{FF}_{\mathrm{i,s}} \cdot \mathrm{ACF}_{\mathrm{s}} \cdot \mathrm{EF}_{\mathrm{s}} \cdot \Delta\mathrm{BF}_{\mathrm{s}}\end{aligned} \tag{6.3}$$

where $\mathrm{FF}_{\mathrm{i,s}}$, $\mathrm{ACF}_{\mathrm{s}}$, and BF_{s} are the corresponding values of these intermediate parameters for the baseline scenario; Δ represents the projected changes in intermediate parameters compared to the baseline period, and the EF was assumed constant for a given mapping unit. The largest term on the right side in Equation 6.3 would be interpreted as the dominant factor in $\Delta\mathrm{CF}_{\mathrm{i,s}}$.

6.2.3.4 Spatial differentiation of characterization factors

To evaluate the influence of spatial differentiation and to facilitate their use by LCA practitioners, the computed CFs for copper terrestrial ecotoxicity at the native resolution of 500 m were aggregated into three lower spatial resolutions: wine-growing regions, European regions, and country. Furthermore, we computed an aggregated CF for the non-calcareous European vineyard soils. The spatial differentiation at the country level was considered because the available datasets from life cycle inventory databases are generally at this level, which may simplify connecting spatialized elementary flows to regionalized CFs (Patouillard et al., 2020). The second spatial differentiation was at the level of European regions, which are defined by the NUTS2 level. The NUTS classification (Nomenclature of Territorial Units for Statistics) corresponds to the European

Union system for defining administrative units at different spatial levels, namely countries, regions, provinces, and municipalities (European Commission, 2022a). The spatial differentiation at the level of European regions was chosen since it is frequently employed for formulating policies, in addition, several environmental indicators have been reported at this scale (Ballabio et al., 2018; Panagos et al., 2018, 2021). The spatial differentiation at the level of wine-growing regions allows using site-dependent CFs in cases where the site of emission is known.

While some impact methods in LCA, such as IMPACT World+, recommend aggregating CFs at native resolution into lower spatial resolution based on emission or extraction data as a proxy for the probability of emissions or extractions occurring in a given point of space (Bulle et al., 2019), we performed the aggregation according to an area-weighted average (Equation 6.4). The reason for this modelling choice is the lack of detail on sales of fungicides per category (Panagos et al., 2018). Still, European vineyards rely heavily on copper-based fungicides (Ballabio et al., 2018; Droz et al., 2021). Moreover, the aggregation criterium used in this study is consistent with previous studies that computed CFs for copper terrestrial ecotoxicity (P Villanueva-Rey et al., 2019; Ivan Viveros Santos et al., 2018). In Equation 6.4, CF_i is the characterization factor of the overlapped vineyard soil *i,* and *Ai* represents the surface of vineyards within a given aggregation region (wine-growing regions, NUTS2, or country). The aggregations of CFs at the spatial resolution of wine-growing regions, NUTS2 regions, and countries were performed using the R package *exactextractr* (Baston, 2020).

$$CF_{area-weighted\,average} = \frac{\sum_{i=1}^{n} CF_i \cdot A_i}{\sum_{i=1}^{n} A_i} \tag{6.4}$$

6.2.3.5 Data source

6.2.3.5.1 Current and future soil organic matter in Europe

Current (2010) and future (2050) soil organic carbon (SOC) stocks in Europe were obtained from the study by Yigini and Panagos (2016), through the European Soil Data Centre (ESDAC) (Panagos et al., 2012). The map of current SOC stocks was derived utilizing the regression kriging geostatistical technique, which relied on 22,300 soil samples (0-20 cm) from the LUCAS Topsoil Database and data on terrain, climate, land cover use, and other soil properties, namely soil texture and available water capacity as environmental predictors (Yigini & Panagos, 2016). Regarding the projections of future SOC stocks, the researchers hypothesized that, by 2050, soil organic carbon is controlled primarily by climate, land cover, and inherent soil properties. Accordingly, maps of future SOC stocks were calculated by using the fitting regression derived from the base model and projections of climate and land cover. The researchers acknowledge that soil organic carbon content is driven predominantly by the balance between net primary production (NPP) from vegetation and the degradation rate of organic matter. Nevertheless, it is projected that climate change will affect SOC levels in the long term, whereas the impact of land-use change and land management practices will have a greater impact in the short term (Yigini & Panagos, 2016).

The projections of SOC are available according to four global climate models (GCMs) and four RCP scenarios. Since the main purpose of our study is to compute the relative magnitude of change in CFs for copper terrestrial ecotoxicity, we considered the projected SOC stocks derived from the GCM IPSL-CM5A-LR, in line which a previous study that addressed the impact of climate change on the environmental performance of viticulture (Ivan Viveros Santos et al., 2022). Besides, to assure consistency across the parameters influenced by climate change considered in this study, we selected the projections under the RCP2.6 and RCP8.5 scenarios in line with Viveros Santos et al., (2022), and included the intermediated RCP4.5.

Because soil organic matter (OM) and dissolved organic carbon (DOC) are required for calculating metal speciation (Equations C6, C7, C8), we first produced maps of soil organic carbon content (*SOC%*) by means of the *raster* package in R (Hijmans et al., 2022), using the maps of SOC stocks (SOC_{stocks}) expressed in tonnes·ha^{-1}, the soil sampling depth (d =20 cm), and a map of soil bulk density (ρ_b) in g·cm^{-3}. The latter computations were performed according to Equation 6.5, which was used in the study by Yigini and Panagos (2016). The map of bulk density was taken from the study by Ballabio et al. (2017) and downloaded from the ESDAC portal (Panagos et al., 2012).

$$\mathrm{SOC\%} = \frac{SOC_{stock}}{\rho_{\mathrm{b}} \times d} \tag{6.5}$$

Based on the derived maps of SOC (%), we generated maps of soil organic matter content (OM) (Figure C3) considering the relationship $\mathrm{OM} = 1.72 \cdot \mathrm{SOC}$, that is, assuming that organic matter (OM) contains 58% of organic carbon, which is in line with previous studies that computed site-dependent CFs for metals terrestrial ecotoxicity accounting for speciation and bioavailability

(Plouffe et al., 2016; Ivan Viveros Santos et al., 2018). Finally, in line with Viveros Santos et al. (2018) and Owsianiak et al. (2013), we computed maps of DOC (mg/l) according to an empirical regression (Equation C3) reported by Römkens et al. (2004).

6.2.3.5.2 Current and future soil erosion rates in Europe

The maps of current (2016) and projected soil erosion rates (2050) in Europe used in this study were computed by Panagos et al. (2021) and made accessible by ESDAC (Panagos et al., 2012). The authors applied the European version of the Revised Universal Soil Loss Equation (RUSLE), incorporating rainfall erosivity change, projections of land-use change, and expected changes in management practices promoted by European policy. Future rainfall erosivity was estimated by employing 19 GCMs according to three RCP scenarios. Even though the researchers reported high variability in changes in soil erosion between the GCMs, the maps of projected soil erosion are provided as an average composite of the 19 GCMs employed under each RCP. Therefore, for projected soil erosion rates we were not able to select data derived according to a specific GCM.

The datasets of soil loss rate in $Mg \cdot ha^{-1} \cdot yr^{-1}$ were converted into units of $mm \cdot yr^{-1}$ to make them suitable for computing fate factors with USEtox (Figure C5). Hence, the layers of soil loss rate ($Mg \cdot ha^{-1} \cdot yr^{-1}$) were divided by the bulk density ($Mg \cdot m^{-3}$) and multiplied by a unit conversion factor of 0.1 ($ha \cdot mm \cdot m^{-3}$).

6.2.3.5.3 Current and future precipitation in Europe

We obtained monthly total precipitation (mm) datasets from WorldClim 2.0 data portal for the current scenario (1970-2000) and the future scenario by 2050 under RCP2.6, RCP4.5, and RCP8.5 as modelled by the GCM IPSL-CM6A-LR (Fick & Hijmans, 2017; Petrie et al., 2021). The spatial resolution of the retrieved maps is 30 seconds (1 km^2 resolution). The prospective scenario by 2050 corresponds to the average climate of the period 2041-2060, which is consistent with the studies that simulated the projections of SOC stocks (Yigini & Panagos, 2016) and soil erosion by 2050 (Panagos et al., 2021) that were used in this study. Since the datasets of precipitation are inputs for the computation of fate factors with USEtox (Equations C1 and C2), layers of average annual precipitation ($mm \cdot yr^{-1}$) were calculated in Python employing the *rasterio* module (Figure C7).

6.2.3.5.4 Auxiliary soil data

The layers of soil properties that were considered constant under both current and future scenarios were retrieved from the ESDAC portal (Panagos et al., 2012), which sources are listed in Table 6.2.

Table 6.2 Source of auxiliary datasets of soil properties for the computation of copper speciation

Parameter	Unit	Database	Source
Bulk soil density	kg/l	-	(Ballabio et al., 2016)
Clay content	% (w/w)		
pH, measured in water	-	Maps of Soil Chemical properties at European scale based on LUCAS	(Ballabio et al., 2019)
Cation exchange capacity	cmolc/kg		
Calcium carbonate content	%		
Base saturation	%	Soil profile analytical database 14 (SPADE 14)	(Breuning-Madsen et al., 2018; Kristensen et al., 2019)
Exchangeable calcium	cmolc/kg		
Exchangeable magnesium	cmolc/kg		
Exchangeable sodium	cmolc/kg		
Exchangeable potassium	cmolc/kg		
Electrical conductivity of soil pore water	dS/m	LUCAS 2015 TOPSOIL data	(Fernández-Ugalde et al., 2020; Orgiazzi et al., 2018)
Copper content	mg/kg		(Ballabio et al., 2018; Droz et al., 2021; Orgiazzi et al., 2018)

6.2.4 Results and discussion

6.2.4.1 Characterization factors under present and future scenarios

Figure 6.1 illustrates the regionalized CFs for copper terrestrial ecotoxicity in non-calcareous European vineyard soils for the baseline period and by 2050 under RCP2.6, RCP4.5, and RCP8.5. CFs were only computed for non-calcareous soils since the TBLMs used to calculate EFs are valid solely for these soils. Still, using TBLMs allowed to account for the metal speciation and bioavailability in the computation of EFs. For the current scenario, the spatial variability of CFs spans 1.96 orders of magnitude between the minimum and the maximum value, with a mean value of 1.46 x 10^3 PAF·m^3·day·kg^{-1}. In comparison, Peña et al. (2018) reported similar findings on the geographical variability of CFs for copper in European vineyard soils, which was found to extend more than 1.5 orders of magnitude with a mean value of 2.34 x 10^3 PAF·m^3·day·kg^{-1}. Viveros Santos et al., (2018) derived CFs employing the method based on the geochemical speciation model WHAM 6.0 and found a spatial variability of around 5.5 orders of magnitude for non-calcareous soils of the world, with a median value of 1.73 x 10^3 PAF·m^3·day·kg^{-1}.

The CFs for both current and future scenarios depict a similar tendency with soil organic matter (Fig. C9). High organic matter content leads to low values of CFs because of the high affinity of copper for organic matter (Droz et al., 2021), which decreases copper mobility, and accordingly the associated FF. Likewise, higher values of CFs occur where pH is low since copper solubility is dependent on soil pH values, and it tends to increase in acidic soils at pH lower than six (Fig. C10). Furthermore, lower FFs are related to lower K_d values (Fig. C12), that is, in cases where copper concentration in the aqueous phase is higher (Equation C10), which results in a higher influence on removal processes, namely runoff and leaching (Equations C1 and C2). This situation leads to potential increases in characterization factors for aquatic ecotoxicity; however, it is out of the scope of this study since this would require computing metal speciation according to freshwater properties to be consistent with the modelling approach of terrestrial ecotoxicity accounting for metal speciation. Moreover, higher values of CFs are associated with lower annual precipitation since the contribution of the removal processes decreases for these values of annual precipitation (Fig. C11).

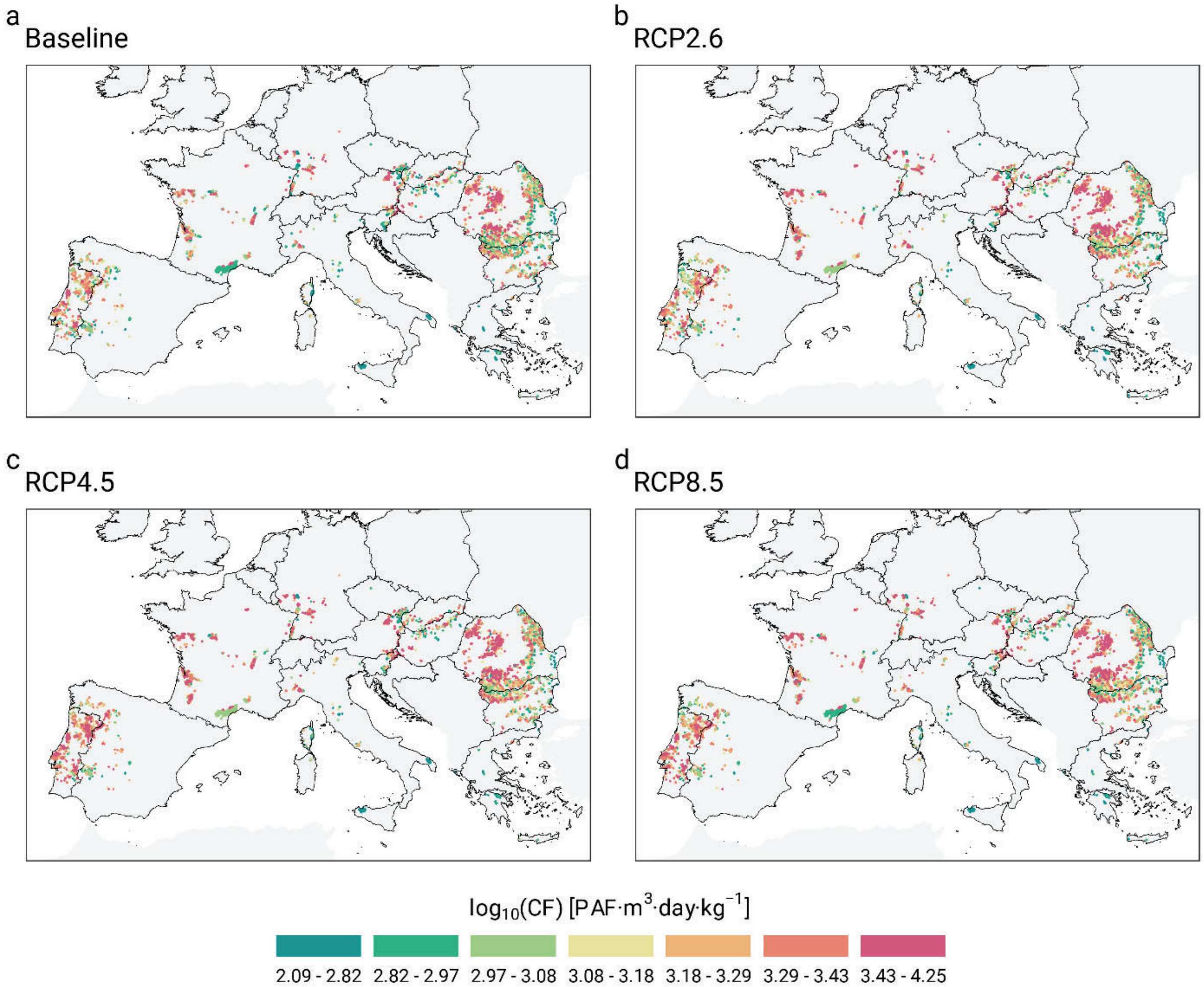


Figure 6.1 Regionalized CFs (PAF·m^3·day·kg^{-1}) for copper terrestrial ecotoxicity under the baseline period (a), RCP2.6 (b), RCP4.5 (c), and RCP 4.5 (d) across European vineyards (coloured by quantiles). The reader is invited to consult the interactive version of this figure at: https://iviveros.github.io/viveros-santos_et_al_2022_jclp/#figure-1

The spatial patterns of CFs shown in Figure 6.1 seem to vary slightly between the analyzed scenarios because the spatial variability of CFs is higher than the temporal variability, the latter resulting from the comparison of CFs for the mid-term future scenario with those for the baseline scenario. Concerning the baseline scenario, CFs in the range of 2.09 to 2.97 orders of magnitude (considering a base-10 logarithmic scale) amount to 46% of the modelled vineyard surface. More specifically, CFs in the range of 2.82 to 2.97 orders of magnitude cover the largest area fraction for this scenario, which corresponds to 29%, while the lowest share of the vineyard surface was found for CFs in the range of 3.43 to 4.25 orders of magnitude, comprising 6% of non-calcareous vineyard soils. In contrast, under RCP2.6 and RCP4.5 scenarios, the highest area fraction corresponds to CFs in the range of 2.97 to 3.08 orders of magnitude, with 44% and 40% shares of land area, respectively. The spatial patterns of CFs under RCP4.5 in the range of 2.09 to 3.29 orders of magnitude resemble the distribution of those under RCP2.6, whereas the former scenario comprises a higher fraction of CFs in the range of 3.29 to 4.25 orders of magnitude, with a 28% share of land area, compared to 21% under RCP2.6. The spatial distribution of CFs under RCP8.5 in the range of 2.82 to 3.29 orders of magnitude roughly mirrors that under the baseline scenario. However, the fraction of CFs in the range of 3.29 to 4.25 orders of magnitude for the former scenario is 23%, while it amounts to 15% for the baseline scenario.

By 2050, under the three RCP scenarios, the median CFs for copper terrestrial ecotoxicity are projected to increase in comparison with that for the baseline scenario. The highest increase of the median CF is projected under RCP4.5 with a value of 1.49 x 10^3 PAF·m^3·day·kg^{-1}, compared to 1.19 x 10^3 PAF·m^3·day·kg^{-1} for the baseline scenario (+27%). This finding is explained by the fact that the RCP4.5 predicts the lowest decreases in soil organic matter content in comparison with the other scenarios. The median CFs for RCP8.5 and RCP2.6 scenarios are respectively 1.40 x 10^3 and 1.34 x 10^3 PAF·m^3·day·kg^{-1}, which corresponds to increases of 19% and 14%, respectively. Moreover, the spatial variability of CFs, calculated as the range between the lowest and the highest value, is predicted to increase under RCP2.6 and RCP8.5, which is 2.13 and 2.06 orders of magnitude, respectively, whereas geographical variability under RCP4.5 will be equal to the current variability (1.96 orders of magnitude).

Figure 6.2a shows the distribution of site-dependent CFs for copper terrestrial ecotoxicity across non-calcareous soils in European countries. Besides, Figure 6.2b illustrates the vineyard surface fraction for which CFs were derived. Because of the range of applicability of the TBLMs for calculating EFs, CFs were derived for around 30% of European vineyard soils. Low area coverage of CFs was obtained for Spain, Greece, and Italy, with values of 9%, 10%, and 13% of the vineyard surface, respectively. For France, Germany, and Bulgaria, we obtained intermediate area coverage of 31%, 33%, and 36%, respectively. Larger vineyard surface coverage was obtained for Slovenia, Romania, and Portugal, with 55%, 64%, and 86% of the vineyard surface, respectively (Figure 6.2b). In section 3.3, we present CFs aggregated at different spatial resolutions that could be used as an approximation of missing CFs. For instance, an aggregated CF at the country level could be used with the corresponding spatial variability set by the range of the minimum and the maximum value to give an approximation of the associated uncertainty of ignoring the corresponding CF for that region.

As shown in Figure 6.2a, the lowest spatial variability of CFs was obtained for Luxembourg under both present and future scenarios, extending over 0.65 orders of magnitude within a 95% interval (2.5th and 97.5th percentiles). This is explained by the low share of the vineyard surface in this country, corresponding to roughly 0.04% of the total European vineyard surface. Likewise, under the baseline scenario, CFs for Czechia exhibited a reduced spatial variability of 0.78 orders of magnitude in a 95% interval, in line with the low share of vineyard surface of this country (0.31%). Nevertheless, the spatial variability of the CFs for Czechia is predicted to increase under future scenarios with an average of 0.87 orders of magnitude within a 95% interval. Spain has roughly a 21% share of the vineyard surface; however, because of the low vineyard surface modelled for this country, the corresponding CFs under the baseline scenario showed a low spatial variability of 0.87 orders of magnitude. Nevertheless, this spatial variability is expected to increase under RCP scenarios to around 0.91 orders of magnitude. Furthermore, despite the largest share of the vineyard surface of France (around 32%), the geographical variability of the CFs for this country was small and corresponded to 1.04 orders of magnitude under the baseline scenario, which is explained in part by the low vineyard surface fraction modelled (31%).

The largest spatial variability of CFs for non-calcareous European vineyard soils was found in Austria, expanding over 1.35 orders of magnitude within a 95% interval under the baseline scenario. Under RCP scenarios this variability is predicted to increase to around 1.40 orders of magnitude (Figure 6.2a). The variability of site-dependent CFs for Austria contrasts with the low share of vineyard surface of this country; however, the corresponding vineyard surface fraction modelled was relatively high (47%), which indicates greater variability in soil properties. Similarly, the CFs for Hungary, under the current scenario, showed a high spatial variability of 1.32 orders of magnitude. Comparably, the CFs for Italy exhibited a high geographical variability of 1.23 orders of magnitude under the current scenario, which is explained by the high share of the European vineyard surface of this country (14%), even though the vineyard surface fraction modelled was relatively low (12%). The spatial variability of CFs for Italy is projected to increase to around 1.31 orders of magnitude under future scenarios. A similar spatial variability of CFs under the current scenario was obtained for Germany and Romania, which is 1.16 orders of magnitude.

Under the RCP4.5 scenario, the median CF values for all countries are projected to increase related to the corresponding ones under the baseline scenario, which indicates a general increase in CFs under the former scenario (Figure 6.2a). Under RCP2.6 and RCP8.5 scenarios, there are some exceptions to the trend of projected increases in median CFs across all countries. Regarding the RCP2.6 scenario, it is expected that the median CF for Portugal slightly decreases in comparison with the baseline scenario. This is explained by the projected increases in soil OM (%) in a larger area of Portugal under RCP2.6 related to other RCP scenarios (Fig. C4). In fact, under the current scenario, CFs for Portugal in the range of 2.97 to 3.29 orders of magnitude extend over an area fraction of 70%, while CFs in the same range under RCP2.6 are projected to comprise an area fraction of 57% (Figure 6.1). Similarly, under RCP8.5 the median CF for Hungary is expected to slightly decline compared to the median CF under the current scenario. While under the baseline scenario, the CFs for Hungary in the range of 3.08 to 4.25 orders of magnitude have a 40% share of modelled area, the RCP8.5 scenario predicts a reduction of this area fraction to 35% because of projected higher increases in soil OM (%) in comparison with the other RCP scenarios.

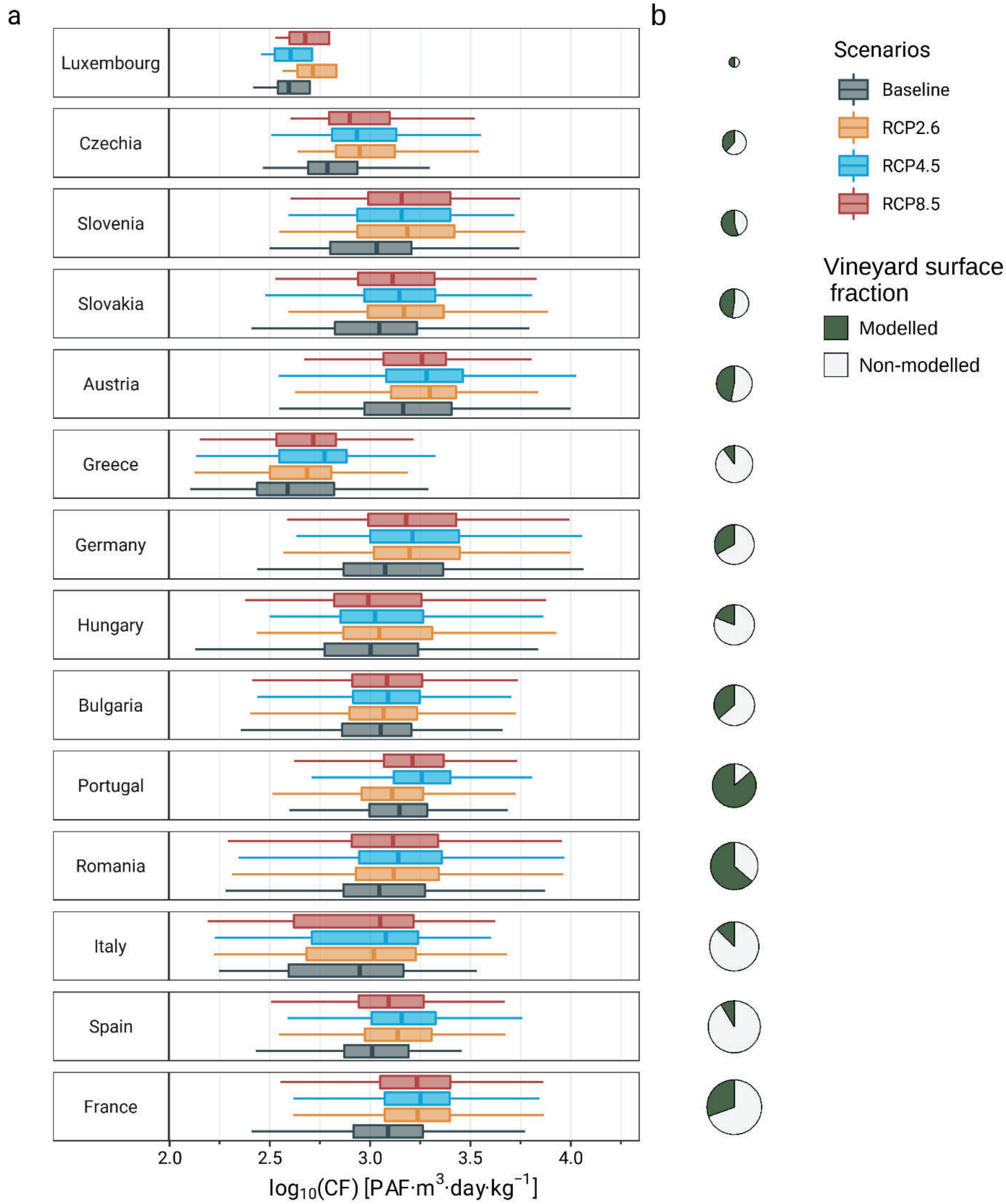


Figure 6.2 Distribution of regionalized CFs (PAF·m^3·day·kg^{-1}) for copper terrestrial ecotoxicity (a). The area of the pie charts is proportional to the soils under vineyards for each country (logarithmic scale), and the green area indicates the vineyard surface fraction for which CFs were computed (non-calcareous soils) (b).

Future scenarios suggest that the changes in soil organic carbon, soil erosion, and rainfall may either increase or decrease CFs for copper terrestrial ecotoxicity by 2050 (Figure 6.3). However, CFs under future scenarios will tend toward an overall increase, which explains the increases in median CFs for most European countries across the RCP scenarios (Figure 6.2).

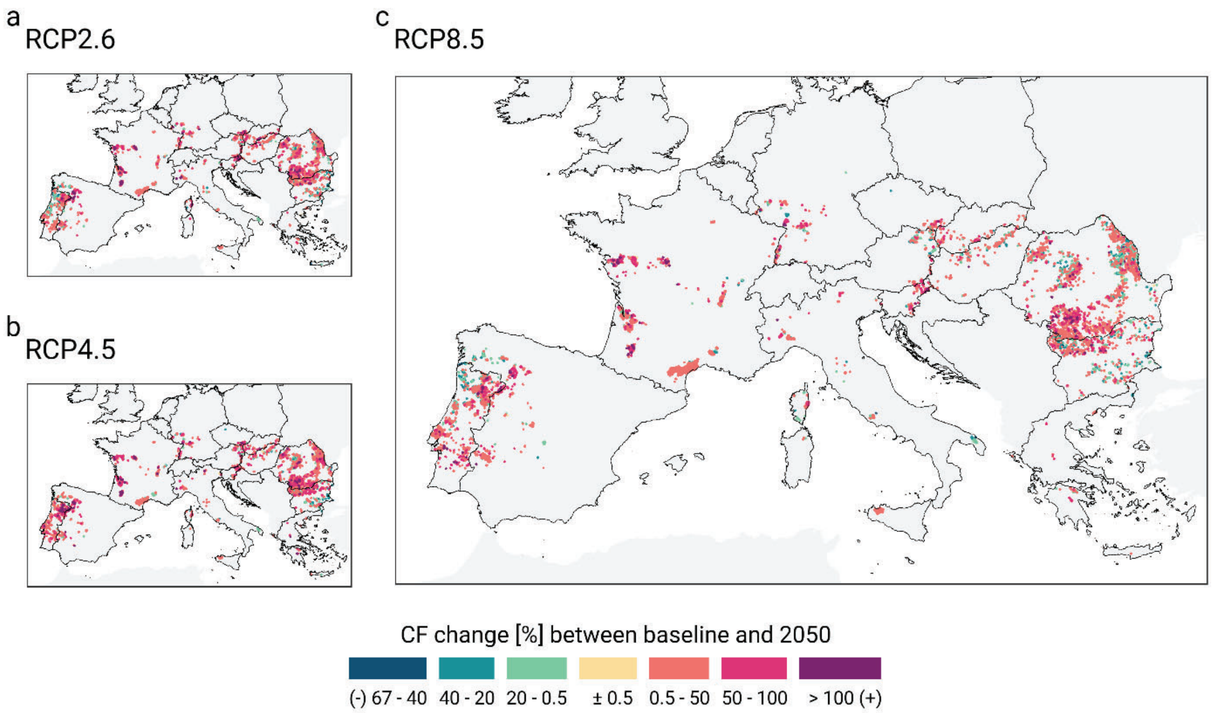


Figure 6.3 Relative change (%) in area-weighted CFs compared to the baseline period for RCP2.6 (a), RCP4.5 (b), and RCP8.5 (c) scenarios across European vineyards. The reader is invited to consult the interactive version of this figure at: https://iviveros.github.io/viveros-santos_et_al_2022_jclp/#figure-3

In terms of area fraction, the higher increases in CFs are projected under RCP4.5 with an 82% share of land area, followed by RCP8.5 with 79.5%, and RCP2.6 with 74% of the non-calcareous vineyard area. Under the RCP4.5 scenario, most of the projected increases in CFs will be in the range of 10%-20%, encompassing a 29% share of the land area, followed by increases by 50%-100% representing 15% of the modelled vineyard surface (Figure 6.3). In contrast, under RCP2.6 the highest increases in CFs are expected in the range of 20%-30%, comprising an area fraction of approximately 26%, followed by increases in CFs by 0.5%-10%, encompassing 15% of the non-calcareous vineyard area. The RCP8.5 scenario projects lower increases in future CFs for copper, which will be mainly in the range of 0.5%-10%, with a 35% share of non-calcareous vineyard surface, which is the highest share for this range of increases among the studied scenarios. Still, the RCP8.5 scenario projects the highest share of the land area for increases in CFs in the range of 30%-40%, extending over a 12% share of non-calcareous vineyard soils. The three considered RCP scenarios predict increases in CFs higher than 100%; however, the area fraction of these increases is low: 1.8%, 2.2%, and 1.3% under RCP2.6, RCP4.5, and RCP8.5, respectively. Likewise, the projected vineyard surface where CFs will not change considerably (in the range of ±0.5%) is low, comprising around a 0.5% share of the land surface.

Regarding the projected declines in CFs under future scenarios, the highest decreases are in the range of -20% to -0.5%. More specifically, the RCP2.6 scenario predicts the highest decrease in CFs in the range of -20% to -0.5% over an area fraction of 17.4%, followed by the RCP4.5 scenario that projects a 14.6% area fraction, and by the RCP8.5 scenario with 12.6%. RCP2.6 and RCP8.5 scenarios predict approximately the same area fraction for expected reductions in CFs by -40% to -20%, which corresponds to nearly 7% of the non-calcareous vineyard surface. RCP4.5 and RCP8.5 scenarios project a 0.4% area fraction for decreases in CFs by -67% to -40%, whereas the RCP2.6 scenario predicts a 1.2% share of the non-calcareous vineyard surface for the same range in declines.

6.2.4.2 Contributions of changes in fate, accessibility, and bioavailability to characterization factors

Changes in future CFs are related to changes in FFs, ACFs, and BFs (Equations 6.1 and 6.3), driven by changes in soil organic matter, rainfall, and soil erosion (Table 6.1). Figure 6.4 shows the major contributors to changes in CFs across non-calcareous European vineyard soils under three RCP scenarios. It was found that the changes in BFs will be the principal contributors to the changes in CFs in around 89% of the modelled European vineyard surface. Particularly, the increases in CFs will be controlled by the increases in BFs in roughly 87% of the total vineyard surface modelled, while the remaining 2% of the vineyard surface will exhibit decreases in CFs resulting from reductions in BFs. The vineyard surface where increases in CFs will be dominated by rises in BFs is the result of projected declines in organic matter under RCP scenarios compared to the baseline scenario (Fig. C4). This is explained by the operational definition of the BF, which corresponds to the free ion fraction of the reactive metal in soil (Equation C12). While lower matter content leads to decreases in reactive copper concentration, it causes increases in free copper concentration (Equation C8), which leads to an overall increase in BFs. The characterization modelling of this study does not account for soil acidification resulting from episodes of extreme rainfall, which would result in a higher fraction of free copper concentration and consequently higher BFs. Besides, soil erosion affects soil organic matter, but the proposed characterization model only accounts for the direct impact of soil erosion on the fate of copper in the soil but neglects its influence on metal speciation, induced by its impact on organic matter content. Among the RCP scenarios considered in this study, it was found that the major contributions of increases in BFs will be observed under RCP4.5 with a 92% share of non-calcareous vineyard surface, whereas the least positive contribution will be expected under RCP2.6 in 81% of the non-calcareous vineyard soils (Figure 6.4).

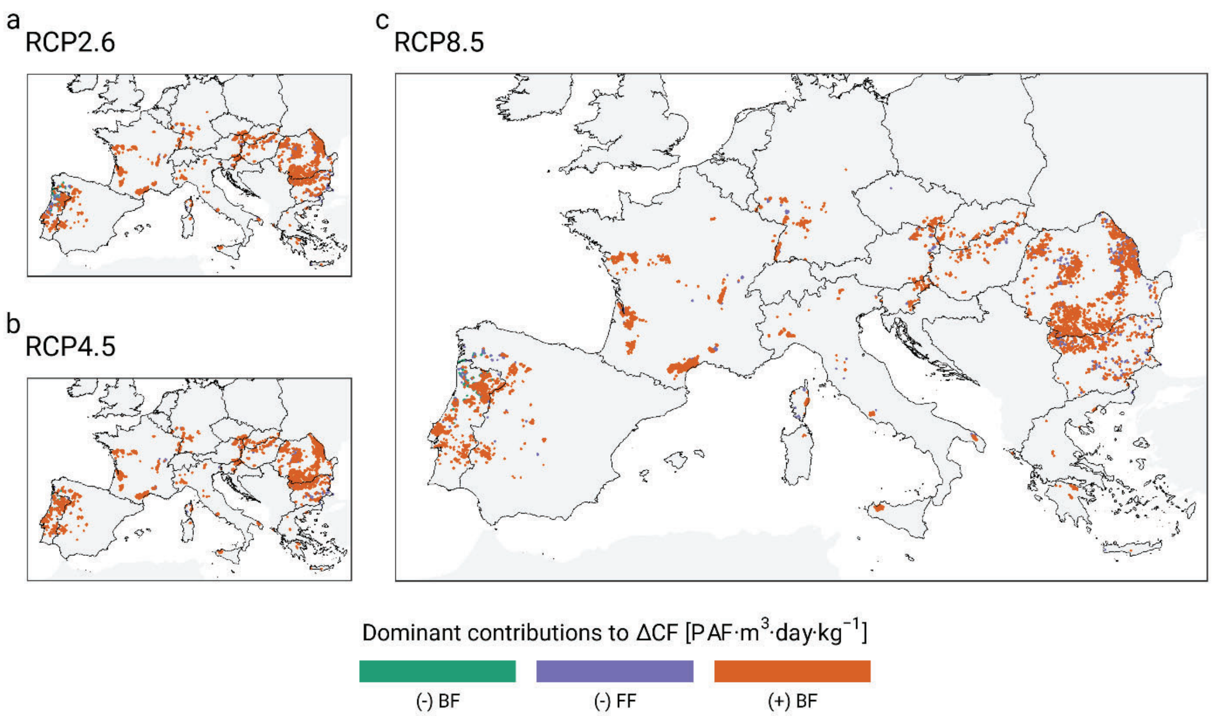


Figure 6.4 Dominant contributions to changes in CFs (PAF·m^3·day·kg^{-1}) under RCP2.6 (a), RCP4.5 (b), and RCP8.5 (c) across European vineyards. (-) and (+) indicate decreases and increases, respectively, in BF or FF.

Changes in FFs are the second principal contributors to projected changes in CFs. Nonetheless, in contrast to BFs, changes in FFs will solely lead to decreases in CFs. This was found for around

11% of the non-calcareous vineyard area. In particular, the highest contribution of changes in FFs towards changes in CFs is expected under the RCP2.6 scenario for approximately 14% of the modelled vineyard surface, whereas the lower contribution of FFs towards decreases in CFs is projected under the RCP4.5 with a vineyard area fraction of 8%. The declines in FFs are explained by the interaction of changes in K_d values, precipitation, and soil erosion projected according to the RCP scenarios. On average across the scenarios, for around 63% of the surface where FFs dominate the changes in CFs, soil erosion is the main contributor to the declines in FFs due to an increase of the removal of copper by runoff (Equation C1). Whereas for roughly 23% of the same area, K_d values are expected to increase; however, their impact is counterbalanced by the projected increases in precipitation, leading to an increased influence of removal processes, namely runoff and leaching (Equations C1 and C2). For the remainder 14% of the area where changes in FFs control the changes in CFs, even though precipitation is projected to decline, the decrease in K_d values is projected to entail lower residence time of copper in the soil, thus decreasing the values in CFs.

6.2.4.3 Characterization factors at different spatial resolutions

The area-weighted CF for non-calcareous European vineyard soils under the current scenario is 1.32×10^3 PAF·m^3·day·kg^{-1}, which is around 10% lower than the mean value obtained for the same scenario. However, the aggregated CFs for European vineyard soils are predicted to increase under RCP scenarios. The highest rise in the area-weighted CF for copper at the European level is expected under RCP4.5, with an increase of 21% related to the baseline scenario (1.49×10^3 PAF·m^3·day·kg^{-1}). This is explained by the fact that a larger fraction of CFs falls in the range of 3.29 to 4.25 orders of magnitude under RCP4.5, with a share of 28%, in comparison to 15% under the baseline scenario, and 21% and 23% under RCP2.6 and RCP8.5, respectively (Figure 6.1). Accordingly, the second-highest increase in the aggregated CF is projected under RCP8.5, with an increase of 14% in comparison with the baseline scenario (1.51×10^3 PAF·m^3·day·kg^{-1}). Whereas the RCP2.6 scenario indicates a rise of 13% in the area-weighted CF compared to the current scenario (1.49×10^3 PAF·m^3·day·kg^{-1}). A potential application of these aggregated CFs for European vineyards is in cases where there are no site-dependent CFs, which is equivalent to using a generic CF, which is a current practice in LCA.

Figure 6.5 depicts the spatial patterns of CFs aggregated at the level of European regions. Based on the spatial analysis performed, European vineyards extend across 124 regions. However, in this study, CFs were derived for only 87 regions because of the range of applicability of the TBLMs employed for computing EFs and due to data unavailability on soil OM (for Croatia and Cyprus). More specifically, 1 region was missing from Cyprus, Czechia, Malta, and Portugal. Whereas 2 and 3 regions were missing from Hungary and Croatia, respectively. In addition, 4 regions were not defined from France and Greece. Italy and Spain are the countries with the largest number of regions excluded from the analysis, with 11 and 9 regions, respectively (Figure C13).

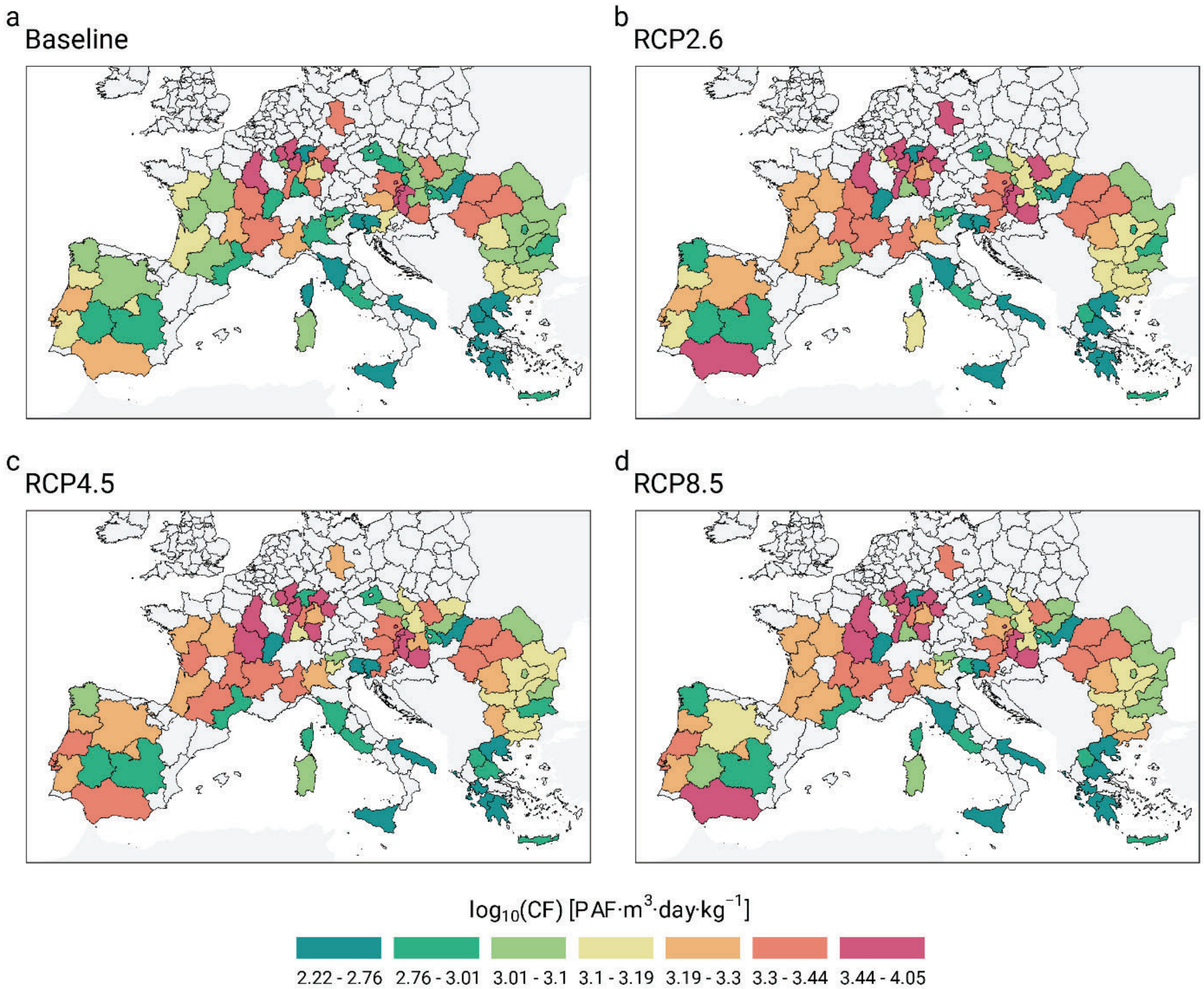


Figure 6.5 CFs (PAF·m^3·day·kg^{-1}) for copper terrestrial ecotoxicity aggregated at the level of European regions (NUTS2) under the baseline (a), RCP2.6 (b), RCP4.5 (c), and RCP 8.5

(coloured by quantiles). The reader is invited to consult the interactive version of this figure at: https://iviveros.github.io/viveros-santos_et_al_2022_jclp/#figure-5

As illustrated in Figure 6.5, the aggregation of CFs at the level of European regions changes the lower bound of the range of CFs at that of wine-growing regions (Figure 6.1) from 2.09 to 2.22 orders of magnitude (Figure 6.5), because the increases in CFs predominate over the projected declines in CFs (Figure 6.3). Apart from RCP4.5, on average across the scenarios, 16% of the European regions have CFs for copper in the range of 2.22 to 2.76 orders of magnitude. Greece and Italy have the highest number of regions with CFs within the latter range of CFs, with six and four regions, respectively under the baseline scenario, but this rank is preserved across future scenarios. Under the baseline scenario, CFs in the range of 2.76 to 3.19 orders of magnitude comprise 53% of the regions, however, under future scenarios this share is predicted to decrease to around 40%. For the latter range of CFs, France, Bulgaria, Italy, Romania, and Spain have the largest number of regions with CFs falling within that range, with seven, six, and five regions for the last three countries, respectively. Nonetheless, future scenarios suggest a sharp decrease in the number of regions under this range of CFs for France, changing from seven regions in the current scenario to two regions by 2050, due to the projected rises in CFs. While CFs in the range of 3.19 to 4.05 orders of magnitude for the baseline scenario cover 31% of the European regions, on average across the future scenarios, this share is expected to increase to 47% (Figure 6.5). Germany, France, and Austria have the highest number of regions with CFs falling in the range of 3.19 to 4.05 orders of magnitude, with 8, 5, and 4 regions, respectively. In the case of France, RCP scenarios project an increase in the number of regions with CFs falling in this range, moving from 5 to 10 regions.

Aggregating CFs at the level of European regions decreases the range of relative change from -67% to >100% at the level of the wine-growing regions (Figure 6.3) to the range of -38 to 85% (Figure 6.6). RCP2.6 and RCP4.5 scenarios project that for one region from Czechia and France, the corresponding aggregated CFs will decrease by -38% to -20%. Moreover, RCP4.5 predicts the same decrease for a region from Germany. Nonetheless, the decreases in CFs in the range of -20% to -10% exhibited an uneven geographical distribution. While the RCP2.6 predicts decreases in aggregated CFs in the latter range for one region from Bulgaria, Greece, Italy, and Spain; the RCP4.5 projects the same level of decrease for one region from Austria and Bulgaria. In contrast, the RCP8.5 predicts a higher number of regions where aggregated CFs fall in the range of decreases by -20% to -10%, more specifically, in one region from Austria, Bulgaria, Germany, Greece, and Italy. Furthermore, the spatial distribution of decreases in aggregated CFs by -10% to -2.5% was irregular. The RCP2.6 scenario projects some decreases in aggregated CFs for one region from five countries by -10% to -2.5%, while the RCP4.5 and RP8.5 forecast some declines in aggregated CFs in one region from six and four countries, respectively. Regarding the number of regions for which the aggregated CF will not change (±2.5% of variation), the RCP2.6 scenario projects this output for nine regions from six countries, while the RCP4.5 scenario forecasts this result for one region from Italy and France. However, future scenarios by 2050 suggest that for around 80% of the European regions, their corresponding aggregated CF will increase by 2.5% to 85% (Figure 6.6). The RCP4.5 scenario predicts that 56 out of 87 European regions will exhibit increases in aggregated CFs by 2.5% to 50%. Followed by the RCP8.5 scenario that projects the same output for 59 regions, while the RCP2.6 scenario predicts this result for 56 regions. In particular, the RCP4.5 scenario forecasts the greatest increases in aggregated CFs in the range of 20% to 30%. Finally, on average across future scenarios, for 12% of European regions, aggregated CFs are projected to increase by 50% to 85% (Figure 6.6).

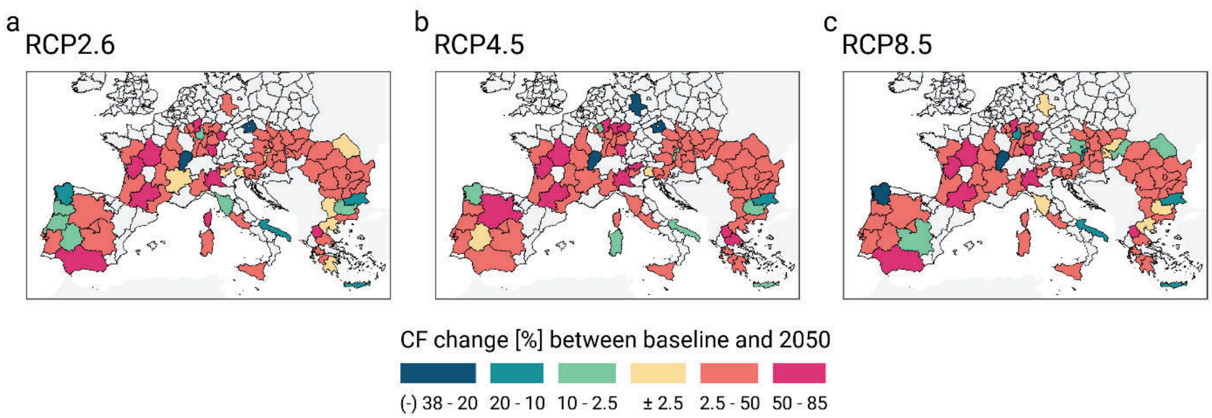


Figure 6.6 Relative change (%) in CFs aggregated at the level of European regions compared to the baseline scenario for RCP2.6 (a), RCP4.5 (b), and RCP8.5 (c) scenarios. The reader is invited to consult the interactive version of this figure at: https://iviveros.github.io/viveros-santos_et_al_2022_jclp/#figure-6

Aggregating CFs at the country level considerably shrank the spatial variability of CFs. While the spatial variability of CFs at the level of wine-growing regions is 1.96 orders of magnitude, aggregated CFs at the country level span over 0.46 orders of magnitude between the minimum and the maximum value. The mean value of aggregated CFs at the country level is 1.30 x 10^3 PAF·m^3·day·kg^{-1}, which is approximately 11% lower than that for CFs at wine-growing regions resolution (Figure 6.7). As shown in Figure 6.7a, 5 out of 14 countries have an aggregated CF higher than the average aggregated CFs as determined under the baseline scenario. More specifically, Germany and Austria have the highest aggregated CFs, with values of 2.36 x 10^3 and 2.16 x 10^3 PAF·m^3·day·kg^{-1}, respectively. This is explained by the high share of vineyard surface of CFs at wine-growing regions resolution in the range of 3.43 to 4.25 orders of magnitude for these countries (around 20%) (Figure 6.1a), which is also shown by the distribution of CFs factors at wine-growing regions resolution shifted to higher values (Figure 6.2). Likewise, Portugal, Slovenia, and Romania have aggregated CFs higher than the mean value and are at the same order of magnitude, with values of 1.61 x 10^3, 1.48 x 10^3, and 1.40 x 10^3 PAF·m^3·day·kg^{-1}, respectively. In fact, in terms of area fraction, most of the CFs at wine-growing regions resolution for Portugal, Slovenia, and Romania fall in the range of 2.97 to 3.29 orders of magnitude, with corresponding shares of vineyard surface of 70%, 43%, and 42%, respectively. In contrast, Italy, Greece, and Czechia have the lowest aggregated CFs with values of 8.04 x 10^2, 8.31 x 10^2, and 8.44 x 10^2 PAF·m^3·day·kg^{-1}, respectively. This corresponds to the high share of the land area of CFs in the range of 2.09 to 2.82 orders of magnitude for these countries, which is 67%, 60%, and 53% for Italy, Greece, and Czechia. Aggregated CFs for Hungary, Bulgaria, Slovakia, France, Spain, and Luxembourg have aggregated CFs in the range of 3 orders of magnitude and are accordingly the mean aggregated CF (Figure 6.7a).

For most countries, future scenarios point to increases in CFs aggregated at the country level. As was the case for the aggregation of CFs at the level of European regions, aggregation at the country level reduced the range of changes in CFs, but the latter operation had a higher influence. On average across the scenarios, the highest increase in aggregated CFs at the country level is predicted by the RCP4.5 scenario with a mean increase of 21%. The RCP2.6 scenario forecast an average increase of 19%, whereas the RCP8.5 scenario predicts a mean increase of 16% in aggregated CFs at the country level (Figure 6.7). Under the RCP4.5 scenario, the higher increases in aggregated CFs are projected for Czechia, Portugal, Slovenia, Slovakia, and Italy, with a mean increase of 33%, while the lowest increases are projected for Hungary, Greece, Bulgaria, Luxembourg, and Austria with an average increase of 11%. One exception to the increases in aggregated CFs at the country level is expected for Portugal under the RCP2.6 scenario, with a decline of 3% compared to the current scenario. The reason is that the RCP2.6 scenario projects an increase in OM in a greater surface over Portugal in comparison to other scenarios, which leads to lower partitioning coefficients, and consequently to lower FFs and CFs (Figure 6.1and Fig. C4). The RCP8.5 scenario also forecast a reduction in the aggregated CF for Austria of 8% related to the one determined according to the baseline scenario.

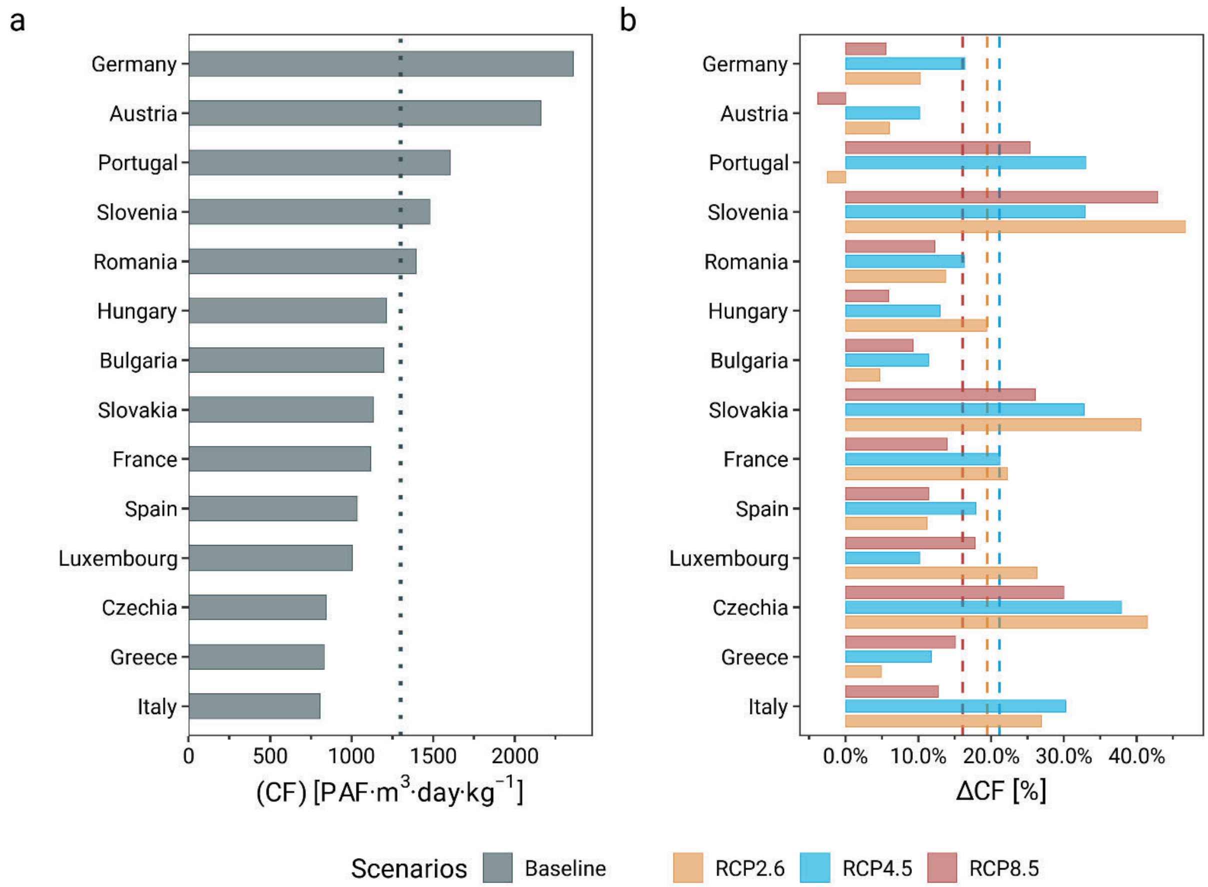


Figure 6.7 Area-weighted average of CFs (PAF·m^3·day·kg^{-1}) at the country level for the baseline period (a), and mean change in CFs for each European country by 2050 compared to the baseline period. The dotted vertical line in (a) indicates the mean area-weighted CF at the country level, whereas the dashed vertical lines in (b) represent the mean change (%) in CFs under RCP scenarios.

6.2.4.4 Limitations and future perspectives

One main limitation of this study corresponds to the restricted application of the developed CFs to non-calcareous vineyard soils, while an important area of European vineyards is settled on calcareous soils. This limitation resulted from the use of TBLMs for computing EFs. While these models permitted to account for metal speciation, and bioavailability, they are only valid for non-calcareous soils (Thakali et al., 2006). Besides, further research is needed on how to account for changing temperature in the computation of EFs. Currently, studies suggest that changing environmental conditions affect the performance of terrestrial organisms, but it is not straightforward to isolate their impact on metal speciation from the impact on species sensitivity. Thus, the reported level of effect on organisms' performance results from the combination of several stress factors, namely increasing temperature, and changes in moisture content (Fu et al., 2018; González-Alcaraz & van Gestel, 2016). Accordingly, in this study, the EFs were assumed constant, to avoid a potential double counting. Still, the influence of changing parameters on the extent of copper effects was introduced via the BF to some degree. Future research may consider a multifactorial approach to account for the combined effect of multiple stressors on soil organisms (Zandalinas et al., 2021).

There are also some uncertainties related to the parameters assumed constant under future scenarios. For instance, under episodes of heavy rainfall, pH may be reduced because of the leaching of basic cations. However, it is not feasible to account for those extreme events in the computation of FFs with USEtox. Still, the rates of changes in pH are slower in comparison to other environmental compartments, namely freshwater and the ocean, owing to the buffer effect of soil minerals (Biswas et al., 2018), which justifies in part assuming a constant pH for future scenarios. A second effect not accounted for in the characterization modelling is the effect of precipitation on DOC, which can be reduced after episodes of increased rainfall due to the increases in its leaching rate. Land-use changes were not integrated into the computation of copper fate in soils, but its influence on copper mobility has been reported and identified as research need to avoid problems related to the mobilization of copper in cases of reconversion of soil use that leads to more acid soil, such as the conversion of vineyards to forests (P Villanueva-Rey et al., 2019). The mobilization of copper driven by the acidification of soils would increase the lixiviation of metal, thus potentially decreasing the associated terrestrial ecotoxicity impacts, but resulting in increases in aquatic ecotoxicity impacts.

6.2.5 Conclusions

The results of this study highlight the pertinence of considering spatial and temporal aspects in the characterization of metals terrestrial ecotoxicity in LCA, as well as the interaction between impact categories. Despite the inherent uncertainties of future projections, temporal variations of soil organic matter, soil erosion, and precipitation were shown to either increase or decrease CFs. Nevertheless, under the future scenarios by 2050, increases in CFs are projected over a larger share of the vineyard surface, which will lead to rises in the median CFs in the order of 27% under RCP4.5 compared to that for the current scenario. However, future research may address extending the range of applicability of the characterization modelling to calcareous soils, since an important fraction of vineyards is established in calcareous soils. Furthermore, more research is needed on how to account for projected changes in temperature on species sensitivity to integrate it into the computation of EFs.

Finally, this study may contribute to the growing interest in performing prospective LCAs of product systems which so far focused mainly on projections at the life cycle inventory level. However, due to the projected changes in environmental conditions associated with climate change, the validity of applying current CFs decreases when using them in the assessment of prospective scenarios. In other words, the development of future CFs will allow performing prospective LCA studies that integrate the temporal dimension at both the inventory and the characterization modelling.

CHAPITRE 7 DISCUSSION GENERALE

Ce chapitre de la thèse commence par une synthèse des contributions du projet doctoral à la recherche scientifique. Ainsi, cette section souligne la manière dans laquelle les objectifs spécifiques émis dans le Chapitre 3 ont été accomplis. Subséquemment, les limites de ces apports scientifiques sont présentées.

7.1 Atteintes des objectifs de recherche

L'objectif général de cette thèse : *« développer une approche d'analyse du cycle de vie prospective permettant d'inclure l'influence du changement climatique à la fois sur l'inventaire du cycle de vie et sur l'évaluation des impacts écotoxiques terrestres du cuivre »* a été accompli au moyen des développements méthodologiques visant à atteindre trois objectifs spécifiques.

La contribution présentée au Chapitre 4 a répondu au premier objectif spécifique de cette thèse : « *évaluer la variabilité spatiale de l'écotoxicité terrestre des fongicides à base de cuivre appliqués en viticulture »*. En effet, la variabilité spatiale exerce une influence prépondérante sur des systèmes sensibles à la variabilité climatique, ainsi qu'à d'autres conditions de l'environnement. C'est le cas des systèmes agricoles, dont les pratiques agricoles varient non seulement en fonction du type de culture, mais aussi en fonction de leur localisation dans l'espace. Cette variabilité est transmise en conséquence aux inventaires du cycle de vie de ces systèmes. En outre, la variabilité spatiale affecte aussi l'ampleur de l'impact entraîné par les polluants. Ainsi, cet objectif spécifique a analysé à la fois la variabilité spatiale associée à l'utilisation des fongicides à base de cuivre, ainsi que celle associée à l'évaluation de l'écotoxicité terrestre causée par le cuivre.

D'abord, des facteurs de caractérisation pour l'évaluation de l'écotoxicité terrestre du cuivre tenant compte de la spéciation du métal selon les propriétés physicochimiques des sols ont été calculés. Il a été montré la pertinence d'inclure le processus de spéciation dans l'évaluation de l'écotoxicité terrestre du cuivre, car le facteur de caractérisation générique obtenu pour l'ensemble des sols non calcaires de la planète est d'environ 3,5 ordres de grandeur inférieurs en comparaison du facteur de caractérisation de la méthode IMPACT 2002+ qui néglige l'influence de la spéciation dans la modélisation de l'écotoxicité terrestre. De plus, les facteurs de caractérisation calculés avec l'approche utilisant le modèle de spéciation géochimique WHAM 6 s'étendent sur 5,5 ordres de grandeur pour l'ensemble des sols non calcaires du monde, ce qui confirme encore une fois la nécessité de tenir compte de la variabilité spatiale des propriétés physico-chimiques des sols dans l'évaluation de cette catégorie d'impact.

De plus, l'utilisation des facteurs de caractérisation dans une étude de cas portant sur quatre régions viticoles des principaux pays fournisseurs du vin consommé au Québec a permis d'évaluer l'influence de la variabilité spatiale associée à l'inventaire du cycle de vie de la production de raisins. Concernant le cas à l'étude, les résultats ont montré que la variabilité de la quantité d'application de fongicides à base de cuivre a une incidence importante sur le score d'impact, car le classement du score d'impact suit l'ordre établi par la quantité de fongicide utilisée. Ceci a montré donc la pertinence de tenir compte de l'aspect spatial au niveau de l'inventaire. Finalement, d'un point de vue pratique, les facteurs de caractérisations développés pour l'écotoxicité terrestre du cuivre ont été rendus disponibles dans un format compatible avec des logiciels de GIS, ainsi qu'avec des logiciels d'ACV qui permettent l'importation des cartes pour la réalisation des ACV régionalisées. Le manuscrit donne plus de détails sur l'accès aux facteurs de caractérisation, ainsi que sur leur utilisation.

Le Chapitre 5 intègre à la fois quelques dimensions spatiales et temporelles en ACV. Le développement méthodologique présentée dans ce chapitre permet d'atteindre le deuxième objectif spécifique de cette thèse : *développer une approche d'analyse du cycle de vie prospective tenant compte de l'influence du changement climatique sur l'inventaire des pratiques en viticulture et sur les stratégies d'adaptation.* En effet, les aspects spatiaux considérés dans ce développement méthodologique correspondent à la définition des inventaires du cycle de vie régionalisés qui tiennent compte des pratiques propres à chaque vignoble du cas à l'étude. En outre, l'aspect spatial a été considéré également dans le calcul des émissions issues des applications de produits phytosanitaires et des engrais, car les conditions environnementales affectent la distribution primaire de ces émissions. De plus, la dimension spatiale est prise en compte lors de la projection des impacts du changement climatique directs sur la production en viticulture, ainsi que ceux entrainés par des événements extrêmes.

L'aspect temporel de l'approche mise en place dans ce chapitre porte sur la définition de scénarios prospectifs en viticulture pour lesquels le changement climatique constitue un enjeu en raison de ses impacts potentiels sur la quantité et la qualité de la récolte. L'influence du changement climatique a été incluse à deux niveaux dans la définition des scénarios prospectifs pour deux vignobles se trouvant dans deux régions viticoles en France (Val de Loire et Languedoc-Roussillon). D'abord, les impacts du changement climatique sur le rendement des vignobles ont été inclus dans l'analyse selon deux scénarios d'émissions de GES et quatre périodes. Il a été montré que pour les régions analysées, les impacts des variations climatiques donnent lieu à des conclusions opposées quant aux impacts environnementaux projetés des vignobles étudiés. Par exemple, selon le scénario à fortes émissions de GES (SSP5-8.5), l'empreinte carbone (kg CO_2eq·kg raisins^{-1}) du vignoble en Languedoc-Roussillon pourrait augmenter d'environ 29% d'ici la fin du siècle, tandis que celle du vignoble en Val de Loire diminuerait d'environ 10%. Pour sa part, le score d'impact pour la catégorie qualité des écosystèmes augmenterait d'environ 33% en Languedoc-Roussillon et diminuerait d'environ 9% en Val de Loire pour le même scénario et horizon de temps. En effet, des études précédentes ont projeté que les impacts du changement climatique ne seront pas homogènes et que bien qu'il cause principalement des effets adverses, il y aura des régions qui en bénéficieront des changements projetés des variables climatiques, car les hausses de température signifieront dans quelques endroits un rapprochement vers la température de croissance optimale.

Néanmoins, dans le deuxième niveau d'analyse de cet objectif, c'est-à-dire, lorsque les impacts des événements extrêmes sont pris en considération dans l'analyse, les conclusions pointent vers la même direction pour les vignobles du cas à l'étude : les impacts environnementaux de la viticulture augmentent de manière importante en raison des impacts entrainés par les événements extrêmes à la fois sur le rendement et sur l'adoption des stratégies d'adaptation contre des extrêmes climatiques. Ainsi, selon le scénario SSP5-8.5, l'empreinte carbone du vignoble en Languedoc-Roussillon serait quatre fois l'empreinte actuelle, tandis qu'elle serait multipliée par un facteur trois dans le cas du vignoble en Val de Loire. Le développement méthodologique a montré donc la pertinence d'inclure non seulement l'impact direct du changement climatique sur le rendement, mais aussi celui engendré par les événements extrêmes.

Le développement méthodologique proposée au Chapitre 5 a permis de considérer la dimension temporelle en ACV à travers de l'évaluation de scénarios prospectifs qui tiennent compte de l'impact du changement climatique sur le rendement et sur les modifications des pratiques agricoles selon l'adoption de stratégies d'adaptation au changement climatique. Pour sa part, le Chapitre 4 a permis d'inclure la dimension spatiale en ÉICV, et plus particulièrement dans l'évaluation de l'écotoxicité terrestre du cuivre. Or, dans le contexte du changement climatique, il est projeté que l'ampleur de l'impact des polluants sera affectée en raison de l'altération des conditions environnementales futures; en conséquence, l'application des facteurs de caractérisation dérivés pour des conditions environnementales actuelles à des scénarios prospectifs est remise en question. Ainsi, le développement méthodologique du Chapitre 5 reprend ceux du chapitre 4, et va un pas plus loin en développant des facteurs de caractérisation futurs pour répondre à l'hypothèse de recherche.

La méthodologie développée dans le troisième article permet de considérer à la fois des aspects spatiaux et certains aspects temporels (ceux reliés à l'influence du changement climatique) en ÈICV, et elle permet de réaliser le troisième objectif spécifique : « développer *des facteurs de caractérisation futurs pour l'écotoxicité terrestre du cuivre intégrant l'influence du changement climatique* ».

Ce chapitre montre que bien que la variabilité spatiale des facteurs de caractérisation pour l'écotoxicité terrestre soit plus importante (1,96 ordre de grandeur) que la variabilité temporelle (celle résultant de la comparaison de facteurs de caractérisation présents et futurs) qui est de seulement 0,78 ordre de grandeur.

On peut également observer que l'omission de l'influence du changement climatique sur l'évaluation de l'écotoxicité terrestre représenterais, de manière générale, une sous-estimation de ces impacts dans l'évaluation de scénarios prospectifs pour la plupart des régions viticoles en Europe. Ainsi, selon le scénario RCP8.5, les facteurs de caractérisation pour l'écotoxicité terrestre des vignobles analysés dans le second article augmenteraient de 60% et 7% respectivement dans les vignobles en Val de Loire et Languedoc-Roussillon. Compte tenu des projections d'augmentation de la quantité de fongicides à base de cuivre utilisée à l'horizon 2050 de 33.5% et 12.5%, respectivement dans ces deux vignobles, si l'on combine les inventaires prospectifs de l'article 2 et les facteurs de caractérisation prospectifs de l'article 3, cela entrainerait une augmentation des impacts écotoxiques terrestres de 113.5% et 20% respectivement par rapport à un scénario qui ne tiendrait compte de l'influence des changements climatiques ni sur les pratiques agricoles ni sur l'écotoxicité terrestre du cuivre. On réalise ici que l'inventaire prospectif et la modélisation prospective des impacts ont une importance du même ordre de grandeur pour convenablement modéliser les impacts écotoxiques de la production viticole à l'horizon 2050 et il serait donc également important de tenir compte des deux phénomènes pour convenablement prédire l'impact futur de la production viticole.

De plus, le Chapitre 6 démontre la faisabilité et la pertinence de considérer l'interaction des catégories d'impact en ÉICV. En effet, la pratique courante en ÉICV est de modéliser chaque catégorie d'impact selon une chaine de cause à effet qui ne tient pas compte des interactions possibles avec d'autres catégories d'impact. Donc, ce chapitre a proposé un cadre méthodologique pour intégrer l'incidence du changement climatique sur les facteurs intermédiaires composant les facteurs de caractérisation pour l'écotoxicité terrestre. Plus spécifiquement, l'utilisation d'une approche basée sur des régressions empiriques pour modéliser la spéciation du métal a permis d'inclure l'influence des projections de la teneur en matière organique des sols, de l'érosion du sol, ainsi que des régimes de précipitations futurs dans le calcul des facteurs de devenir, d'accessibilité et de biodisponibilité.

Les résultats présentés aux Chapitres 5 et 6 ont permis de confirmer l'hypothèse de recherche de cette thèse, selon laquelle : « *Négliger l'influence du changement climatique sur les pratiques agricoles et sur l'évaluation de l'écotoxicité terrestre de cuivre dans l'analyse du cycle de vie prospective de la viticulture entraine une sous-estimation des impacts environnementaux potentiels* ». Tout de même, il y a quelques limites associées aux développements présentés dans cette thèse, lesquelles sont présentées dans la section suivante.

7.2 Limites de la thèse

7.2.1 Limites méthodologiques

Tout d'abord, les facteurs de caractérisation pour l'écotoxicité terrestre du cuivre obtenus avec les deux méthodes, soit avec celle basée sur des régressions empiriques ou celle basée sur l'utilisation du modèle de spéciation géochimique WHAM 6.0, comportent une limite commune : ces facteurs de caractérisation sont uniquement valables pour les sols non calcaires. Cette limite découle du choix méthodologique d'employer les modèles TBLM pour calculer des valeurs de EC_{50}, pour ensuite dériver des facteurs d'effet. Les modèles TBLM permettent de calculer des EC_{50} tenant compte de la spéciation du métal, de plus, dans le cas du cuivre, la disponibilité des modèles satisfait les conditions de représentativité recommandée par la méthode AMI pour le calcul des facteurs d'effet. Or, une fraction importante du vignoble mondial se trouve sur des sols calcaires.

Par conséquent, le manque de facteurs de caractérisation pour ce dernier type de sols constitue une limite importante de cette étude. Une autre limite des facteurs de caractérisation correspond à la non-prise en considération de l'essentialité des métaux. Néanmoins, cela nécessiterait plus des résultats de laboratoire sur la sensibilité d'un ensemble plus large d'organismes terrestres en fonction de leur distribution spatiale.

En ce qui concerne les facteurs de caractérisation futurs pour l'écotoxicité terrestre, la méthode développée pour intégrer l'influence du changement climatique a montré la faisabilité et la pertinence de considérer ce type d'interaction entre catégories d'impacts en ÉICV. Cependant, il y a des limites associées avec cette méthode. Avant tout, l'influence du changement climatique sur le facteur d'effet n'a pas été considérée dans la paramétrisation des facteurs de caractérisation afin d'éviter un potentiel double comptage. En effet, les études portant sur des tests *in vivo* simulant des conditions altérées par le changement climatique concluent que les impacts observés des métaux sous ces conditions sont le résultat de l'effet combiné des modifications de la spéciation des métaux, ainsi que des modifications de la sensibilité des espèces. Par conséquent, le choix méthodologique de cette thèse a été de ne pas considérer un pourcentage d'augmentation du niveau d'effet par changement de variable (par °C, par exemple), car les résultats rapportés dans la littérature ne permettent pas de départager la contribution des changements dans la température de celle des changements associés à des modifications de la spéciation. Or, il convient de préciser que ce choix méthodologique donne lieu à une sous-estimation potentielle du facteur d'effet, laquelle se propage donc dans les facteurs de caractérisation futurs pour l'écotoxicité terrestre du cuivre.

Une troisième limite de la thèse est en lien avec le calcul des facteurs de caractérisations futurs pour l'écotoxicité terrestre. En effet, quelques propriétés physicochimiques des sols influant sur la spéciation des métaux ont été considérées constantes sous des scénarios prospectifs. C'est le cas en particulier du pH qui peut être modifié par des périodes de forte précipitation, auquel cas les valeurs de pH sont réduites à cause de la lixiviation des bases cationiques. Cependant, le choix méthodologique de considérer le pH constant pour une cellule spatiale a été fait en raison du manque des projections du pH. Bien que les variations du pH affectent la mobilité du cuivre, la dynamique du pH dans les sols est lente en comparaison d'autres compartiments tels que l'eau de surface et l'océan. Une quatrième limite en lien avec les facteurs de caractérisation est la non-prise en compte des changements d'utilisation des sols, lesquels pourraient engendrer la mobilité des métaux dans le cas où le changement d'utilisation de sols donnerait lieu à des sols plus acides (par exemple, lors de la conversion d'un vignoble en forêt).

Une limite associée à la modélisation de l'impact du changement climatique sur le rendement des raisins dans des scénarios futurs est introduite par l'utilisation de la méthode statistique employée pour évaluer l'influence des changements des variables climatiques. En effet, la technique de dissociation statistique utilisée permet de calculer la variation du rendement par changement de variable climatique, mais elle ne permet pas de tenir compte d'autres facteurs qui affectent également le rendement de raisins comme la radiation solaire, des précipitations extrêmes, la gestion de la canopée et des systèmes de conduite de la vigne.

Une autre limite de ce projet de recherche est associée avec la modélisation de l'impact des événements extrêmes. En effet, les événements extrêmes ont été traités comme des événements indépendants et en conséquence l'impact total entrainé par ces événements a été calculé par une simple addition des impacts associés à chaque événement extrême. Or, des études ont rapporté que des impacts synergistes peuvent survenir, par exemple, lorsque des périodes de sècheresse et des ondes de chaleurs surviennent simultanément. Dans cette situation, les impacts calculés dans le cadre de cette thèse seraient potentiellement sous-estimés.

7.2.2 Limites sur les résultats

La prise en compte de l'influence du changement climatique à la fois sur le calcul des facteurs de caractérisation futurs pour l'écotoxicité terrestre et sur l'inventaire des pratiques en viticulture comporte une limite en commun, correspondant à l'incertitude inhérente à l'analyse prospective. En effet, il convient de rappeler qu'une étude d'impact du changement climatique comporte inévitablement une cascade d'incertitudes. D'abord, les modèles climatiques, bien que complexes, représentent une description simplifiée de la réalité. Ensuite, les scénarios d'émissions de GES utilisés pour projeter le climat futur sont basés sur une série d'hypothèses sur le développement démographique, économique et technologique. Puis, il y a aussi des incertitudes sur la conversion des émissions de GES en des concentrations atmosphériques en raison des incertitudes découlant de la modélisation du cycle du carbone. Subséquemment, ces scénarios d'émissions de GES alimentent des modèles climatiques qui posent des hypothèses physiques et font des approximations numériques lors de la projection du climat. Ensuite, les sorties des modèles climatiques sont régionalisées selon différentes méthodes de *réduction de la résolution,* lesquelles introduisent aussi des incertitudes en raison des approximations faites. Finalement, il y a des incertitudes liées aux modèles d'évaluation d'impact du changement climatique. Malgré les incertitudes inhérentes à l'analyse prospective, les résultats obtenus contribuent à la discussion scientifique sur les impacts du changement climatique. De plus, il est important de rappeler qu'un des objectifs principaux des modèles scientifiques est de contribuer à la compréhension ou à l'anticipation du futur.

Une autre limite sur l'ACV prospective de la viticulture est la gamme des options d'adaptation considérées. En effet, la méthode permet d'évaluer l'impact des stratégies d'adaptation technologiques, comme l'installation d'un système d'irrigation pour pallier la sécheresse, mais cette méthode ne permet pas de tenir compte des changements au niveau de la canopée ou de la couverture des sols qui pourraient atténuer les impacts de la rareté d'eau. Cependant, à l'heure actuelle, les effets de ces stratégies d'adaptation ne sont pas encore rapportés dans la littérature scientifique.

CHAPITRE 8 CONCLUSION ET RECOMMANDATIONS

Les principales contributions de ce projet doctoral ont permis de répondre à l'objectif principal de développer une approche d'analyse du cycle de vie prospective permettant d'inclure l'influence du changement climatique à la fois sur l'inventaire du cycle de vie et sur l'évaluation des impacts écotoxiques terrestres du cuivre. La méthode a été appliquée dans l'ACV prospective de la viticulture où le changement climatique influe à la fois sur l'inventaire et sur l'ampleur de l'impact des polluants dans des conditions environnementales futures projetées par des modèles climatiques. Ainsi, les principales contributions de cette thèse sont :

- Des facteurs de caractérisation pour l'écotoxicité terrestre du cuivre intégrant la spéciation du métal en fonction de la variabilité des propriétés physicochimiques des sols non calcaires du monde. Ceci contribue donc aux efforts d'amélioration de l'ÉICV grâce à la prise en compte de la dimension spatiale en ACV. De plus, l'application de ces facteurs de caractérisation a montré la nécessité de tenir compte de la régionalisation dans l'évaluation de l'impact causé par l'utilisation de fongicides à base de cuivre en viticulture.
- Une approche méthodologique permettant d'inclure l'influence du changement climatique sur le rendement et la mise en œuvre des stratégies d'adaptation dans des scénarios prospectifs en viticulture. Ceci a contribué à la discussion sur l'évaluation prospective des systèmes vulnérables aux conditions climatiques projetées par les scénarios d'émission du GIEC.
- Une méthode de caractérisation de l'écotoxicité terrestre intégrant l'influence du changement climatique. Cette méthode a montré la faisabilité et la pertinence de considérer l'interaction entre des catégories d'impact en ÉICV dans le cas particulier du calcul des facteurs de caractérisation futurs pour l'écotoxicité terrestre du cuivre.

Tout de même, comme décrit à la section 7.2, les approches méthodologiques développées dans ce projet de recherche comportent des limites liées aux choix méthodologiques et aux résultats. Ainsi, quelques recommandations sont émises à la section suivante.

8.1 Recommandations

Cette thèse a permis d'améliorer la prise en compte de quelques aspects spatiaux et temporels en ACV; plus particulièrement, grâce au développement des facteurs de caractérisation régionalisés pour l'évaluation de l'écotoxicité terrestre du cuivre. De plus, deux approches méthodologiques ont permis d'évaluer l'influence du changement climatique sur l'inventaire des pratiques en viticulture et sur l'impact futur des émissions de cuivre dans les sols viticoles. Tout de même, des limites ont été identifiées et rapportées à la section 7.2. Ces limites, en revanche, peuvent porter vers d'autres perspectives de recherche :

Le calcul des facteurs d'effet pour l'écotoxicité terrestre avec une méthode alternative

L'utilisation du modèle TBLM bien qu'elle permette de tenir compte de la spéciation des métaux et quelle satisfasse les critères de la méthode AMI pour le calcul des facteurs d'effet, elle est limitée à deux métaux (cuivre et nickel) et aux sols non calcaires. Ainsi, bien qu'en développement, l'utilisation des *voies menant à des effets nocifs* (AOP pour *Adverse Output Pathway*) est prometteuse pour l'évaluation des effets combinés du changement climatique et des facteurs de stress chimiques. De plus, cela permettrait de combler quelques lacunes de l'évaluation des impacts écotoxiques en ACV : la couverture limitée des substances, l'incertitude qui n'est pas rapportée, la sensibilité des espèces qui n'est pas prise en compte ; et le manque de considération de la distribution spatiale des espèces.

Le calcul des impacts du changement climatique sur la viticulture avec un modèle phénologique

Dans le but de réduire l'incertitude de l'impact du changement climatique sur le rendement de la vigne et sur l'adoption des stratégies d'adaptation, une solution de rechange à la méthode statistique pour projeter les impacts des changements climatiques sur le rendement, et à l'approche d'analyse de risque pour estimer la nécessité d'adopter certaines stratégies d'adaptation, est l'utilisation d'un modèle de culture, aussi appelé modèle phénologique, car il décrit les différents étapes de développement des cultures. Dans le cas de la vigne, le modèle STICS pourrait combler ce besoin, et il permettrait de tenir compte de l'influence des concentrations de CO_2 atmosphériques sur le rendement. En outre, il serait possible de coupler ce modèle avec des modèles de projections d'événements extrêmes afin de réduire l'incertitude des dommages causés par ces événements. Cependant, il faudra tenir en considération que les gains en matière de réduction d'incertitude impliqueront plus de ressources informatiques et du temps de calcul pour le déploiement de ce type de modèle.

La modélisation de l'interaction du changement climatique avec d'autres catégories d'impact

Étant donné l'intérêt grandissant envers la réalisation des ACV prospectives, et compte tenu des impacts du changement climatique sur les caractéristiques de l'environnement, qui à leur tour modifient le sort, biodisponibilité et niveau d'effet des polluants ; il serait intéressant de modéliser l'interaction du changement climatique avec d'autres catégories d'impact ou d'étendre la méthode développée dans cette thèse à d'autres substances. Cependant, des variations de température jouent un rôle déterminant dans la demi-vie des polluants organiques et dans ce cas il faudra y tenir compte pour modifier en conséquence l'approche proposée dans cette thèse.

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ANNEXE A INFORMATIONS SUPPLÉMENTAIRES POUR L'ARTICLE PRÉSENTÉ AU CHAPITRE 4

Table A.1 Ranges of calculated factors and comparison with those reported by Owsianiak et al. (2013).

Method:	**WHAM**			**Regression Models**			**Owsianiak *et al.* (2013)**		
Parameter\ Percentiles	2.5th	50th	97.5th	2.5th	50th	97.5th	2.5th	50th	97.5th
$\log_{10}(K_d)$ [L/kg]	0.48	2.6	2.99	2.64	3.1	3.34	2.48	2.96	3.40
$\log_{10}(FF_{soil,air})$ [day]	-	-	-	-	-	-	4.15	4.59	4.96
$\log_{10}(FF_{soil,soil})$ [day]	2.52	4.46	5.23	4.52	5.18	5.58	-	-	-
$\log_{10}(ACF)$ [$kg_{reactive}/kg_{total}$]	-	-	-	-0.41	-0.34	-0.25	-0.46	-0.38	-0.19
$\log_{10}(BF)$ [$kg_{free}/kg_{reactive}$]	-	-	-	-6.54	-5.34	-4.27	-6.77	-5.64	-4.32
$\log_{10}(BF)$ [kg_{free}/kg_{total}]	-9.43	-5.51	-3.75	-	-	-	-	-	-
$\log_{10}(EF)$ [$PAF.m^3/kg_{free}$]	4.01	4.88	5.6	4.01	4.87	5.6	3.81	4.64	4.89
$\log_{10}(CF)$ [$PAF.m^3.day/kg_{total}$]	-0.06	3.23	5.44	2.78	4.27	5.16	2.23	3.15	4.30
CF_{global} [$PAF.m^3.day/kg_{total}$]	32707.8			32696.4			-		
Database	HWSD			HWSD			ISRIC-WISE3		
Number of soil mapping units	7907			7841			760		

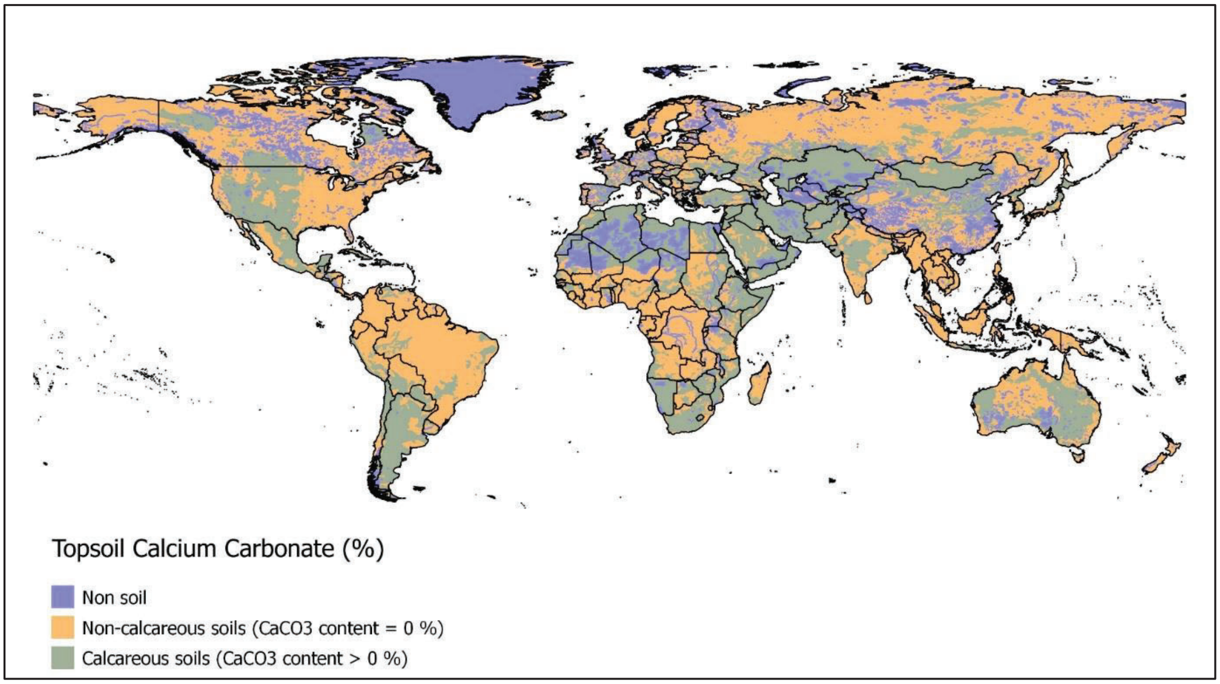

Figure A.1 Calcium carbonate content (%) in topsoil according to the HWSD (FAO/IIASA/ISRIC/ISS-CAS/JRC, 2012).

In the following figures, black dots correspond to 2.5th and 97.th percentiles of the values. Boxes and bars correspond to 5th, 25th, 50th, 75th, and 95th percentiles.

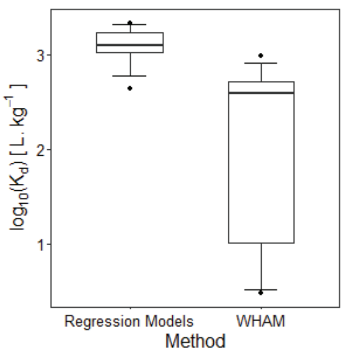

Figure A.2

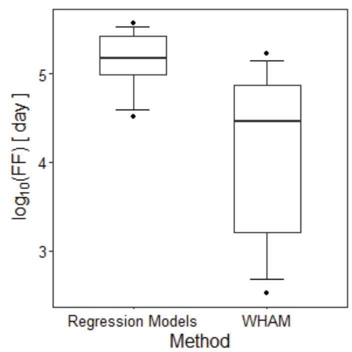

Figure A.3

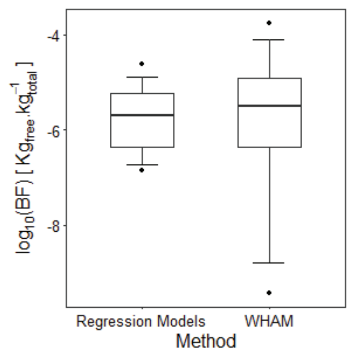

Figure A.4

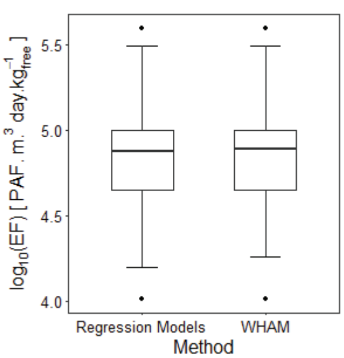

Figure A.5

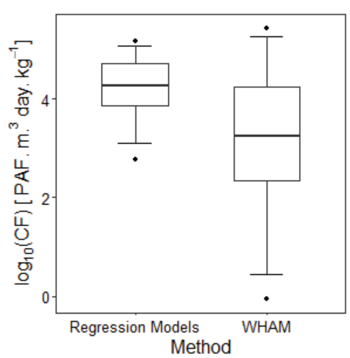

Figure A.6

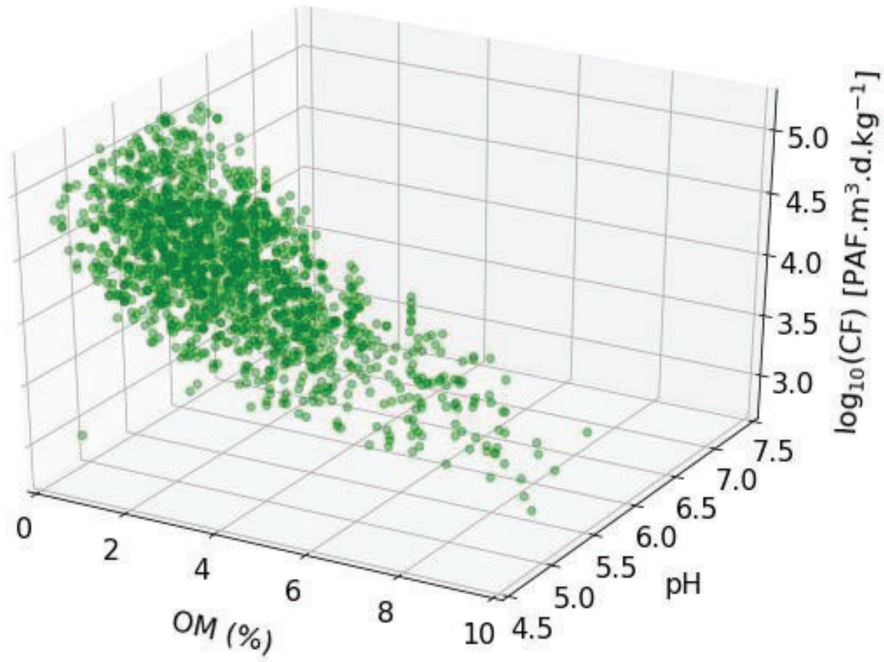

Figure A.7 CFs derived from Empirical Regression Models Method.

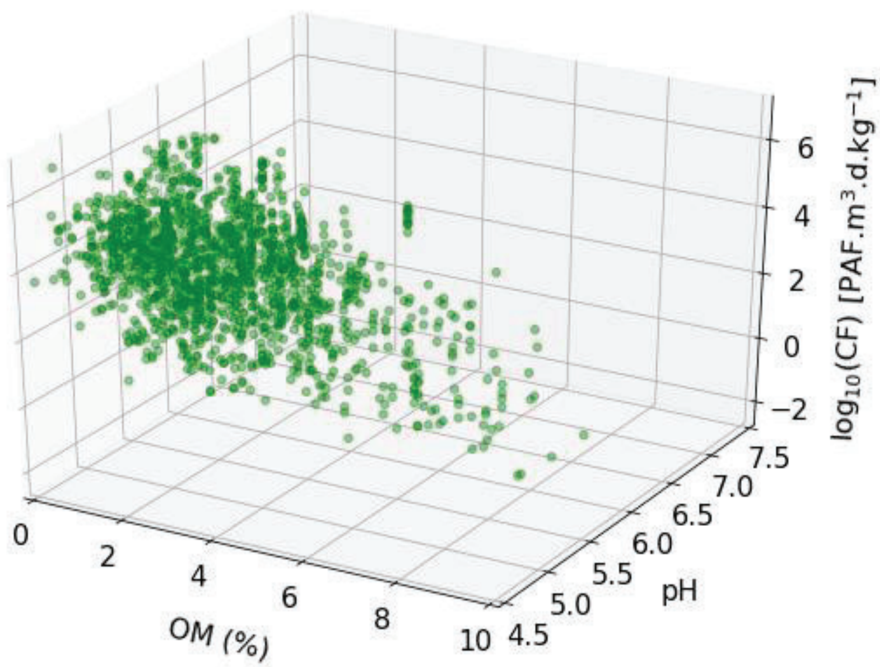

Figure A.8 CFs derived from WHAM 6.0 method.

ANNEXE B INFORMATIONS SUPPLÉMENTAIRES POUR L'ARTICLE PRÉSENTÉ AU CHAPITRE 5

B1 Study description

Table B.1 Main characteristics of the POTs implemented on the vineyards analyzed in this study.

Case study	**Loire Valley**	**Languedoc-Roussillon**
Yield (kg/ha)	7500	7829
Grape variety	Chenin Blanc	Syrah
Vintage	2011	2017
Pesticides applications		
Number of pesticide applications	5	19
Total amount of fungicides sprayed (kg/ha)	3	71.7
Amount of organic pesticides (kg/ha)	3	9.5
Amount of copper-based products (kg/ha)	0	5.4
Amount of sulfur-based products (kg/ha)	0	56.8
Total amount of insecticides sprayed (kg/ha)	0	3.9
Type of sprayer used	Pneumatic	Pneumatic
Fertilizer management		
Type of fertilizer used	N, P, K	N, P, K
Amount of fertilizer applied (kg/ha) (N, P, K)	(100, 58, 126)	(42, 13, 84)
Application method	Soil surface application	Soil surface application
Weed management		
Inter-row management	1 out of 2 grassed, 1 out of 2 mowed	1 out of 2 grassed, 1 out of 2 mechanically weeded
Row management	Chemical weeding	Mechanical weeding
Number of chemical weeding operations	1	1

Case study	Loire Valley	Languedoc-Roussillon
Total amount of herbicides sprayed (L/ha)	6	3
Number of mechanical weeding operations	0	3
Number of grass mowing operations	0	0
Canopy management		
Number of mechanical trimming operations	2	0
Harvest		
Mechanical or manual harvest?	Manual	Manual

In Loire Valley, the year 2011 was characterized as a dry and warm climate, which restricted the development of pest diseases and favoured the precocity of the harvest, that is, harvest occurred before the usual time (C Renaud-Gentié et al., 2014). Whereas in Languedoc-Roussillon, the year 2017 was characterized as a wet climate, which promoted high fungal disease pressure (namely powdery and downy mildew).

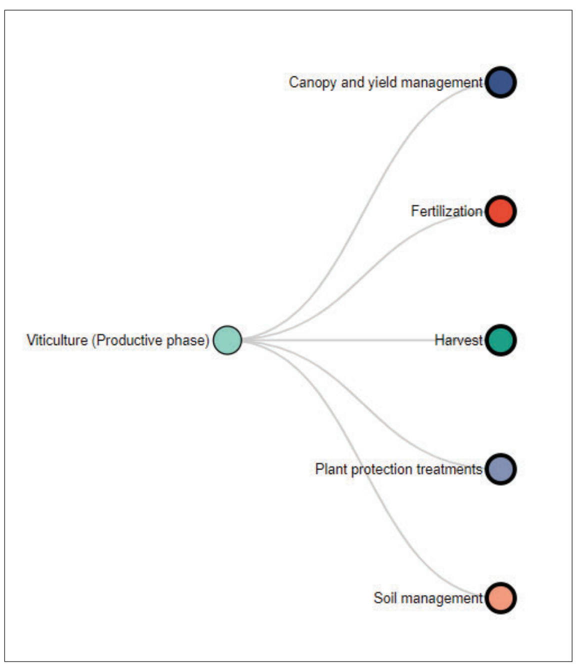

Figure B.1 Product system of the vineyard from Loire Valley.

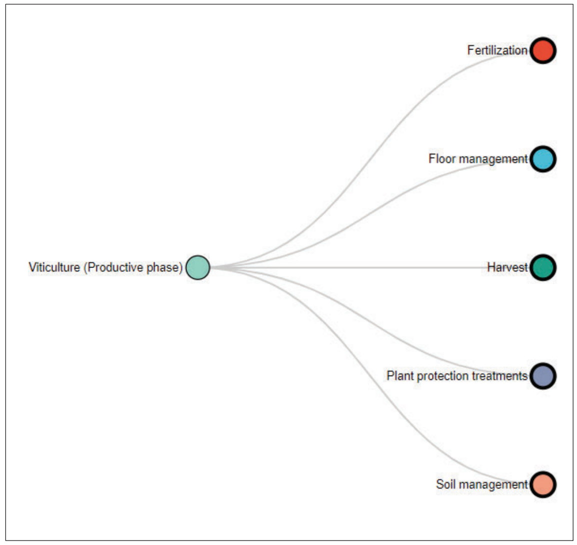

Figure B.2 Product system of the vineyard from Languedoc-Roussillon.

B2 Data sources

B2.1 Grape yield and vineyard surface

The database of viticultural yield obtained from the French Ministry of Agriculture and Food Statistics Service (Agreste, 2021) has 11 categorical columns and a numeric column ($valeur$) (Table B.2). The latter column gives the value for each indicator ($n027\_lib$) across the reported years ($annref$), the geographical resolution ($geographie$), and the categories of vines ($n304\_lib$), for a total of 36450 rows. The columns that have unique values were removed, since they do not give relevant information. Furthermore, the columns $geographie\_lib$, $n304\_mod$, and $n027\_mod$ were discarded since they correspond to the labels or codes of the columns $geographie$, $n304\_lib$, and $n027\_lib$, respectively.

Table B.2 Structure of the database of viticultural yield obtained from Agreste (2021).

Column	**Count**	**Unique values**
nom	1	SAA_VIGNE
annref	21	2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, 2010, 2011, 2012, 2013, 2014, 2015, 2016, 2017, 2018, 2019, 2020
geographie	121	01, 02, 03, 04, 05, 06, 07, 08, 09, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 21, 22, 23, 24, 25, 26, 27, 28, 29, 2A, 2B, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 971, 972, 973, 974, 976, DOM, FR, METRO, NR01, NR02, NR03, NR04, NR06, NR11, NR24, NR27, NR28, NR32, NR44, NR52, NR53, NR75, NR76, NR84, NR93, NR94
geographie_lib	121	Pattern: Code - Label
n304	1	Production de vins
n304_mod	3	1, 1.11, 1.12
n304_lib	3	Vigne à raisin de cuve, Vigne à raisin de table, Vignes et pépinières viticoles

Column	Count	Unique values
n027	1	Groupe d'indicateurs
n027_mod	5	020.01, 020.02.06, 050, 050.07.01, 070.01
n027_lib	5	Production (volume), Production pour le fruit (volume), Rendement, Superficie en production, Surface
qualite	1	OUI
valeur	NA	Continuous variable

Yield ($n027\_lib = Rendement$) of wine grapes ($n304\_lib = Vigne$ à $raisin\ de\ cuve$) for the period 2000-2021 of Departments of Metropolitan France were extracted from Agreste database of viticultural yield, which produced a subset of 1260 records (Table B.3). Column $geographie$ was filtered according to the polygon shapefiles of Metropolitan France and French Departments (Table B.4).

Table B.3 Filter criteria to subset the database of viticultural yield.

Column	Count	Unique values
annref	21	From 2000 to 2020 (inclusive)
geographie	60	01, 02, 03, 04, 05, 07, 10, 11, 13, 16, 17, 18, 21, 24, 26, 2A, 2B, 30, 31, 32, 33, 34, 36, 37, 38, 39, 40, 41, 42, 44, 45, 46, 47, 49, 51, 52, 54, 57, 58, 63, 64, 65, 66, 67, 68, 69D, 70, 71, 72, 73, 74, 77, 79, 81, 82, 83, 84, 85, 86, 89
n304_lib	1	Vigne à raisin de cuve
n027_lib	1	Rendement
valeur	NA	Continuous variable

For subsequent spatial analyses, French Vineyard area (Figure B.3-b) was extracted from the layer of European vineyard surface (Figure B.3-a) obtained from CORINE program (EEA, 2017b), subsequently, French vineyard surface was split by Departments of France (Figure B.3-d). GeoPandas Python module (Jordahl et al., 2020) was used to assist in these operations. In total, there is available data on grape yield for 60 Departments of Metropolitan France (Figure B.3).

Table B.4 Summary of spatial data sets used to relate vineyard surface and French Departments.

Descriptive name	Source
Vineyard area over Europe	European Environment Agency (2017b): https://www.eea.europa.eu/data-and-maps/data/corine-land-cover-2000-clc2000-seamless-vector-database-1/221-vineyards/221-vineyards
Metropolitan France	Global administrative area boundaries (2021): https://gadm.org/download_country.html
Departments of France	Open platform for French public data (2021): https://www.data.gouv.fr/en/datasets/contours-des-departements-francais-issus-d-openstreetmap/
Regions of France	Open platform for French public data (2021): https://www.data.gouv.fr/fr/datasets/contours-des-regions-francaises-sur-openstreetmap/

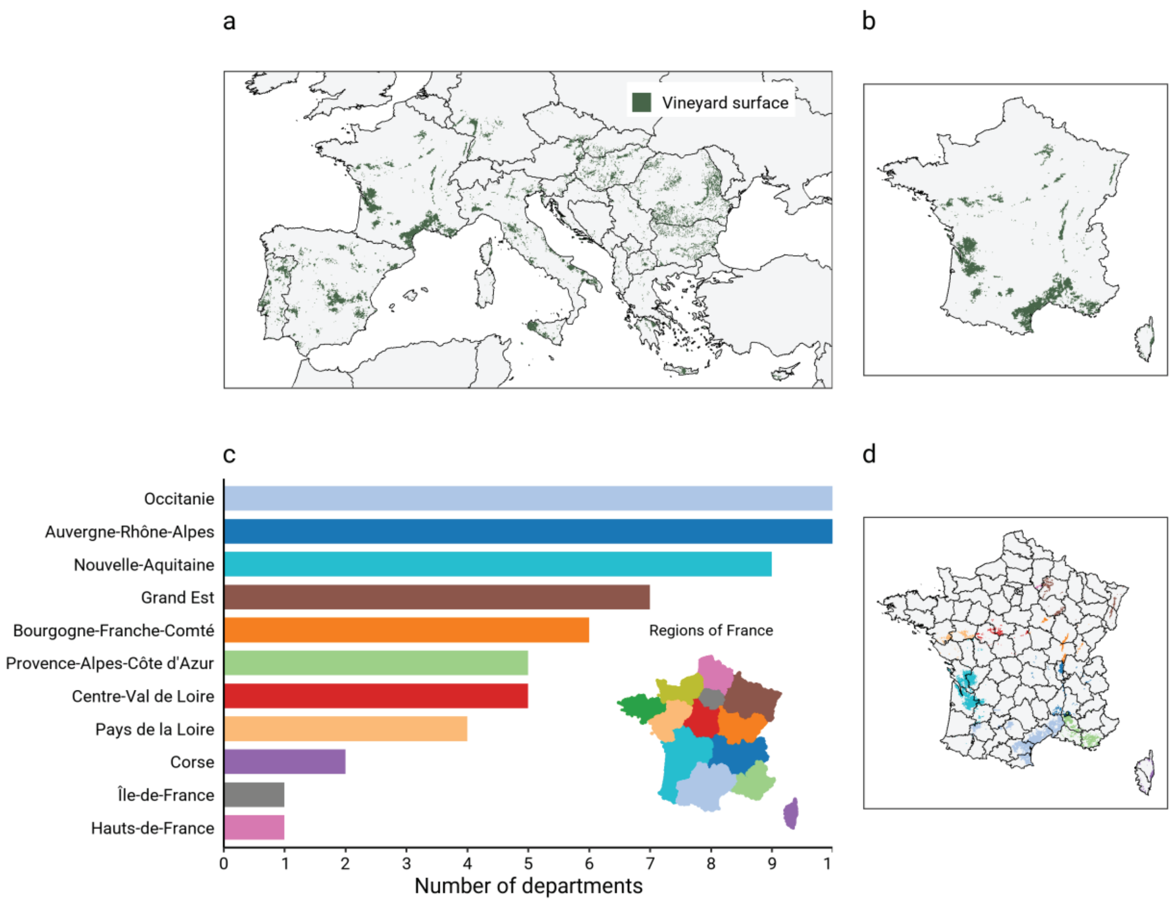

Figure B.3 European vineyard area according to CORINE land Cover data sets (EEA, 2017b) (a). French vineyard area (b). French vineyard area split by Department (coloured by Administrative

Region) (d). Number of French Departments that reported viticultural yield, by Regions of France (c) (Agreste, 2021).

B2.2 Climate data

For the investigation of the impacts of climate on grape yield, we used daily climate data from the Agri4Cast Resource portal (Biavetti et al., 2014). From the available data (Table B.5), we collected mean air temperature (°C) and total precipitation (mm/day) for the period 2000-2020. Figure B.4 shows the Agri4Cast grid points across France.

Table B.5 Description of the Agri4Cast data set (Biavetti et al., 2014).

Property	**Detail**
Version	3.1
Date published	15/01/2021
Publisher	Joint Research Centre European Commission https://agri4cast.jrc.ec.europa.eu/dataportal/
Grid Spatial Projection (Grid EPSG Code)	Lambert Azimuthal Equal Area (3035)
Grid Resolution	25 km
Period	From: 01/01/1979 - To: 31/12/2020
Time Resolution	1 day
Variables	maximum air temperature (°C), minimum air temperature (°C), mean air temperature (°C), mean daily wind speed at 10m (m/s), vapour pressure (hPa), total precipitation (mm/day), potential evapotranspiration from a crop canopy (mm/day), total global radiation (KJ/m^2/day)

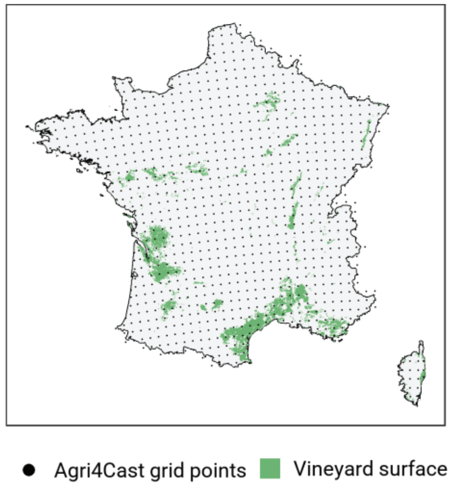

Figure B.4 French vineyard area according to CORINE land Cover data sets (EEA, 2017b) with the Agri4Cast grid points (Biavetti et al., 2014) used in the interpolation of the climate variables.

To compute projected change in average temperature and total precipitation during the growing season of wine grapes we used data from WorldClim. Table B.6 details the source and characteristics of historical and future climate data used to calculate projected climate change. Figure B.5 and Figure B.6 show respectively average temperature and total precipitation over the grape-growing season according to the baseline period (1970-2000), and future periods (2021-2100) driven by two Shared Socioeconomic Pathways (SSPs) (SSP1-2.6 and SSP5-8.5).

Table B.6 Sources and characteristics of climate data sets for computing change in average temperature and total precipitation during the growing season of wine grapes.

	Historical climate data	**Future climate data**
Source	Fick and Hijmans (2017) https://www.worldclim.org/data/worldclim21.html	Fick and Hijmans (2017) and Petrie et al. (2021) https://www.worldclim.org/data/cmip6/cmip6climate.html
GCM	NA	IPSL-CM6A-LR
Period	1970-2000	2021-2040 2041-2060 2061-2080

	Historical climate data	Future climate data
		2081-2100
Spatial resolution	2.5 minutes	2.5 minutes
Shared Socioeconomic Pathways (SSPs)	NA	SSP1-2.6 SSP5-8.5

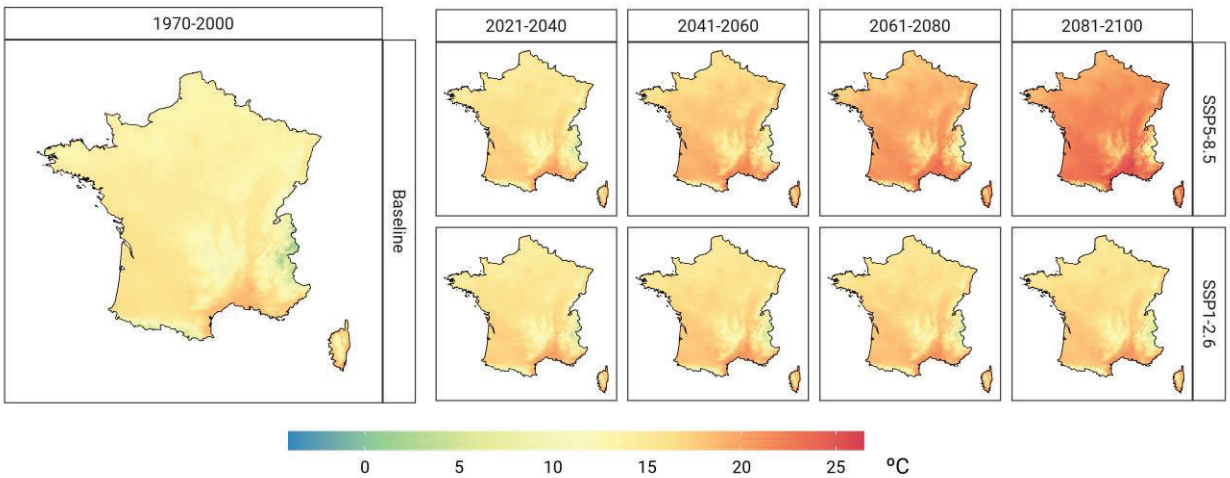

Figure B.5 Average temperature (°C) during the growing season of wine grapes. Source of data: Fick and Hijmans (2017).

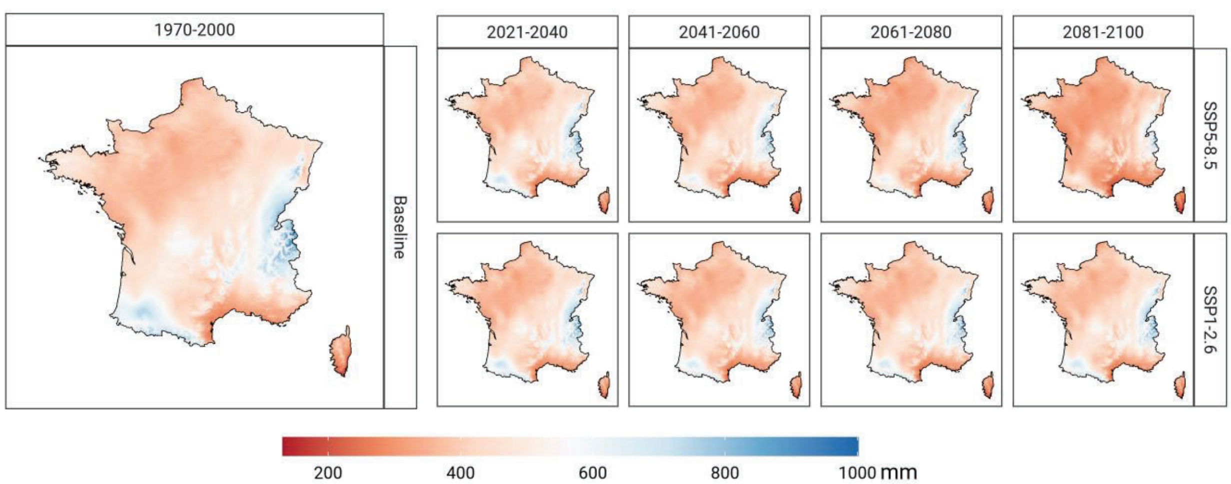

Figure B.6 Total precipitation (mm) during the growing season of wine grapes. Source of data: Fick and Hijmans (2017).

B2.3 Agroclimatic indicators

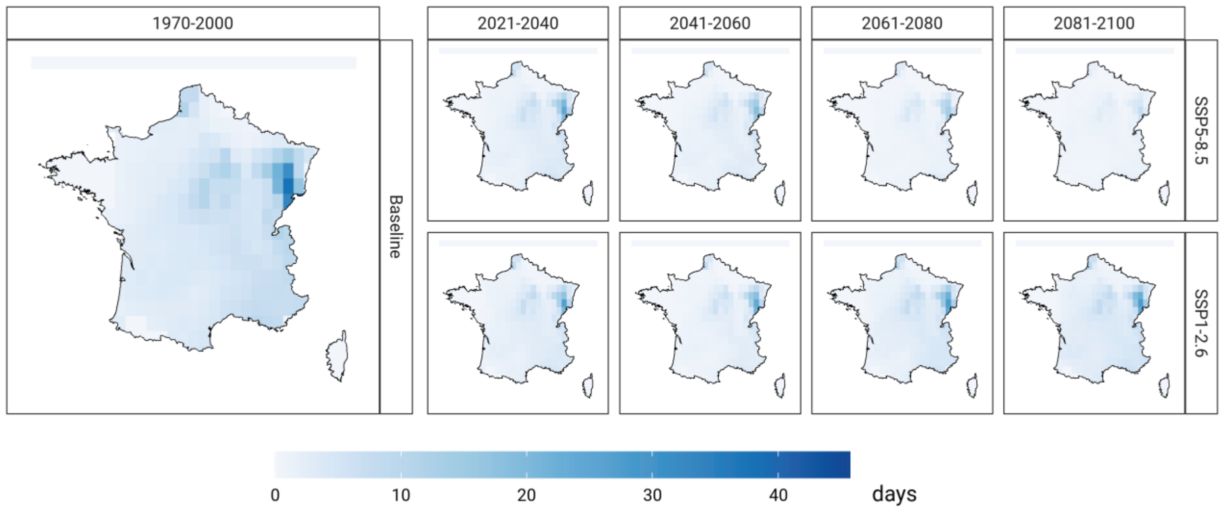

Figure B.7 Consecutive frost days (CFD, cold spell) during the spring season. Source of data: Copernicus (2021).

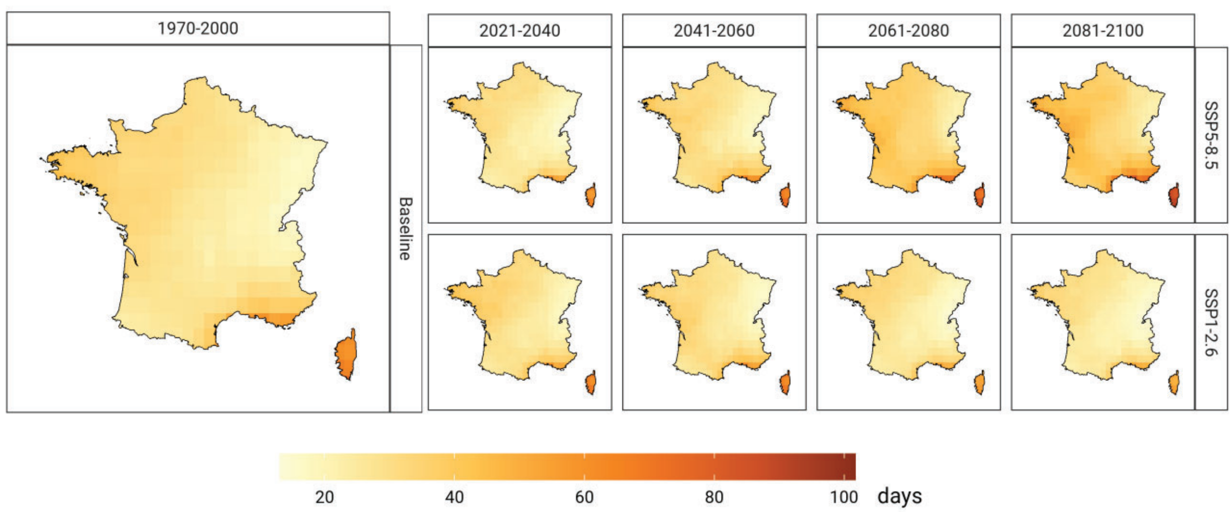

Figure B.8 Consecutive dry days (CDD) during the growing season of wine grapes. Source of data: Copernicus (2021).

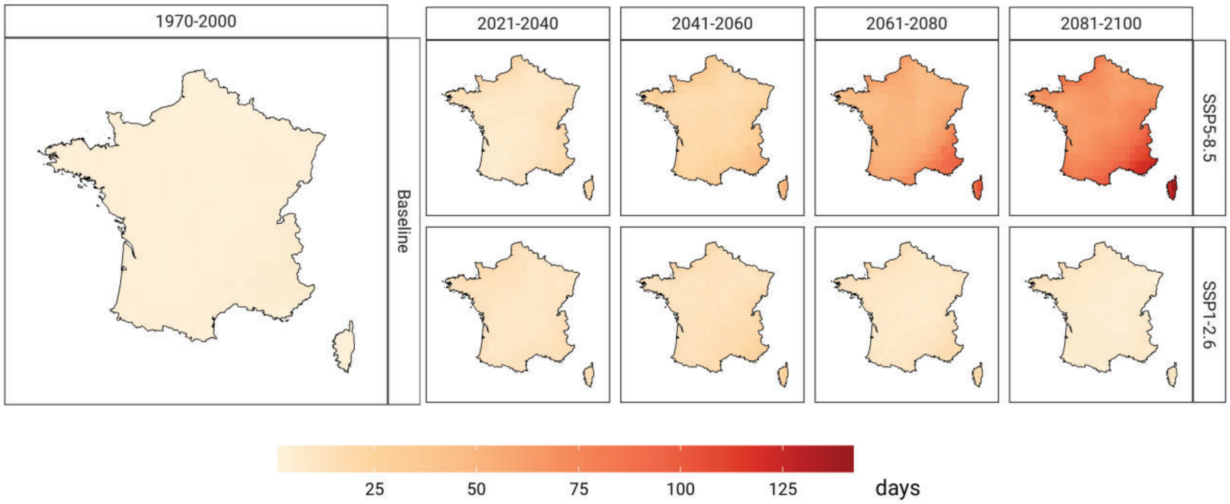

Figure B.9 Warm-spell duration index (WSDI) during the growing season of wine grapes. Source of data: Copernicus (2021).

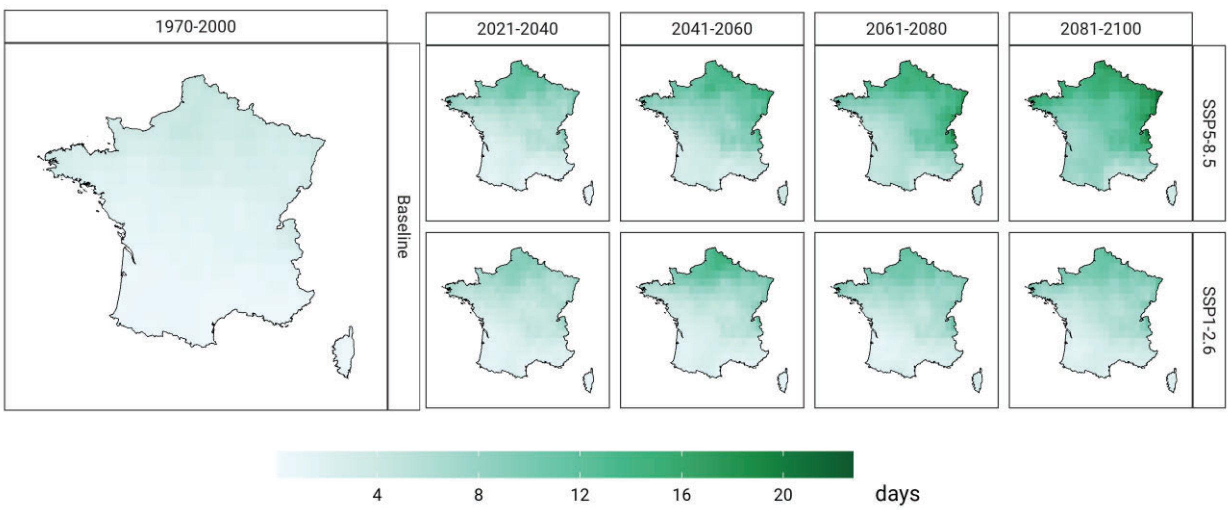

Figure B.10 Warm and wet days (WW) during the growing season of wine grapes. Source of data: Copernicus (2021).

B3 Methods and results not shown in the main document

B3.1 Historical climate

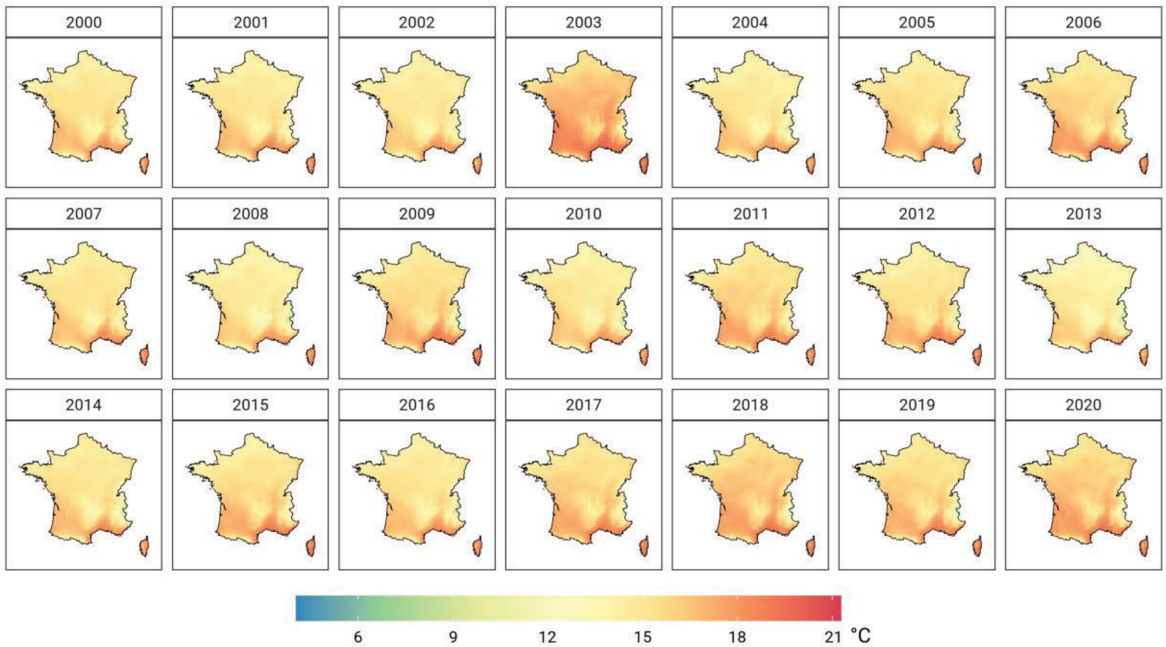

Figure B.11 Spatial distribution of average temperature (°C) during the growing season of wine grapes (2000-2020). Derived from Biavetti et al., (2014).

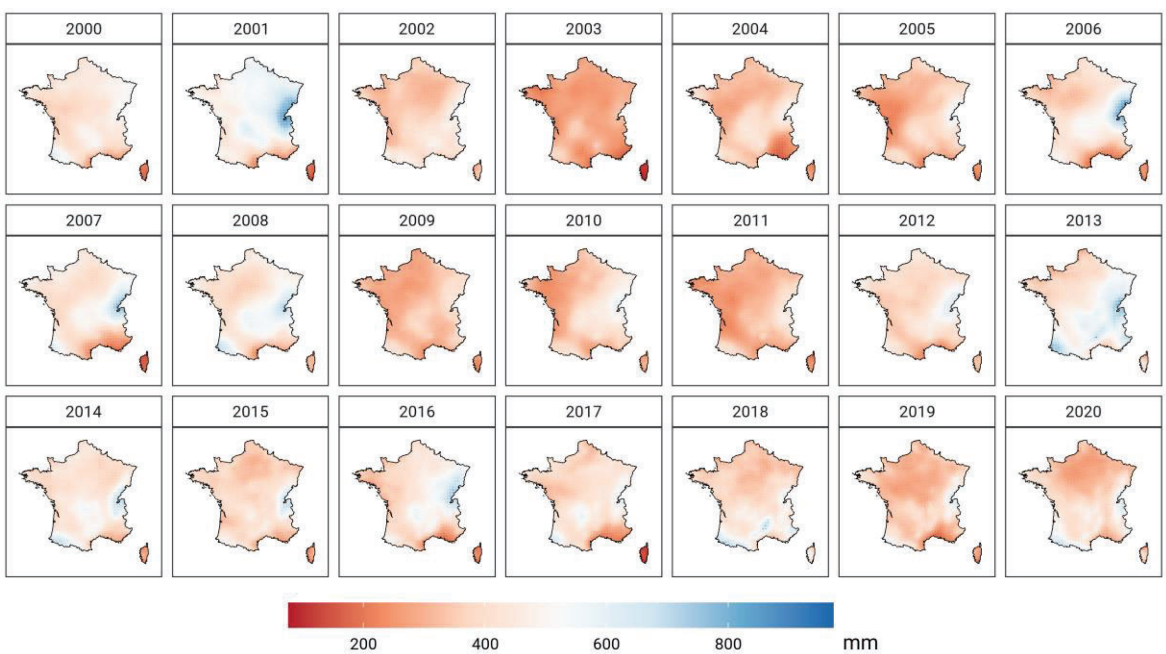

Figure B.12 Spatial distribution of total precipitation (mm) during the growing season of wine grapes (2000-2020). Derived from Biavetti et al., (2014).

B3.2 Climate-induced yield impacts

As described in the main text, to evaluate the sensitivity of grape yield to climate variables, namely the average temperature and total precipitation during the growing season, we applied a widespread method based on the first-difference time series for yield and climate (Kukal & Irmak, 2018; Lobell & Field, 2007). First, we computed the first differences of yield, mean temperature, and precipitation, that is, the difference in values from one year to the next. This resulted in residuals of yield (kg ha^{-1} yr^{-1}), average temperature (°C yr^{-1}) and total precipitation (mm yr^{-1}). Subsequently, we computed linear regressions with residuals of yield (kg ha^{-1} yr^{-1}) as the response variable, and the residuals of average temperature (°C yr^{-1}) and total precipitation (mm yr^{-1}) as explanatory variables (Figure B.13 and Figure B.14).

Figure B.13 Scatter plots of yield residuals (kg ha^{-1} year^{-1}) and residuals of average temperature (°C yr^{-1}) during the growing season of wine grapes, along with best-fit linear regression (black line) and standard error (gray shadow).

Figure B.14 Scatter plots of yield residuals (kg ha-1 year-1) and residuals of total precipitation (mm yr-1) during the growing season of wine grapes, along with best-fit linear regression (black line) and standard error (gray shadow).

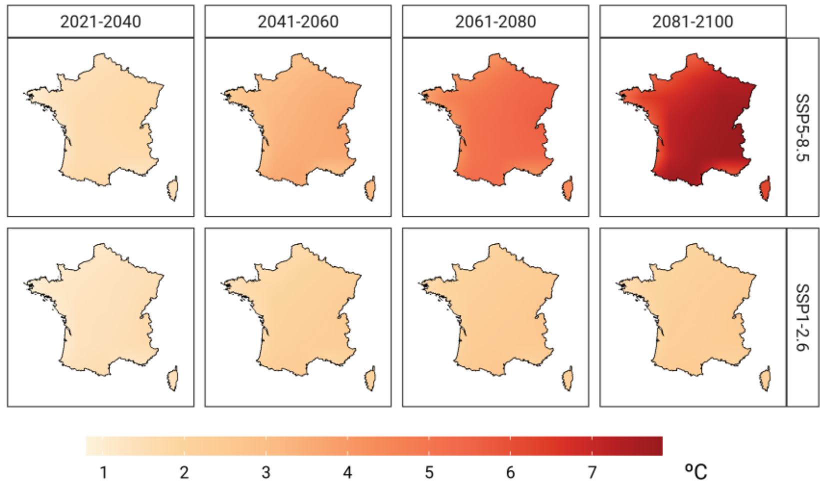

Figure B.15 Projected changes in mean temperature (°C) during the growing season of wine grapes.

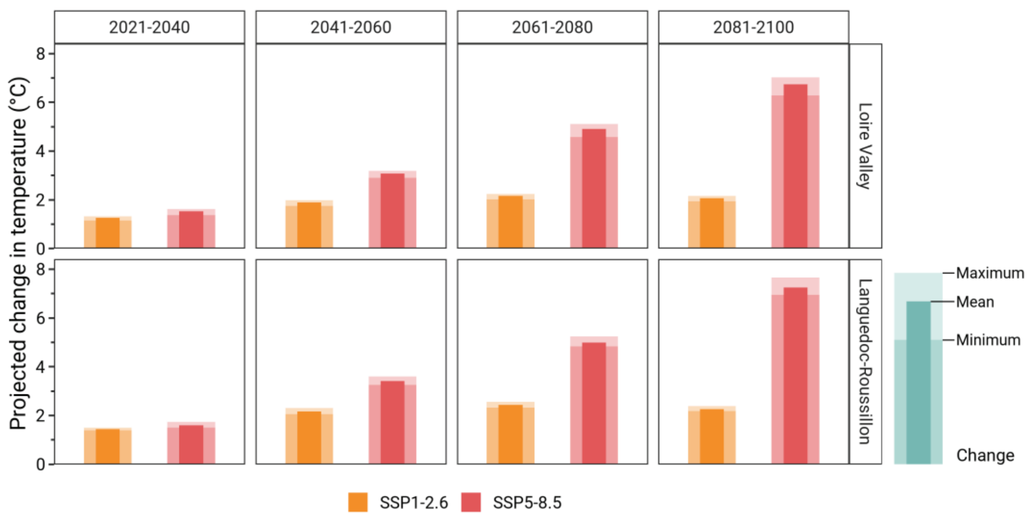

Figure B.16 Projected change in mean temperature (°C) during the growing season of wine grapes for the vineyards of the case study.

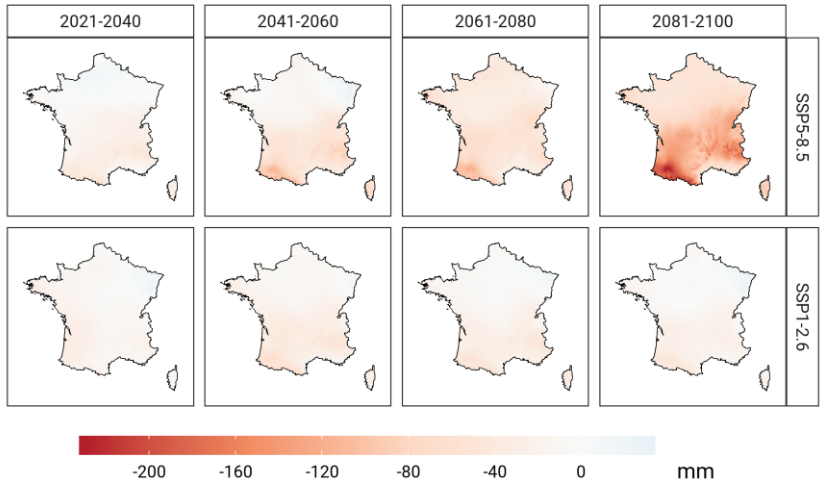

Figure B.17 Projected changes in total precipitation (mm) during the growing season of wine grapes.

Figure B.18 Projected change in total precipitation (mm) during the growing season of wine grapes for the vineyards of the case study.

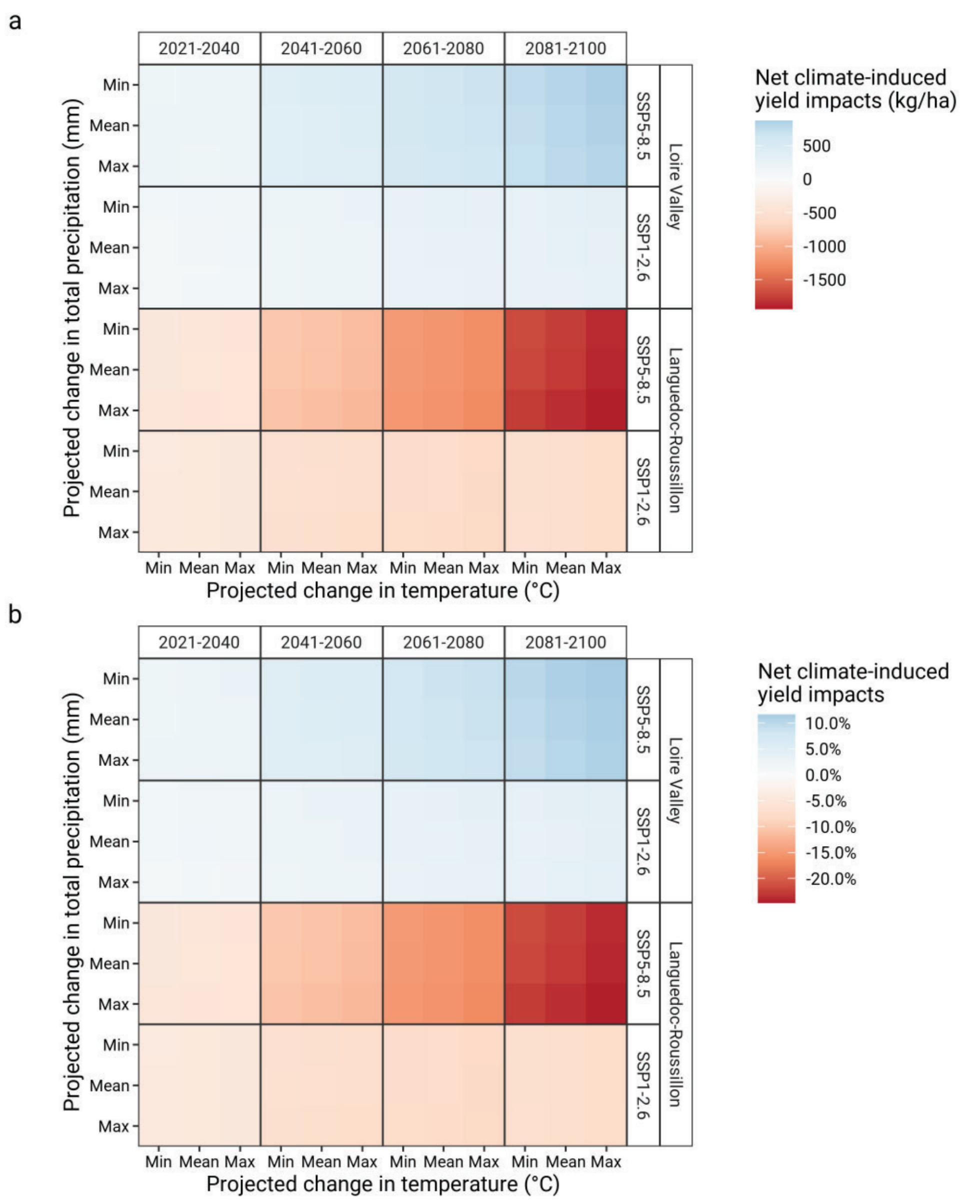

Figure B.19 Projected change in grape yield for the vineyards of the case study: (a) absolute value (kg/ha), (b) relative to current yield (%).

B3.3 Coverage of active ingredients of pesticides by the impact assessment method

Table B.7 and Table B.8 show the active ingredients of the pesticides applied respectively in the vineyards from Languedoc-Roussillon and Loire Valley. The column status of these columns indicates whether the potential ecosystem quality and human health impacts were evaluated (*characterized*), that is, if there are characterization factors available for the active ingredients.

Table B.7 Status of the active ingredients of the pesticides applied in the vineyard from Languedoc-Roussillon.

Active ingredient	**Status**
Chlorpyrifos methyl	Characterized
Copper	Characterized
Cymoxanil	Characterized
Folpet	Characterized
Fosetyl-aluminium	Characterized
Glyphosate	Characterized
Mancozeb	Characterized
Tebuconazole	Characterized
Flazasulfuron	Characterized
Quinoxyfen	Characterized
Emamectin benzoate	Missing characterization factors
Sulfur	Missing characterization factors
Trifloxystrobin	Missing characterization factors
Flumioxazin	Missing characterization factors
Fluopyram	Missing characterization factors
Myclobutanil	Missing characterization factors
Spiroxamin	Missing characterization factors

Table B.8 Status of the active ingredients of the pesticides applied in the vineyard from Loire Valley.

Active ingredient	**Status**
Aminotriazole	Characterized
Difenoconazole	Characterized
Dimethomorph	Characterized
Glyphosate	Characterized
Mancozeb	Characterized

Ammonium thiocyanate	Missing characterization factors
Flazasulfuron	Missing characterization factors
Trifloxystrobin	Missing characterization factors
Meptyldinocap	Missing characterization factors

B3.4 Impacts of extreme events

B3.4.1 Hail

Table B.9 Expected impact of hail on grape yield (%YLR) for the vineyards of the case study by SSP and period.

Vineyard	SSP1-2.6				SSP5-8.5			
	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	0.9	1.3	1.4	1.4	1.1	2.1	3.3	4.7
Languedoc-Roussillon	1.1	1.5	1.6	1.5	1.1	2.3	3.4	5

B3.4.2 Frost

Table B.10 Frost probability (%) for the vineyards of the case study by SSP and period.

Vineyard	SSP1-2.6				SSP5-8.5			
	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	1.7	0.8	1.9	2.9	1.5	1.3	0.4	0.4
Languedoc-Roussillon	0.6	0.3	1.2	1.1	0.9	0.3	0.1	0.1

Table B.11 Expected impact of frost on grape yield (%YLR) for the vineyards of the case study by SSP and period.

Vineyard	**SSP1-2.6**				**SSP5-8.5**			
	2021-2040	**2041-2060**	**2061-2080**	**2081-2100**	**2021-2040**	**2041-2060**	**2061-2080**	**2081-2100**
Loire Valley	0.7	0.3	0.7	1.1	0.6	0.5	0.2	0.2
Languedoc-Roussillon	0.2	0.1	0.5	0.4	0.4	0.1	0	0

B3.4.3 Drought

Table B.12 Drought probability (%) for the vineyards of the case study by SSP and period.

Vineyard	**SSP1-2.6**				**SSP5-8.5**			
	2021-2040	**2041-2060**	**2061-2080**	**2081-2100**	**2021-2040**	**2041-2060**	**2061-2080**	**2081-2100**
Loire Valley	76.4	71.2	72.2	67.5	68.4	73.3	86.6	96.4
Languedoc-Roussillon	70.7	77.4	63.6	61.6	64.7	85	92	97.9

Table B.13 Expected impact of drought on grape yield (%YLR) for the vineyards of the case study by SSP and period.

Vineyard	Baseline	SSP1-2.6				SSP5-8.5			
	1970-2000	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	21.6	22.9	21.4	21.7	20.2	20.5	22	26	28.9
Languedoc-Roussillon	20.5	21.2	23.2	19.1	18.5	19.4	25.5	27.6	29.4

B3.4.4 Heat waves

Table B.14 Heat waves (spell of nine consecutive days) probability (%) for the vineyards of the case study by SSP and period.

Vineyard	SSP1-2.6				SSP5-8.5			
	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	80	60	70	30	50	100	100	100
Languedoc-Roussillon	60	100	70	30	100	100	100	100

Table B.15 Number of multi-day heat waves (spell of nine consecutive days) for the vineyards of the case study by SSP and period.

Vineyard	SSP1-2.6				SSP5-8.5			
	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	0.8	0.6	0.7	0.3	0.5	1.4	4.4	5.9
Languedoc-Roussillon	0.6	1.2	0.7	0.3	1	2.5	5.7	7.4

Table B.16 Expected impact of heat waves on grape yield (%YLR) for the vineyards of the case study by SSP and period.

Vineyard	SSP1-2.6				SSP5-8.5			
	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	28	21	24.5	10.5	17.5	35	35	35
Languedoc-Roussillon	21	35	24.5	10.5	35	35	35	35

B3.4.5 Phytopathology

Table B.17 Projected mean warm and wet days for the vineyards of the case study by SSP and period.

Vineyard	SSP1-2.6				SSP5-8.5			
	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	4.9	5.4	6	5.3	5.8	7.1	7.8	9.9
Languedoc-Roussillon	1.5	1.6	2	2.6	1.4	2.5	4.1	4.8

Table B.18 Expected increase of phytosanitary treatments (%) for the vineyards of the case study by SSP and period.

Vineyard	SSP1-2.6				SSP5-8.5			
	2021-2040	2041-2060	2061-2080	2081-2100	2021-2040	2041-2060	2061-2080	2081-2100
Loire Valley	24.5	27	30	26.5	29	35.5	39	49.5
Languedoc-Roussillon	7.5	8	10	13	7	12.5	20.5	24

B3.4.6 Status of adaptation strategies

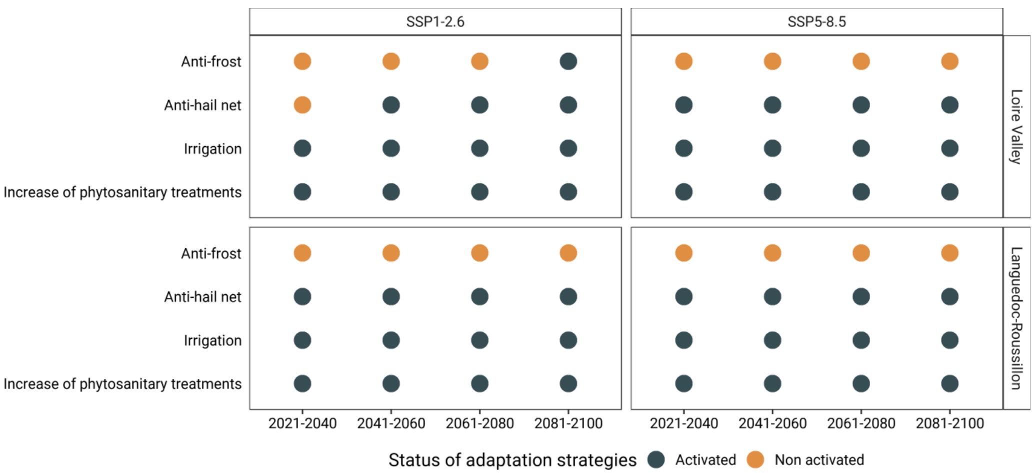

Figure B.20 Status of adaptation strategies in two French vineyards under future periods according to two emissions scenarios.

Table B.19 Projected irrigation demands for the vineyards of the case study by SSP and period.

Vineyard	**SSP1-2.6**				**SSP5-8.5**			
	2021-2040	**2041-2060**	**2061-2080**	**2081-2100**	**2021-2040**	**2041-2060**	**2061-2080**	**2081-2100**
Loire Valley	415	439	371	282	323	365	572	833
Languedoc-Roussillon	344	449	459	394	416	610	529	896

B3.5 Influence of extreme events and adaptation strategies on LCIA results

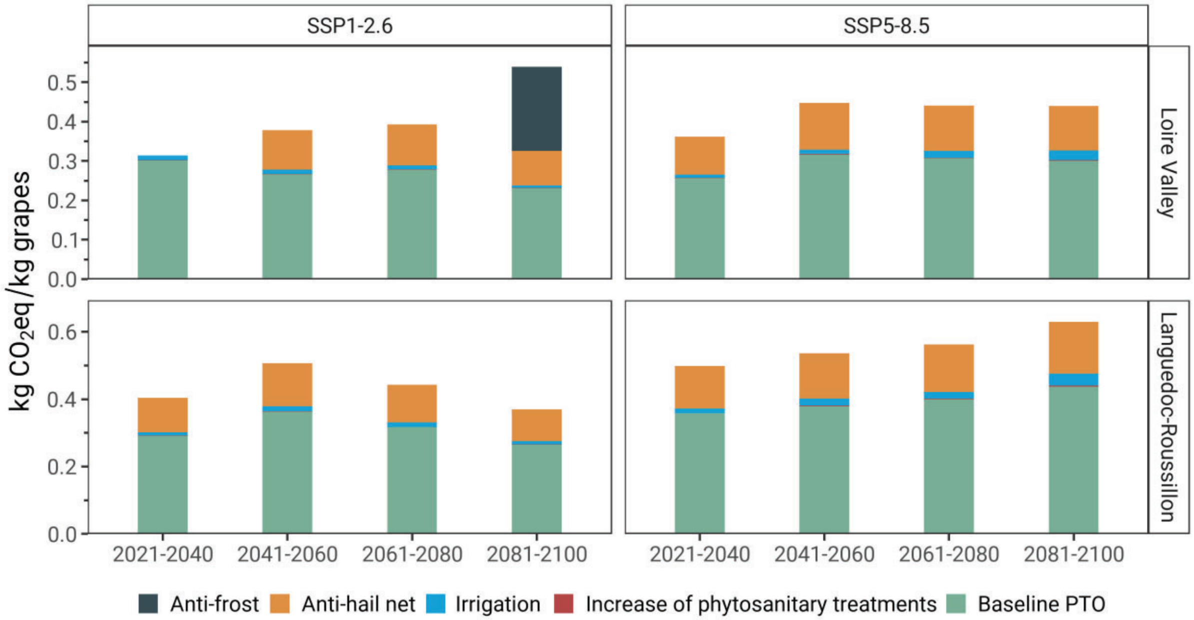

Figure B.21 Carbon footprint (kg $CO_2eq \cdot kg\ grapes^{-1}$) of grape production for future periods under two emissions scenarios in two French vineyards. The bars show the contribution of adaptation levers.

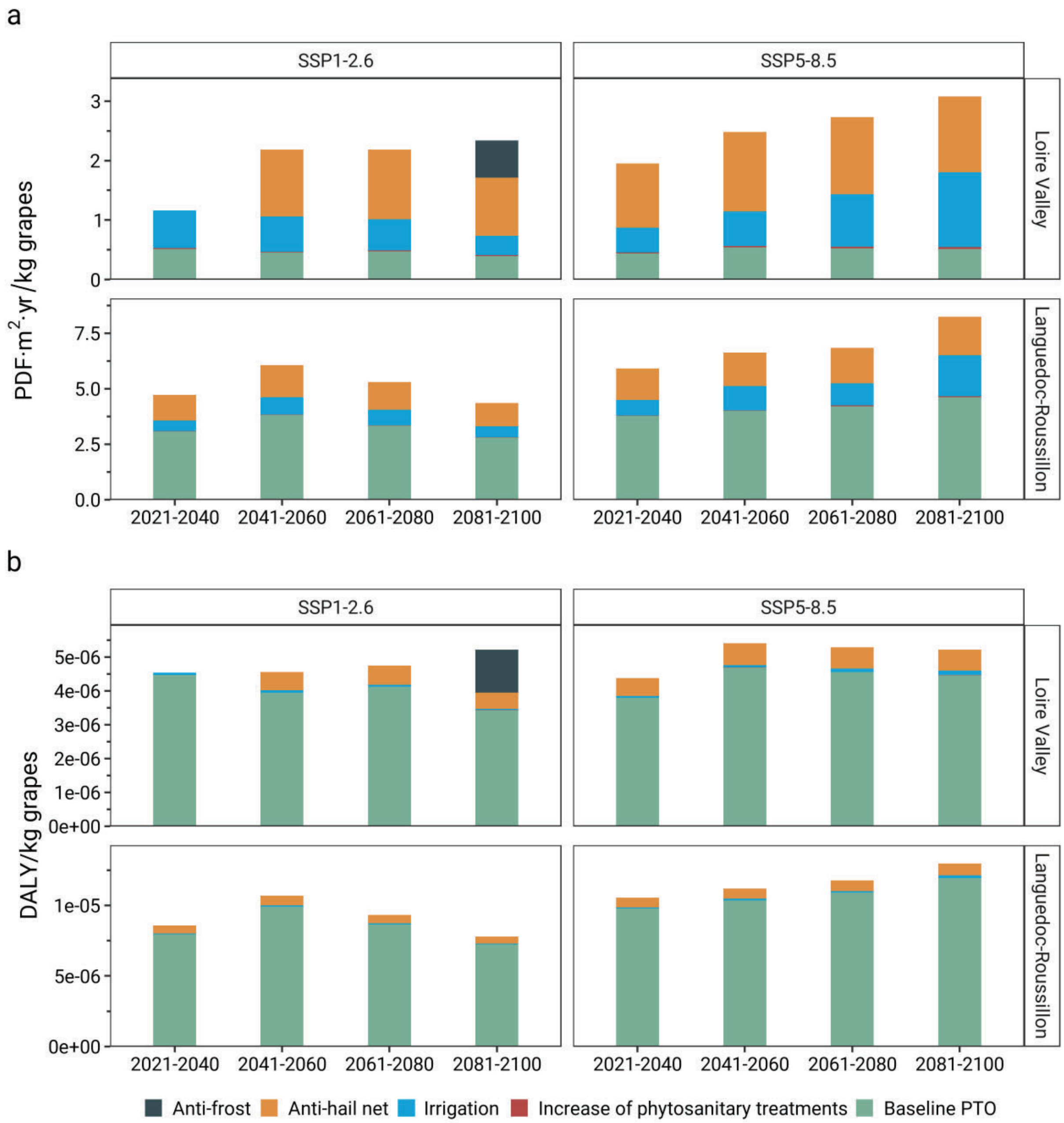

Figure B.22 Ecosystem quality (EQ) impacts (PDF·m2·yr·kg grapes-1) (a) and human health (HH) impacts (DALY·kg grapes^{-1}) (b) of grape production for future periods under two emissions scenarios in two French vineyards. The bars show the contribution of adaptation levers.

ANNEXE C INFORMATIONS SUPPLÉMENTAIRES POUR L'ARTICLE PRÉSENTÉ AU CHAPITRE 6

C1 Study area and parameters affected by climate change

C1.1 Study area definition and copper concentration in European soils

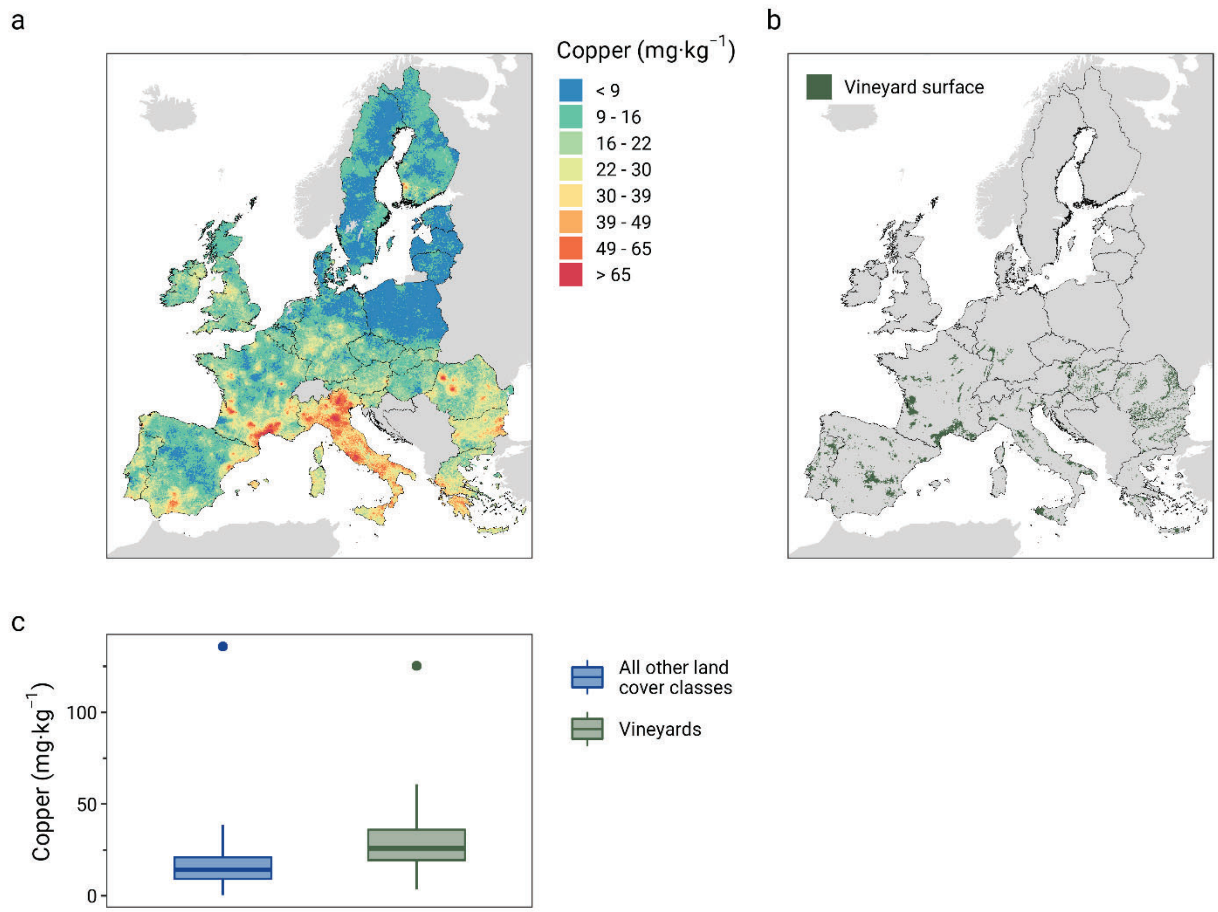

Figure C1 Topsoil copper concentration (mg kg^{-1}) in European soils (Ballabio et al., 2018) (a). European vineyard area according to CORINE land Cover data sets (EEA, 2017) (b). Distribution of topsoil copper concentration (mg kg^{-1}) according to land cover classes based on data from (Ballabio et al., 2018) (c).

C1.2 Preliminary sensitivity analysis of fate factors for a direct emission of copper to soil to partitioning coefficients, precipitation, and soil erosion

As described in the main text, the fate of direct emission of copper to the soil, either natural or agricultural soil, is controlled by the following removal processes:

- Transfer from soil to freshwater compartment
- Leaching.

Equation C1 describes the transfer rate of a chemical from soil ($k_{sl\rightarrow fw[S]}$ [d^{-1}]) to freshwater (P. Fantke et al., 2018).

$$k_{sl\rightarrow fw[S]} = \frac{\left(\frac{v_{\mathrm{rain[S]}} \cdot fr_{V_{\mathrm{rain,runoff,sl[S]}}}}{K_{sl|w,sl[\mathrm{S}]}} + v_{\mathrm{sl[S]}}\right)}{h_{\mathrm{sl[S]}}} \cdot (3600 \cdot 24) \qquad \text{(Equation C1)}$$

Equation C2 describes the transfer rate of a chemical from soil ($k_{\mathrm{leach,sl[S]}}$ [d^{-1}]) by leaching (P. Fantke et al., 2018).

$$\mathrm{k}_{\mathrm{leach,sl[S]}} = \frac{\left(\frac{\mathrm{v}_{\mathrm{rain[S]}} \cdot fr\_V_{\mathrm{rain,inf,sl[S]}}}{K_{\mathrm{sl|w,sl[S]}}}\right)}{h_{\mathrm{sl[S]}}} \cdot (3600 \cdot 24) \qquad \text{(Equation C2)}$$

Definition of the parameters of Equations C1 and C2:

$v_{\mathrm{rain[S]}}$	average annual precipitation [$m \cdot s^{-1}$]
$fr\_V_{\mathrm{rain,runoff,sl[S]}}$	volume fraction of precipitation running off from continental and global natural and agricultural soil [-]
$v_{\mathrm{sl[S]}}$	erosion of continental and global natural and agricultural soil [$m \cdot s^{-1}$]
$k_{\mathrm{sl\|w,sl[S]}}$	partition coefficient continental and global natural and agricultural soil/water
$h_{\mathrm{sl[S]}}$	depth of continental and global natural and agricultural soil [m]

$3600 \cdot 24$	conversion factor [$s \cdot d^{-1}$]
$fr\_V_{\mathrm{rain,inf,sl[S]}}$	volume fraction of precipitation infiltrating to continental and global natural and agricultural soil [-]

In USEtox, the fraction of precipitation running off from soil ($fr\_Vrain,runoff,sl_{[S]}$) is a landscape parameter that was calculated for each landscape archetype based on empirical data from the Global Runoff Data Centre (Kounina et al., 2014). To the best of our knowledge, no data sets are projecting the future fraction of precipitation running off from soil that are consistent with the datasets describing the current state of the environment in USEtox. Consequently, for modelling both baseline and future scenarios, we selected the value of $fr\_Vrain,runoff,sl_{[S]}$ corresponding to the European landscape (W13).

With respect to the volume fraction of precipitation infiltrating to soil ($fr\_Vrain,inf,sl_{[S]}$), the USEtox model does not consider spatial differentiation for this parameter and uses a constant value of 0.269 ($mm \cdot yr^{-1}$). Accordingly, we kept the same value for modelling both the current and future scenarios.

Apart from the depth of soil ($h_{\mathrm{sl[S]}}$), which we kept constant for both the baseline and future scenarios, the reaming factors of Equations C1 and C2 are vulnerable to climate change. Therefore, we performed a preliminary sensitivity analysis to show that the USEtox model allows to account for the influence of projected changes in the partitioning coefficient soil-water (Equation C10), rain (Figure C5), and erosion (Figure C4) on the fate factor for a direct emission of copper to soil (Table C1 and Figure C2).

Table C1 Description of preliminary sensitivity analysis performed on the influence of substance and landscape parameters on the fate factor for a direct emission to agricultural soil.

Parameter (class)	**Unit**	**Default value**	**Tested values (min, max)**	**Change in parameter**	**Source**
Partitioning coefficient (Kd) (substance)	l/kg	530	(437, 2188)	2.5th percentile value of Kd 97.5th percentile value of Kd	(Ivan Viveros Santos et al., 2018)
Rain rate (landscape)	mm/yr	552.6	(100, 865)	Low rain (lower bound of the first quantile) High rain (lower bound of second-to-last quantile)	Figure C5 (Fick & Hijmans, 2017)
Soil erosion (landscape)	mm/yr	0.03	(0.023, 0.035)	Low erosion rate (decrease of 25%) High erosion rate (increase of 60%)	(Panagos et al., 2021)

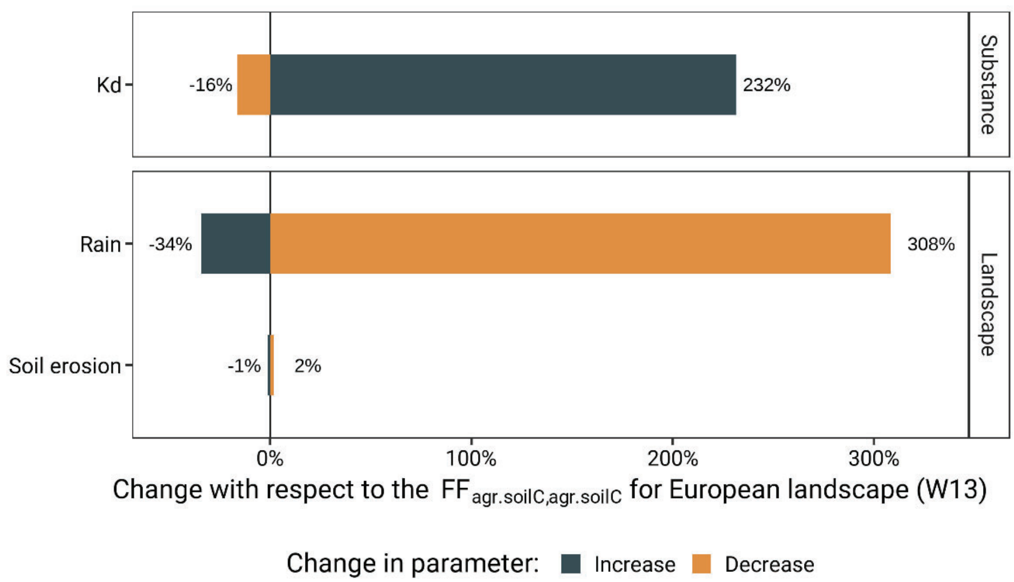

Figure C2 Tornado plot of the sensitivity analysis performed on fate factors for a direct emission of copper to agricultural soil.

C1.3 Current and future soil organic matter in Europe

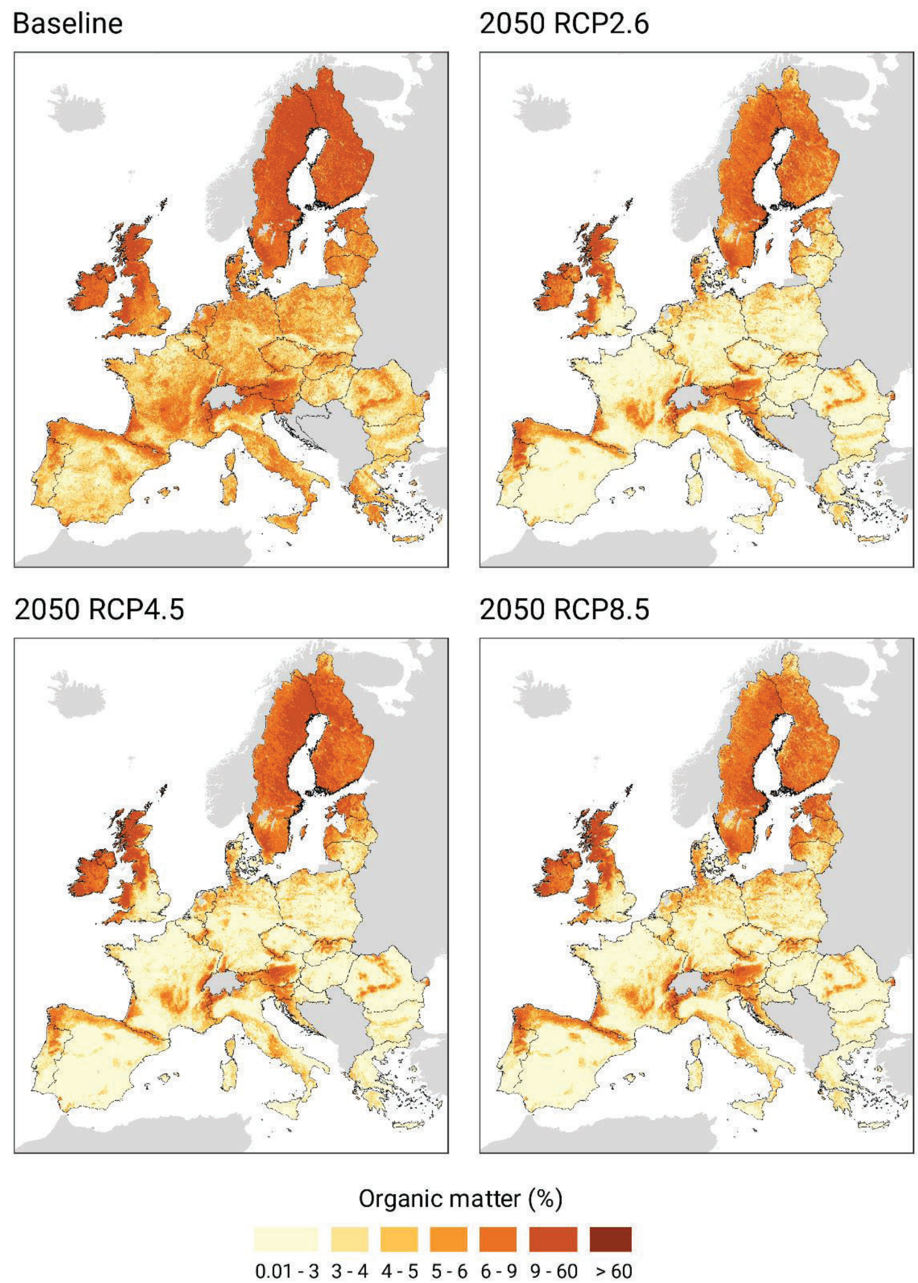

Figure C3 Soil organic matter (%) (coloured by quantiles) for the baseline period and Representative Concentration Pathways (RPCs) according to the Global Climate Model IPSL-CM5A-LR. Our computations, based on data from Yigini and Panagos (2016).

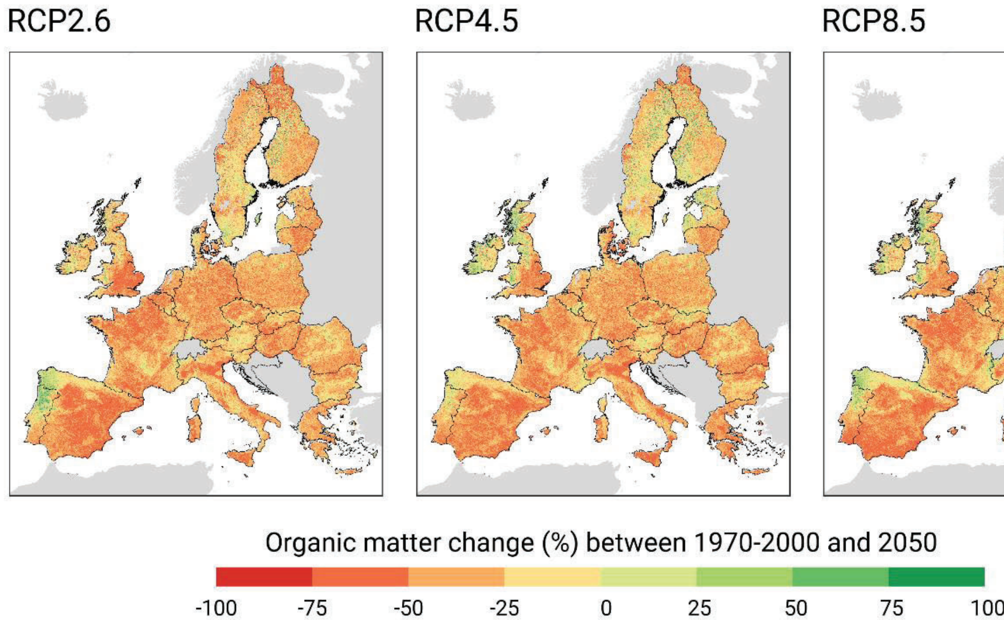

Figure C4 Organic matter change (%) between 1970-2000 and 2050 according to the Global Climate Model IPSL-CM5A-LR. Our computations, based on data from Yigini and Panagos (2016).

C1.4 Current and future soil erosion rates in Europe

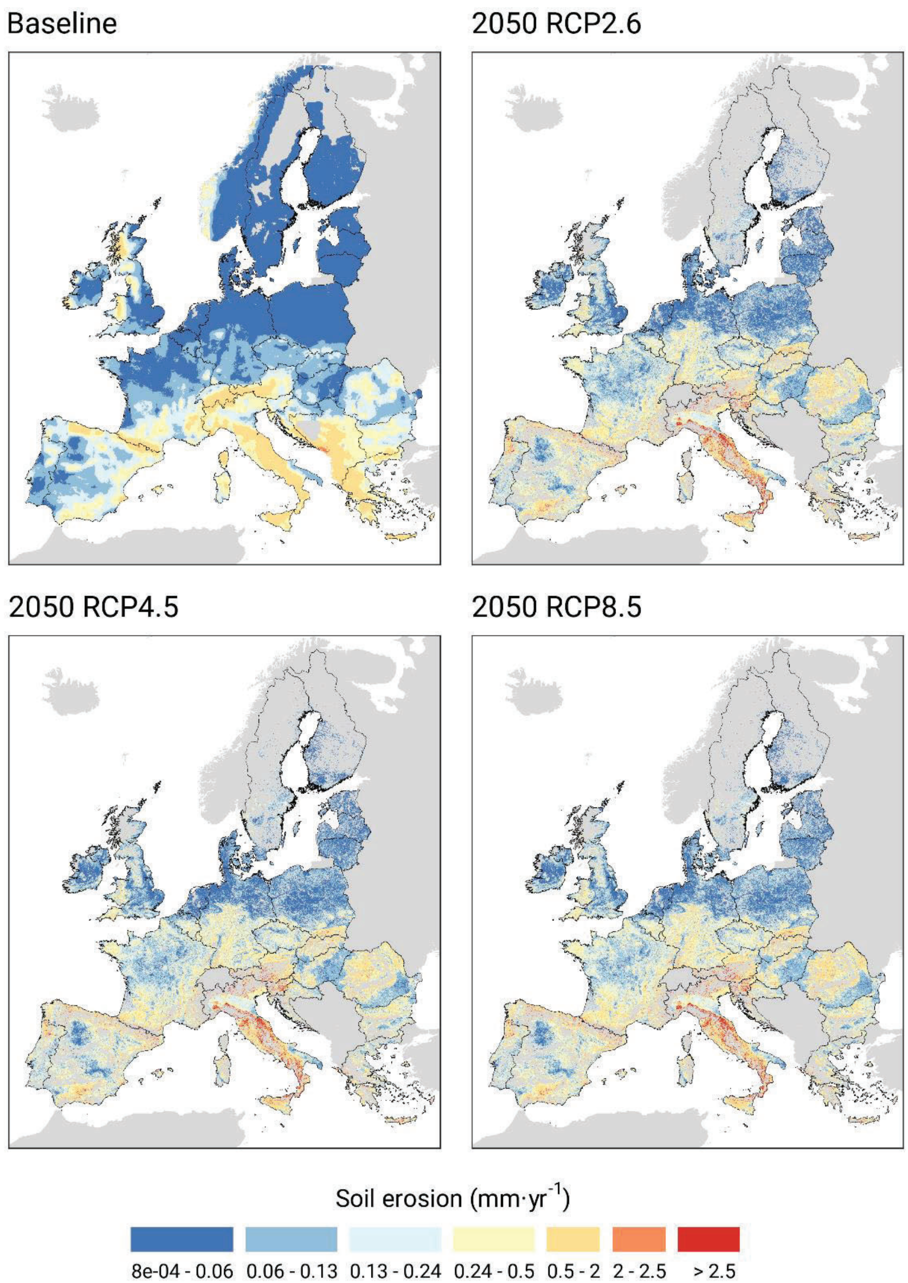

Figure C5 Soil erosion (mm·yr^{-1}) (coloured by quantiles) for the baseline period and Representative Concentration Pathways (RPCs) according to an average composite of 19 Global Climate Models. Our computations, based on data from Panagos et al. (2021).

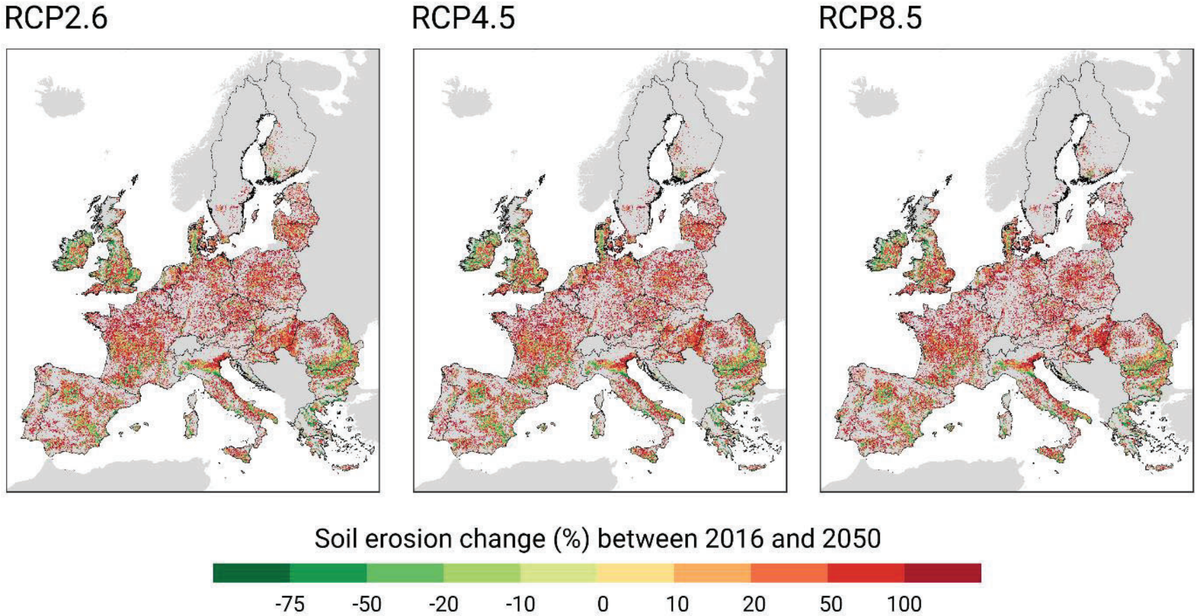

Figure C6 Soil erosion change (%) between 2016 and 2050 according to an average composite of 19 Global Climate Models. Our computations, based on data from Panagos et al. (2021).

C1.5 Current and future precipitation in Europe

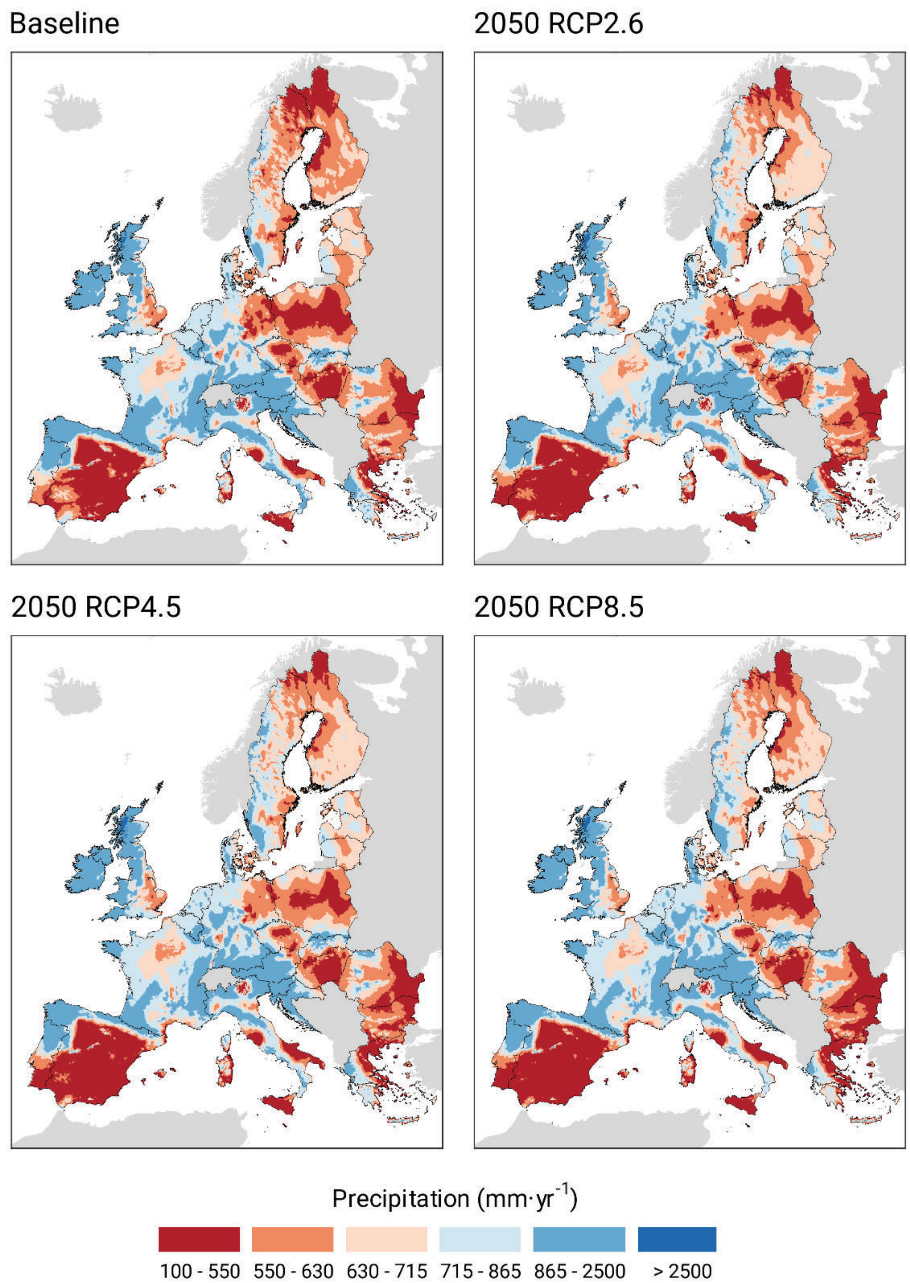

Figure C7 Average annual precipitation (mm ·yr-1) (coloured by quantiles) for the baseline period and Representative Concentration Pathways (RPCs) according to the Global Climate Model IPSL-CM5A-LR. Our computations, based on data from Fick and Hijmans (2017).

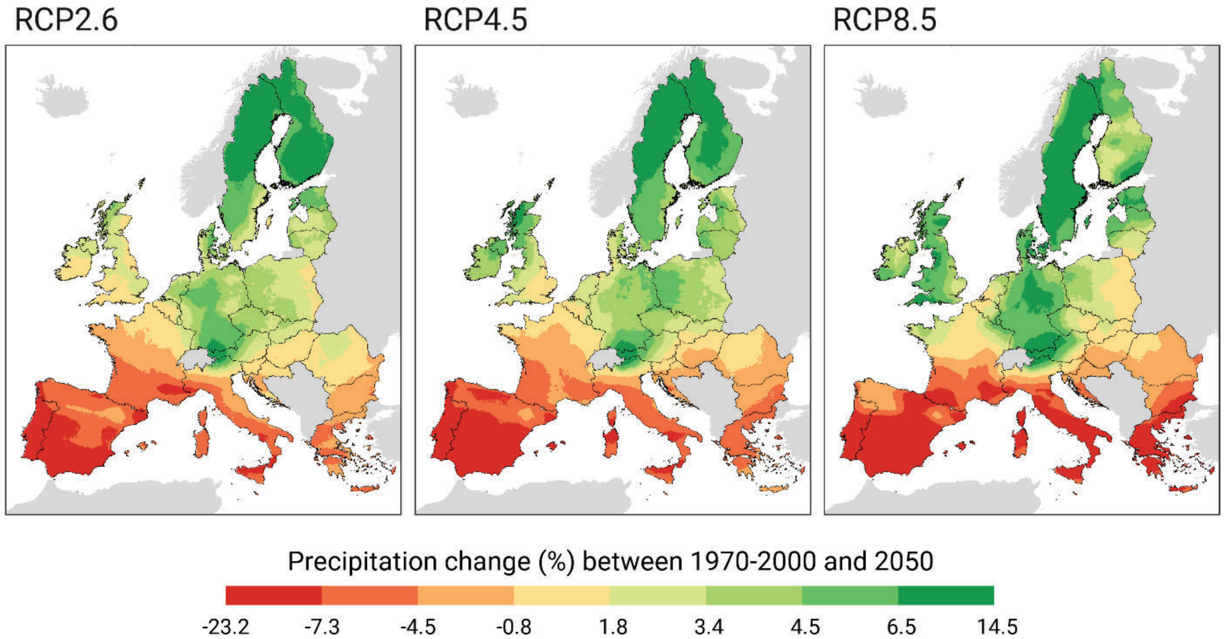

Figure C8 Precipitation change (%) between 1970-2000 and 2050 according to the Global Climate Model IPSL-CM5A-LR. Our computations, based on data from Fick and Hijmans (2017).

C2 Parameters and equations for calculating metal speciation

C2.1 Topsoil copper concentration (mg/kg)

Total copper concentration in topsoil (Cu_{total}) is required for computing the reactive metal concentration ($Cu_{reactive}$) (Equation C6). The former concentration was obtained from a raster layer at 500m resolution (Figure C1-a) (Ballabio et al., 2018), which is available from the European Soil Data Centre (European Commission, 2022b; Panagos et al., 2012).

C2.2 Sum of amorphous Fe and Al(Hydr)Oxides (mmol/kg)

This parameter is needed for calculating the total dissolved fraction from reactive metal (Equation C7). In agreement with a previous study (Ivan Viveros Santos et al., 2018), this parameter was set constant and equivalent to the value reported by Owsianiak et al. (2013).

$$FeAlox = 89\ mmol/kg$$

C2.3 Dissolved organic carbon (DOC - mg/L)

In agreement with Owsianiak et al. (2013) and Viveros Santos (2018), dissolved organic carbon (*DOC* – mg/l) was computed according to an empirical regression reported by Römkens et al. (2004).

$$log_{10}(DOC) = 2.25 + 0.75 \cdot \ log_{10}(OM) - 0.20 \cdot \ pH - 0.30 \cdot \ log_{10}(ECW) \quad \text{(Equation C3)}$$

With *OM* expressed in % and the electrical conductivity of soil pore water (*ECW*) in dS/m.

C2.4 Activity coefficient for cations

Activity coefficients (Equation C4) were used to relate cations activities (in curly braces in Equation C5, which is an example for magnesium) and cation concentrations (in square brackets in Equation C5). The activities of cations are parameters required for computing EC_{50} based on the Terrestrial Biotic Ligand Models (Table C2). The activity coefficients were computed with the modified Debye-Hückel equation, as reported by Owsianiak et al. (2013).

$$\log_{10}(f_i) = -0.512 \cdot z_i^2 \cdot \left[\frac{IS^{0.5}}{1 + IS^{0.5}} - 0.3 \cdot IS\right] \quad \text{(Equation C4)}$$

$$\{Mg^{2+}\} = f_{di}[Mg^{2+}] \quad \text{(Equation C5)}$$

With f_i, the activity coefficient for a cation (*i*: monovalent; *di*: divalent) and *IS* the effective ionic strength of soil pore water (mol/l).

C2.5 Reactive copper concentration (mg/kg)

In line with Owsianiak et al. (2013) and Viveros Santos (2018), reactive metal concentration ($Cu_{reactive}$) was calculated according to an empirical regression reported by Römkens et al. (2004).

$$log_{10}(Cu_{reactive}) = -0.331 + 0.023 \cdot \ log10(OM) \ - 0.171 \cdot \ log10(CLAY) + 1.152 \cdot \ log10(Cu_{total}) \quad \text{(Equation C6)}$$

The units of $Cu_{reactive}$ and Cu_{total} are mg/kg, and % for *OM* and *CLAY*.

C2.6 Total dissolved metal concentration (mol/L)

Total dissolved metal concentration ($Cu_{total\ dissolved}$) was calculated with an empirical regression published by Groenenberg et al. (2012), following Owsianiak et al. (2013) and Viveros Santos (2018).

$$log_{10}(Cu_{total\ dissolved}) = -3.74 + 0.60 \cdot \log_{10}(Cu_{reactive}) - 0.28 \cdot \log_{10}(OM) - 0.79 \cdot log_{10}(FeAlox) + 0.79 \cdot \log_{10}(DOC) \quad \text{(Equation C7)}$$

The units of $Cu_{reactive}$ are mol/kg, % for *OM* and *CLAY*, mmol/kg for *FeAlox* and mg/L for *DOC*.

C2.7 Free ion activity from reactive metal (mol/L)

The free ion activity (Cu_{free}) was calculated with an empirical regression model reported by Groenenberg et al. (2012).

$$\log_{10}\left(Cu_{free}\right) = 0.48 + 0.81 \cdot \log_{10}(Cu_{reactive}) - 0.89 \cdot \log_{10}(OM) - pH \quad \text{(Equation C8)}$$

The units for Cu_{free} are mol/L, mol/kg for reactive metal ($Cu_{reactive}$), and % for OM.

C3 EC50 from terrestrial biotic ligand models (TBLM)

We used the TBLMs reported by Thakali et al. (2006) to calculate EC_{50} activities for six endpoints by applying Equation C9.

$$\{M^{2+}\}_{EC50} = \frac{f_{50}}{(1 - f_{50}) \cdot K_{MBL}} \cdot \left(1 + \sum K_{XBL} \cdot \{X^{z+}\}\right) \quad \text{(Equation C9)}$$

f_{50} is the fraction of the total biotic ligand (BL) sites occupied by the metal, which translates into a 50% effect level. For the six available biological endpoints, f_{50} takes the value of 0.05. $\{X^{z+}\}$ is the activity of cation *X* and K_{XBL} is its corresponding conditional binding constant. Finally, K_{MBL} is the conditional binding constant of metal M^{2+} (Thakali et al., 2006).

Table C3 Parameters and biological endpoints used for the TBLMs for copper ecotoxicity modelling (Thakali et al., 2006).

Biological endpoint	**log10 KCuBL**	**log10 KHBL**	**log10 KMgBL**
Barley root elongation (BRE)	7.41 (0.23)	6.48 (0.26)	-
Tomato shoot yield (TSY)	5.65 (0.10)	4.38 (0.21)	-
F. candida juvenile production (FJP)	4.62 (0.12)	2.97 (0.62)	-
E. Fetida cocoon production (ECP)	6.50 (0.25)	5.90 (0.29)	-
Glucose induced respiration (GIR)	6.69 (0.10)	7.5	-
Potential nitrification rate (PNR)	4.93 (0.48)	4.45 (0.58)	1.64 (5.80)

C4 Intermediate factors for calculating characterization factors for copper terrestrial ecotoxicity

C4.1 Fate factor

Kd corresponds to the ratio of metal concentration in the solid phase over the concentration of solubilized metal. Where $C_{solution\ phase}$, the concentration of copper in the solution phase is calculated according to Equation C7, and $C_{solid\ phase}$ is the copper concentration in soil (Table 6.2 from the main text).

$$K_d = \frac{C_{solid\ phase}}{C_{solution\ phase}} \qquad \text{(Equation C10)}$$

C4.2 Accessibility factor

The accessibility factor (ACF) was computed according to Equation C11, where $\Delta C_{reactive}$ is the concentration of reactive metal and was calculated according to Equation C6. ΔC_{total} is the copper concentration in soil (Table 6.2 from the main text).

$$ACF = \frac{\Delta C_{reactive}}{\Delta C_{total}} \quad \text{(Equation C11)}$$

C4.3 Bioavailability factor

The bioavailability factor (BF) was computed according to Equation C12. ΔC_{free} is the concentration of free ion copper, calculated according to Equation A8, and $\Delta C_{reactive}$ is the concentration of reactive metal and was calculated according to Equation C6. θ_w and ρ_b are the volumetric water content and bulk soil density, respectively. The source of the latter parameter is shown in Table 6.2 of the main text.

$$\mathrm{BF} = \frac{\Delta C_{free} \cdot \theta_w}{\Delta C_{reactive} \cdot \rho_b} \quad \text{(Equation C12)}$$

C5 Complementary results

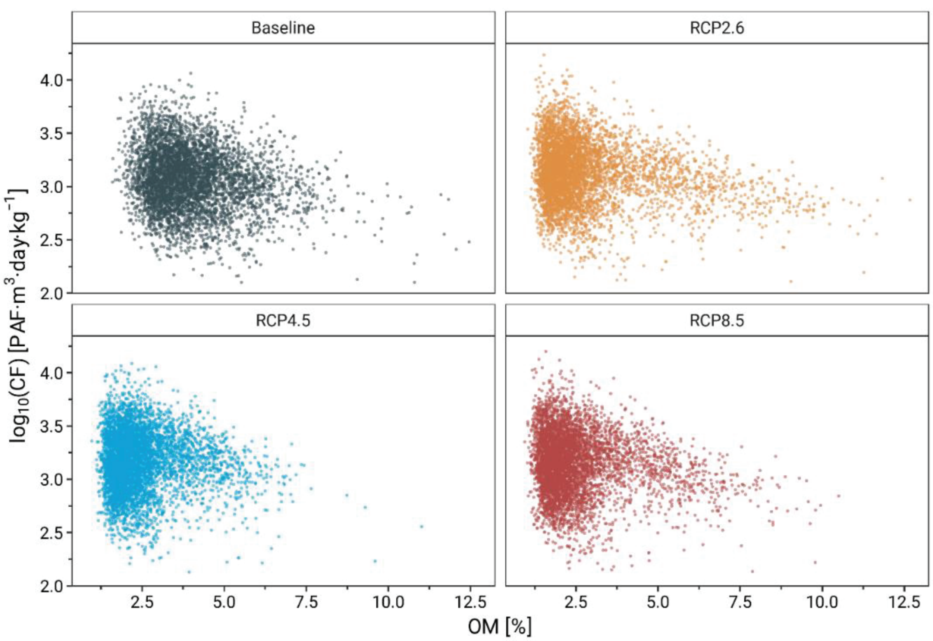

Figure C9 Correlation between regionalized CFs (PAF·m^3·day·kg^{-1}) and soil organic matter (%) across non-calcareous vineyard soils.

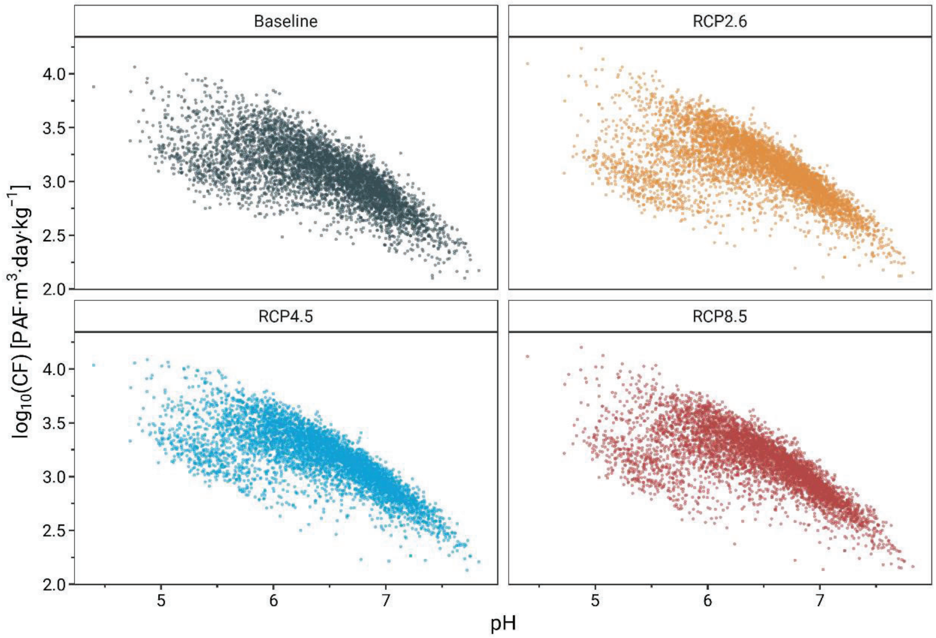

Figure C10 Correlation between regionalized CFs (PAF·m^3·day·kg^{-1}) and soil pH across non-calcareous vineyard soils.

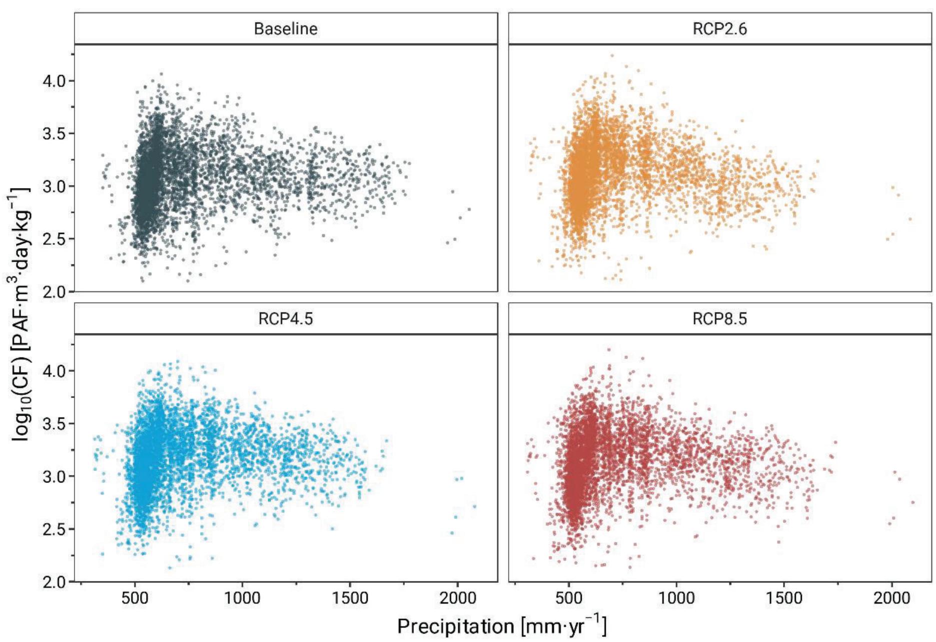

Figure C11 Correlation between regionalized CFs (PAF·m^3·day·kg^{-1}) and average annual precipitation (mm ·yr^{-1}) across non-calcareous vineyard soils.

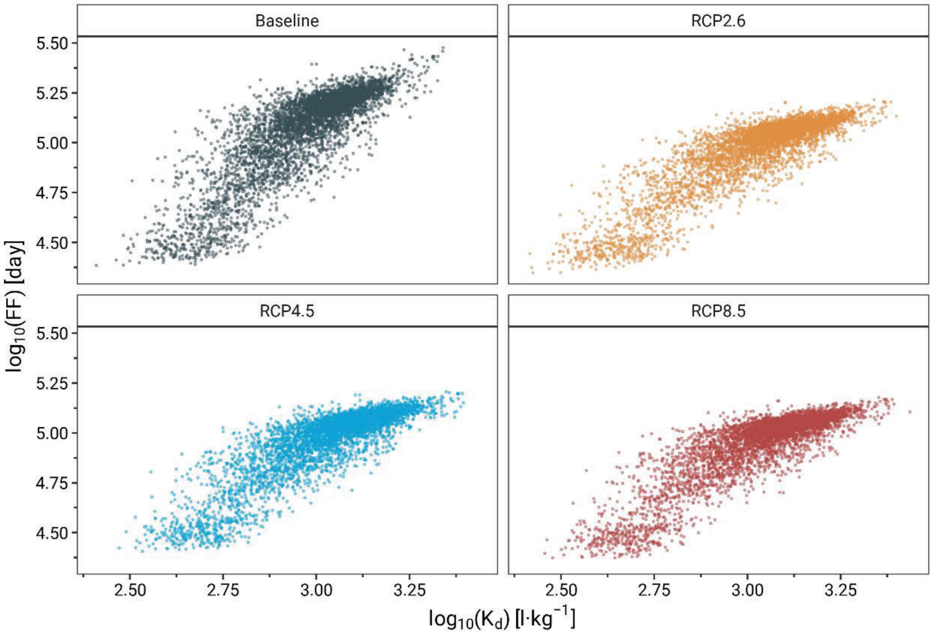

Figure C12 Correlation between fate factors (day) and log10-transformed values of partitioning coefficients (Kd) ($l \cdot kg^{-1}$).

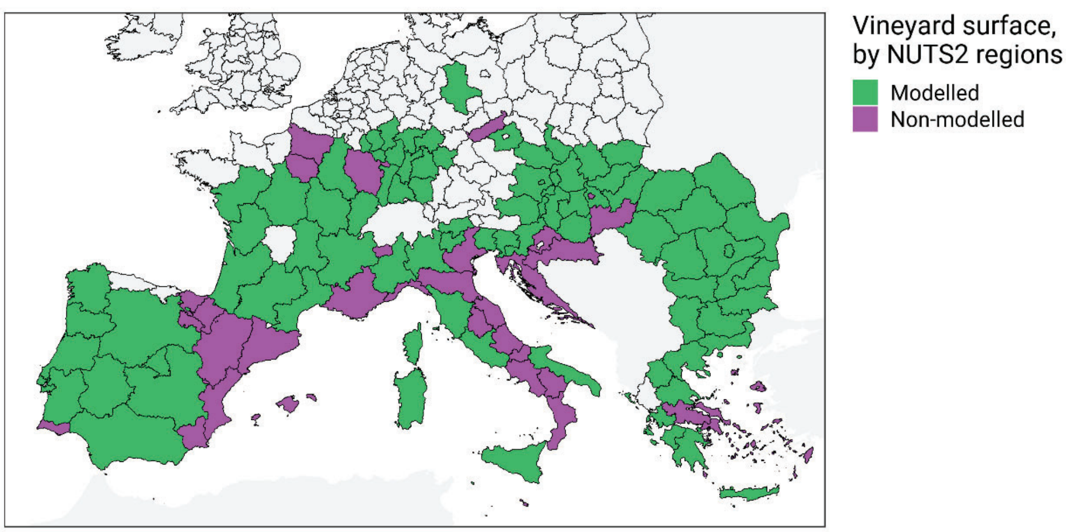

Figure C13 Vineyard surface by NUTS2 regions. The legend indicates whether CFs were determined for a given European region according to the NUTS2 level.