Environmental trade-offs in fresh-fruit cold chains by combining virtual cold chains with life cycle assessment

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ABSTRACT

Refrigeration is vital in fresh-produce supply chains for minimizing food losses. However, it requires energy and impacts the environment. To optimize the control and logistics of postharvest cold chains, we need to better identify trade-offs between maintaining fruit quality and reducing environmental impacts. Therefore, we propose a novel computational method, by combining life cycle assessment with virtual cold chains. This holistic approach allows us, on the one hand, to track the thermal history of the cooling process and fruit quality decay of each single fruit in an entire pallet throughout the cold chain, using computational fluid dynamics. On the other hand, the carbon footprint of the supply chain is quantified. This pioneering method enriches life cycle assessment with more customized input data from multiphysics modeling, and at the same time assesses food quality evolution. Significant differences between ventilated carton designs (63 g CO_2-eq/kg) and cold chain scenarios (11 g CO_2-eq/kg) were identified, namely 10% and 1.6% of the environmental impact of the entire supply chain, respectively. If solar electricity is used for precooling, the environmental impact was reduced by 55 g CO_2-eq/kg of fruit (or 8.5%), while still providing similar fruit quality retention. By combining climate impact with the predicted quality retention, this method will help retailers to choose the most optimal package design and cold chain scenario to make their food supply chains more sustainable. This approach can be applied as well for life cycle assessment of biogas conversion of food waste, amongst others.

NOMENCLATURE

Symbols

A quality attribute [-]

c constant, 3600 [kJ kWh^{-1}]

c_p specific heat capacity of the produce [kJ kg^{-1}K^{-1}]

E_e consumed electricity per month [kWh mo^{-1}]

E_A activation energy [J mol^{-1}]

k rate constant [s^{-1}]

k_0 constant [s^{-1}]

M total mass of all produce that is cooled per month [kg mo^{-1}]

n reaction's order [-]

R ideal gas constant, 8.314 [J mol^{-1} K^{-1}]

T_i product temperature at the start of the cold chain [K]

T_f product temperature at the end of the cold chain [K]

T absolute temperature [K]

t time [s]

Abbreviations

CFD   computational fluid dynamics

CO_2-eq/kg   carbon dioxide equivalent per kg of fruit

EC   energy coefficient

LCA   life cycle assessment

VCC   virtual cold chain

1 INTRODUCTION

A large share of produced fruit and vegetables are lost between leaving the farm and arriving at the retailer. These postharvest losses in fresh produce supply chains vary from 13% in Europe up to 38% in sub-Saharan Africa [1]. Proper refrigeration helps to reduce these losses as temperature is the single most important environmental factor affecting the produce deterioration rate and thereby the postharvest life. A decrease in product temperature by 10°C from ambient conditions typically doubles the shelf life [2,3]. Therefore, a rapid removal of the field heat by cooling after harvest, and the maintainance of optimum cold temperatures throughout the supply chain are essential to preserve fruit quality and minimize losses.

Refrigeration, however, consumes energy, and accounts for 8 % of all electrical energy used in the food industry [4]. With over 400,000 reefer containers and 1,000,000 refrigerated vehicles currently in use [5], the postharvest transport of such refrigerated cargo is responsible for a large share of this energy consumption. With every product that is lost within the supply chain, the corresponding energy used to preserve, and agriculturally produce it, is thereby also lost [1,6]. The cold chain thus plays an important role in the food-energy-water nexus [7,8]. Therefore, optimizing postharvest cold chains by prolonging produce shelf life, thereby reducing losses, and lowering energy consumption is essential to reduce the environmental impact. To achieve these goals, new cold chain scenarios [9,10] or ventilated package design [11,12] have recently demonstrated promising potential. However, the currently used methods to evaluate these innovative technologies still suffer from key limitations, which are discussed in the two paragraphs below.

Advanced experiments in refrigerated containers [13,14] or precooling facilities [15] have been used, as well as numerical modeling with computational fluid dynamics (CFD) [16,17] . These experimental and computational techniques [18] enabled to identify and optimize the thermal

history of individual products, arranged in larger bulks (e.g. a pallet), and their associated quality evolution. These thermophysical methods provide a very detailed insight into, and understanding of, cooling behavior in the supply chain. As a novel step in this field, a virtual cold chain (VCC) method was recently developed [15,19]. Employing the VCC method enables tracking of the thermal history and associated fruit quality of every individual fruit in an entire pallet of packaged fruit throughout the entire postharvest cold chain of interest using CFD. In addition, information on the energy use for ventilation and cooling can also be extracted. These high-resolution numerical or experimental methods, however, lack a quantification of the environmental impact of the different cooling scenarios, supply-chain itineraries and ventilated package types (e.g. cardboard vs. plastic). This is a key bottleneck of such methods, as new cooling strategies can be devised that maintain food quality better, but the data do not enable to quantify how sustainable the new processes are. For that, the entire supply chain needs to be targeted, including differences in travel times, the amount of containers or lorries needed to transport a certain amount of fruit, the food losses, and the used amount of packaging material.

Life cycle assessment (LCA) [20] is capable of providing this information, and is widely used as a decision-support tool by retailers [21], food companies and policy makers [22,23] to reduce the carbon footprint of their supply chains [24,25], including the impact of food losses [26]. The recent development of LCA methodologies and dissemination programs by international and local bodies is the basis for LCA's increased use on agricultural and industrial food products [27]. In a related context, LCA is also used to evaluate the conversion of food waste into biogas by recycling [28,29] or the use of renewable energy in distribution networks of perishables [30]. LCA enables identification of trade-offs between sourcing regions, transport itineraries, energy technologies or material usage, amongst others [31,32]. LCA however relies on inventory

databases, such as ecoinvent [33,34], which often only include generic inventory data of postharvest unit operations (precooling, refrigerated container transport or cold storage) [35]. Dynamic modeling of food products depending on the region, seasonality, food waste rates, or other parameters in the food supply chain is rare, and especially information on the energy consumption is not specific enough [36], despite the significance of the food system as an energy consumer [21]. For instance, no differentiation in energy use is made between ventilated package designs or container stowing strategies, although these differences have been recently identified to be relevant [37]. As such, the energy and quality gains from better cooling processes and better package systems are rarely explicitly incorporated in LCA [38]. In addition, although there are many studies on food waste management [27], the avoidable food losses and waste, and the associated embodied energy, are often not accounted for or only covered by approximate assumptions in most LCA studies [39]. In a recent combined effort, fruit quality, energy use, and the global warming impact of food cold chains were evaluated together [40]. However, this method did not provide a sufficient degree of detail to compare either the different package designs or the quality heterogeneity between individual fruits, for example. For retailers or food companies, it would be very useful to have a tool or method that can provide the overall environmental impact of their supply chains, and at the same time information on the temperature-dependent fruit quality evolution, by means of a food quality assessment that is linked to the cooling processes. This could help these stakeholders in the perishables supply chain to choose the most optimal package and cold chain scenario to make their food supply chains more sustainable, and to optimize logistics.

As a pioneering step towards a more holistic evaluation of fresh-produce cold chains, a combination of VCC with LCA is proposed to enrich life cycle assessment with more

customized input data from multiphysics modeling. This link between these models provides us with unique information on the temperature-dependent fruit quality reduction of each fruit in a palletized cargo, together with the environmental impact of the complete postharvest part of the supply chain. This holistic method is demonstrated for the case of an overseas citrus cold chain to identify the best-performing and most eco-friendly cold-chain scenario and package.

2   MATERIALS AND METHODS

The strategy to combine VCC with LCA is depicted in Figure 1. The different methods (VCC and LCA) are detailed below, as well as the way in which they are linked.
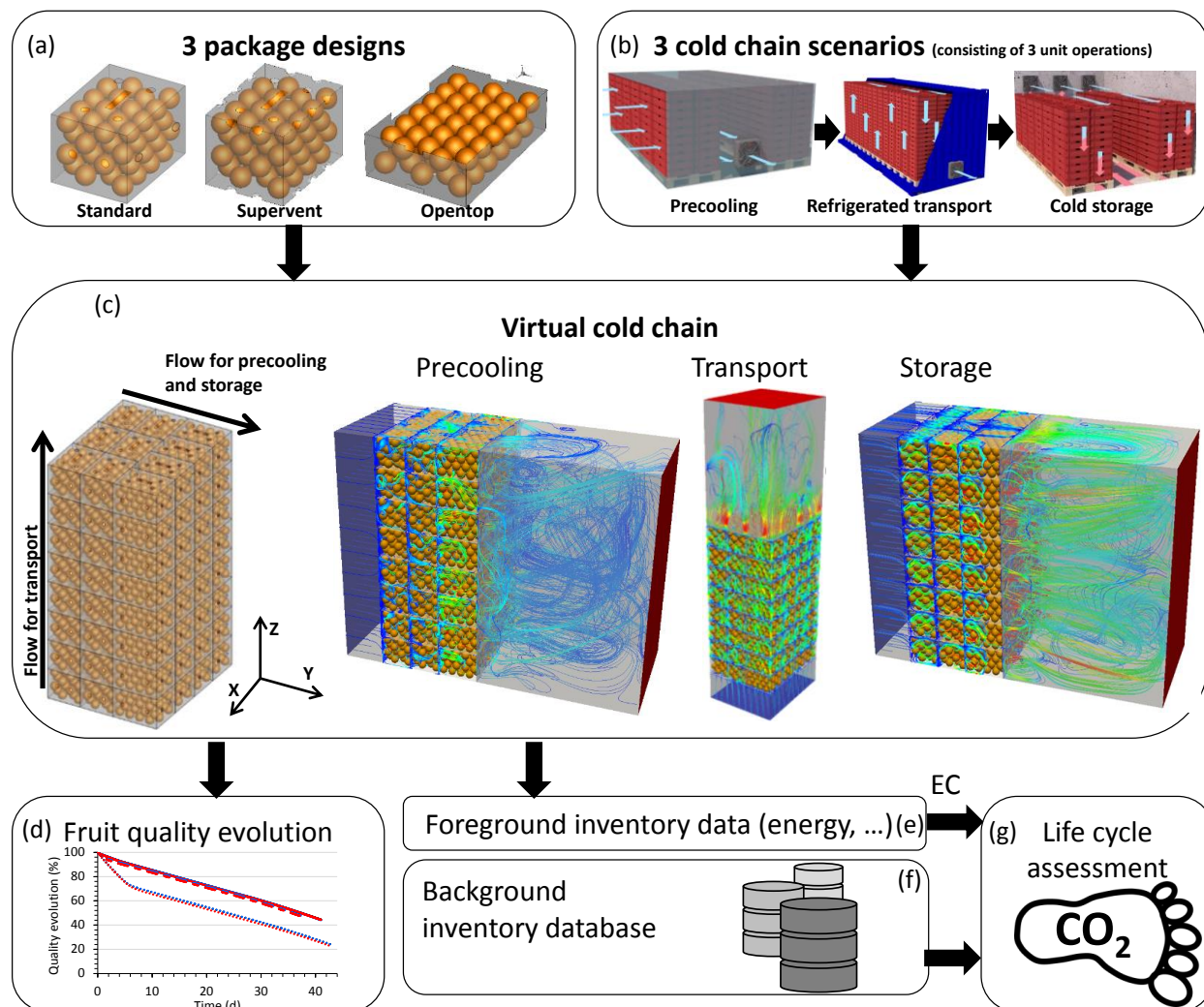
Figure 1. Schematic overview of the methodology combining virtual cold chains (VCCs) for different unit operations (precooling, refrigerated transport, cold storage) with life cycle assessment (LCA) to assess the individual fruit quality evolution within a pallet and the environmental impact of different cold chain scenarios and ventilated packag designs.

2.1 Cold chain scenarios and ventilated package type

An overseas citrus cold chain is targeted, from South Africa to Switzerland, in particular for orange fruit. Multiple cold chain scenarios and ventilated package designs are evaluated. The

three carton box designs (Figure S1) are Standard, Supervent and Opentop, which are stacked on high-cube pallets (Figure S2). The targeted cold chains are composed of three refrigerated unit operations: (1) precooling, (2) refrigerated transport, and (3) refrigerated storage (see Figure S3). By combining these unit operations, we simulate three scenarios of the cold chain for each package design:

- Forced-airflow precooling, where a precooler facility is used to rapidly remove the field heat after packaging and palletization, by forcing cold air horizontally at high airflow rates through the package. This is currently the standard practice in the South African citrus industry.

- Ambient cooling, also called static cooling, where fruits are cooled in a large cold room before shipment. This practice is often employed if the capacity of the precooler facilities is exceeded. The lower airflow rates, however, induce slower fruit cooling.

- Ambient loading, where fruits are loaded into the refrigerated container at the ambient temperature (<22 °C fruit pulp temperature) and are cooled using the container's cooling unit [9,37,41]. This novel scenario is explored in South Africa as a way to relieve pressure on the precooling facilities as well as for its logistical advantages due to reduced handling.

To serve as a comparative contrast to the South-African situation, the impact of an alternative sourcing region for citrus fruit is also evaluated, namely, fruit coming from Valencia (Spain, Europe).

2.2 Coupling VCC and LCA

The VCC and LCA methods (detailed below) are linked to enable a more holistic evaluation of fresh-produce cold chains in terms of the aforementioned package designs (Figure 1a) and cold chain scenarios (Figure 1b). The following workflow is adopted.

First, the VCC approach is applied to obtain the cooling behavior and resulting thermal history of every fruit packed inside a pallet of fruit (Figure 1c) throughout the complete refrigerated supply chain. From that, the associated quality evolution is extracted for each cold chain (Figure 1d).

In a second step, on the basis of the VCC model, the energy efficiencies for cooling orange fruit in a precooling facility, in a refrigerated container during maritime transportation, and in a cold store are estimated for the three different box types, which are required as an input for LCA. For this purpose, the energy coefficient (EC) is used to quantify the energy consumption of cold chain operations (Figure 1e). The EC represents the heat that has to be extracted from the fruit (in kJ) per kJ of electricity that is consumed to achieve this goal. It was defined originally for entire cooling facilities [42] and is defined as:

$$EC = \frac{Mc_p\left(T_i - T_f\right)}{E_e c} \tag{1}$$

where M is the total mass of all produce that is cooled per month [kg mo^{-1}], c_p the specific heat capacity of the produce [kJ kg^{-1} K^{-1}], T_i the product temperature at the start of the cold chain [K], T_f the product temperature at the end of the cold chain [K], E_e the consumed electricity per month for operating the facility for fruit cooling [kWh mo^{-1}], and c is 3600 kJ kWh^{-1}. The calculation of the EC, which is based on the VCC model, is detailed in Supplementary Material 1. In conventional LCA, the energy use is assumed to increase linearly with time for a specific unit operation [21], that is, the required power is assumed to be constant. A main merit of

combining VCC with LCA is that package-specific ECs could be determined via the VCC method, together withmore accurate values for each unit operation.

In a third step, this energy consumption data, via the EC, as well as more detailed package-specific data (dimensions, fruit capacity, material weight) (Figure 1f) are fed into the LCA model. LCA is subsequently used to quantify the environmental footprint of the different package designs and cold chain scenarios (Figure 1g). By applying this strategy of linking the LCA and VCC methods, a unique insight is provided into the thermophysical behavior of each single fruit in the cargo, together with a more detailed environmental impact quantification than is currently possible with the conventional LCA.

2.3 <u>VCC modeling of fruit cooling and quality evolution</u>

The VCC method was presented recently [15,19], and only its key features are highlighted. The VCC method evaluates the thermal evolution of a pallet of fruit during convective cooling throughout the cold chain. To this end, each unit operation of each cold chain scenario is calculated sequentially with CFD (computational fluid dynamics, Figure 1c). Although these models are in some way a simplified representation of reality, they capture the differences in cooling kinetics for the individual fruit between the different unit operations. In that way, the temperature history of each single fruit inside the pallet is quantified throughout the complete postharvest cold chain. Using the data of the temperature of each fruit, a kinetic rate law model is used to predict the temperature-dependent fruit quality evolution (Figure 1d), as detailed below. In that way, heterogeneities in differential cooling behavior between fruits in the pallet and the resulting quality can be identified.

For the CFD simulations, computational models of a pallet of orange fruit (spheres with diameter 75 mm) and the surrounding air domain are generated for each unit operation. Each single fruit inside the pallet is modeled explicitly. The geometrical details (vent openings) of the ventilated package design are also explicitly included (Figure S1 and S2). A pallet contains 5120, 5120 and 3900 fruit for the Standard, Supervent, and Opentop package, respectively. The computational models are meshed with 40 million tetrahedral control volumes for each computational model. The boundary conditions for the airflow rate and delivery air temperature are defined on the basis of commercial practices, and are specified in Table S1. Precooling is characterized by horizontal flow at high airflow rates, refrigerated transport by vertical flow at moderate airflow rates, and cold storage by horizontal flow at low airflow rates.

The CFD simulations are executed with the software OpenFOAM 2.4.0. Turbulent airflow and heat transport through the ventilated package are solved, as well as heat transport inside the fruit and package. The air and solid domains convectively exchange heat via the boundary layer. To this end, the Reynolds-averaged Navier-Stokes (RANS) equations are solved together with the shear stress k-ω turbulence (SST k-ω) model (Menter, 1994) and wall functions. The current CFD model was validated on multiple occasions times by the authors for fruit cooling [11,43]for the same turbulence model and a similar geometrical model as used in the present study. All the details of the validation procedures can be found there. A good agreement with experimental data was found.

Fruit quality evolution is modeled by means of a kinetic rate law [3,44]. Such a model quantifies the change of a particular quality attribute A over time, for instance, vitamin content, and is temperature-dependent:

$$\frac{-dA}{dt} = kA^n$$

(2)

$$k(T) = k_0 e^{\frac{-E_A}{RT}} \tag{3}$$

where k is the rate constant [s^{-1}], n is the reaction's order (0 in this case, zero order), t is the time [s], k_0 is a constant [s^{-1}], E_A is the activation energy [J mol^{-1}], R is the ideal gas constant (8.314 J mol^{-1} K^{-1}) and T is the temperature (absolute, [K]). To include the dependency of quality decay to the temperature, the rate constant k is made a function of temperature (Eq. (3)), for which typically an Arrhenius relationship is used [3,44]. This temperature is, for example, the core temperature or the volume-averaged fruit temperature. To calculate $k(T)$, k_0 and E_A were calibrated on the basis of quality decay data, and are both assumed to be independent of temperature. In this study, the model was calibrated for A being the overall fruit quality. A quality of 0% implies that the shelf life is completely lost. From literature, we assume that the quality of the orange fruit is completely lost after 56 days of storage at 4 °C [45], thus A_{end}(56 d, 4 °C) = 0%, or a shelf life of 56 d. Such data is typically obtained by shelf-life experiments at a certain temperature. For model calibration, information on the temperature dependency of the rate constant is also required. This information is determined via the Q_{10} value:

$$Q_{10} = \frac{k_{T+10}}{k_T} \tag{4}$$

where k_T and k_{T+10} are the rate constants at temperatures T and T+10K. Van't Hoff's rule states that the rate of a biological reaction doubles or triples for every 10°C rise in temperature [2]. As such, the Q_{10} value is about 2-3 for fruit degradation reactions [2,3]. In this study, a Q_{10} value of 2 was chosen. This implies that an increase in temperature of 10°C doubles the rate constant, so halves the time until the fruit is lost, if stored at a constant temperature. This means that in our study, the citrus fruit can be stored for approximately 28 d at 14 ºC. Note that the model above

was explicitly calibrated on the basis of experimental data, so no validation is required in this case.

More details on the CFD simulations and the fruit quality model are specified in Supplementary Material 1. Note that the VCC simulation data used in the present publication were presented as a part of a larger simulation study on ventilated package design and cold chain scenarios [46], where more details can be found.

2.4 Environmental impacts by LCA

The second part of the methodology consists of a life cycle assessment (LCA) of the citrus fruit supply chain and fruit waste treatment, using the software SimaPro 8.3 [47]. Life cycle assessment starts from inventory data of specific supply chains, including orange fruit growing, fertilizer, pesticides, machinery inputs, electricity and fuel consumption of the different unit operations, the material consumption for packaging, storage, transport distances and the means of transport, the amount of food waste, and the treatment method of the wasted fruits (in regard to food waste from agricultural production to composting, and from trade and retail to anaerobic digestion). In this study, LCA receives input from the VCC simulations on the energy use of different unit operations and for different packages. The other output of the VCC simulations, namely, the fruit quality loss, was not used in LCA for predicting the resulting amount of food waste at this stage, but is a focus of our future research. Hence, the food waste amounts at different stages in the food supply chain are based on the average estimations by Beretta et al. for Switzerland [26]. The datasets used for the background processes of the life-cycle inventory are based on the LCA databases ecoinvent 3.2 ("allocation recycled content") [33] and the World

Food LCA Database 3.0 [34]. The functional unit of the various cooling scenarios to be compared was defined as 1 kg of orange fruit at the retailer, ready to be sold in Switzerland.

On the basis of the life-cycle inventory data, LCA calculates the climate change impacts with the *global warming potential 100a* method [48]. The impacts are expressed in kilogram CO_2 equivalents. In addition, in Supplementary Material 3, the aggregated environmental impacts are analyzed with the *ReCiPe* method [49]. A list of the datasets and their functional units is provided in Table S5 in Supplementary Material 2, which also contains information on how energy consumption, electricity generation and food waste are implemented.

3   RESULTS AND DISCUSSION

### 3.1   Cooling behavior of cooling operations and package designs

From VCC computations, the cooling of fruit in each unit operation is quantified via the seven-eighths cooling time (SECT) for all package designs. The SECT ($t_{7/8}$) is the time that is needed to reduce the difference between initial-fruit and cooling-air temperature to seven eighths of the initial temperature difference. The SECT is often applied in commercial cooling processes because when it is reached, the fruit is almost at the targeted storage temperature [50]. We use the fruit pulp (core) temperature to evaluate the cooling progress by determining the SECT for each individual fruit. We do this because the core (pulp) temperature is the last position in the fruit where the target temperature is reached. As such, this core pulp temperature is often measured in various commercial operations to monitor cooling processes, which is typically performed by placing a point probe in the fruit pulp.

In Figure 2, the average SECT of all fruit inside a pallet is given for all unit operations and package designs, together with the standard deviation. This standard deviation is calculated on the basis of all SECT values of the individual fruit. In Figure 3, the SECT values, averaged over each box in the pallet, are shown for the Supervent package for the three unit operations. Each box is represented by a colored dot, boxes are depicted in Figure S2.
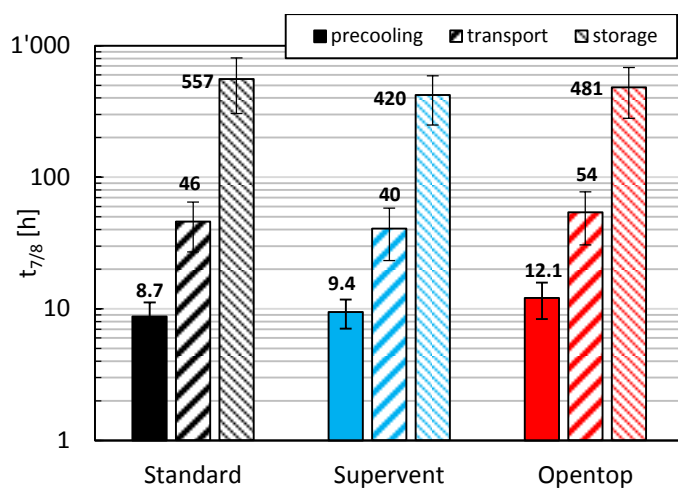
Figure 2. Seven-eighths cooling time of the individual fruits for different cold chain unit operations and package designs: average value of an entire pallet (also depicted quantitatively) and standard deviation (logarithmic scale).
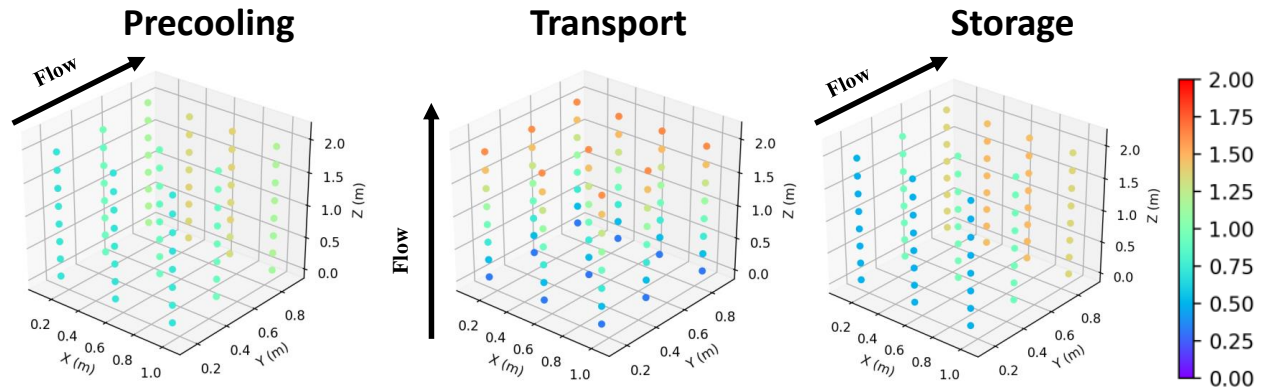
Figure 3. Seven-eighths cooling time (SECT) for a pallet of Supervent packages for all cooling operations (scaling is done with the total SECT for the entire pallet for that cooling operation, which is $SECT_{avg}$). Each colored point indicates the $SECT/SECT_{avg}$, averaged over each single box (Figure adjusted from [46]).

For precooling, Standard and Supervent packages cool quite similarly. Opentop on the other hand cools slower and in a less uniform way (Figure 2, 3). The slower and more heterogeneous cooling of Opentop is intuitively surprising as this package has the highest area of vent openings on its long as well as short side (see Supplementary Material 1, Figure S1), compared to the Standard and the Supervent packages. This finding is attributed to the vent opening configuration, where these are distributed not so homogeneous on both the long and short sides, in comparison to the other two packages. This non-uniform distribution of vent holes introduces preferential pathways. As an example, cold airflow is directed mainly over the fruit that is placed in the top layer, which induces preferential cooling here (Figure S1). Thus, the fruit at the bottom of the package cools more slowly. Furthermore, the air speeds are lower for Opentop due to its lower density of fruit packing [46]. During refrigerated transport, the Supervent package outperforms the others. This is mainly due to the optimal vent opening configuration, namely

openings located along the edges of the carton. Thus, aligned ventilation channels for cold air are present in the vertical direction along the sides of the package. Since there is also a central vent opening at both the bottom and top surfaces, uniform fruit cooling is achieved. For refrigerated storage, Supervent also performs better than the other cartons. As such, it is, in an overall sense, the carton that provides the most rapid and homogeneous cooling of the fruit.

From Figure 3, the spatial non-uniformity of fruit cooling in each pallet is clearly distinguishable for each unit operation. Precooling (meaning high air speeds) clearly provides better cooling uniformity when comparing individual boxes, compared to refrigerated storage (low air speeds). This indicates that the complete pallet is more uniformly cooled at elevated airflow rates. The closer the box is to the inlet, and thus upstream, the faster the cooling is for all carton types. The boxes cool progressively slower with increasing distance from the inlet, i.e. when they are located in more downstream positions in the pallet.

3.2 <u>Reduction in fruit quality for various cooling scenarios and package designs</u>

Using the sequential thermal history of the various cooling operations presented, the reduction in fruit quality is determined within the pallet. This is done for the aforementioned postharvest scenarios for each box design. To this end, the volume-averaged fruit pulp temperature within the full pallet is used instead of the single fruit core pulp temperatures, as this gives a better approximation of the general quality evolution. In Figure 4a, this volume-averaged (pallet-based) fruit temperature is shown for all package designs to illustrate the temperature history evolution. In Figure 4b, the corresponding fruit quality evolution is given.

When comparing the postharvest scenarios, forced-airflow cooling as well as ambient loading exhibit a quite similar reduction in fruit quality. One reason is that citrus fruit is quite resilient and has an inherently long storage life, so that the fruit-quality-decay timescales are much higher than the ones for cooling. Thus, the enhanced quality achieved by a more rapid cooling by using precooling, in comparison to refrigeration inside the container, does not significantly affect the quality reduction of citrus fruit. However, since the postharvest chain with ambient loading has a reduced duration, the final quality is higher compared to the forced-airflow cooling chain. Ambient loading, with its clear logistical advantage, therefore also results in enhanced fruit quality, which also implies less food losses. This possibly can also increase the marketing time window. Ambient cooling, however, results in a larger quality loss in comparison to the two other cold chains. This is related to the long cold storage period at higher fruit temperatures, as a result of which the cooling rates are much lower. As such, this practice is not recommended, but is often the only option due to the limited access or availability to precooling facilities in some fruit supply regions. Ambient cooling will however induce higher quality loss, thus food waste, compared to forced air precooling. This cold chain scenario will also imply a larger economic impact for the retailers, who will have to import a larger amount of fruit to have the same net supply for their customers. However, the quality loss can also remain invisible throughout the cold chain for a resilient species such as orange fruit. This is the case if ambient cooling does not necessarily lead to additional food losses in the cold chain, but just results in a reduced number of shelf life days for the consumer. This invisible quality loss can however lead to increased food waste in households.

The differences in timescales in the cooling process and fruit quality decay also explain the fact that differences between carton designs are rather limited. For all cold chains, Opentop

exhibits the lowest quality due to the overall worst cooling behavior over all unit operations (Figure 2, 3). For other fruit species, such as berries, avocado, or mango, which are more sensitive to temperature-driven quality loss, the differences between package designs or cold chain scenarios are expected to be more pronounced.
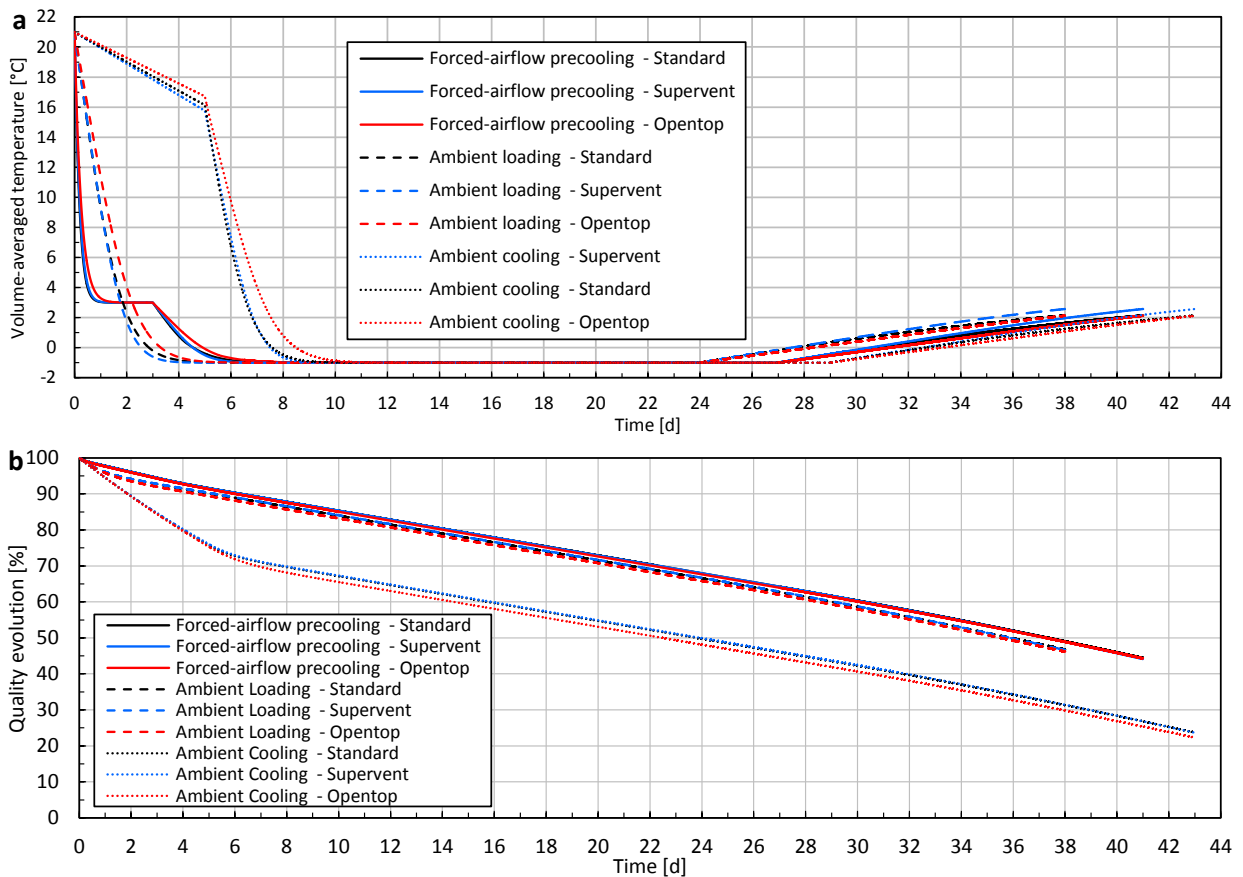
Figure 4. (a) Volume-averaged temperature of all palletized fruit as a function of time for various cold chain scenarios and package designs (Figure adjusted from [46]); (b) corresponding fruit quality evolution in the pallet as a function of time. The remaining overall quality is depicted, where the initial quality was 100% and where the fruit is considered to be lost when the quality level reaches 0%.

3.3 <u>**Environmental impact of different cold chain scenarios and package designs**</u>

Using LCA, the environmental impact is quantified for all three package designs and the three cold chain scenarios. The climate impacts are represented in Figure 5 in grams CO_2 equivalent per kg of fruit [48] and are split up into various processes of the supply chain. For processes that have the same climate impact for each cold chain scenario, no explicit value is quantified in Figure 5. As a fourth cold chain scenario, the use of solar energy to precool the fruit is also shown. In Figure 6, the differences with the base case (i.e. forced air precooling for the standard package) are quantified for each process in the supply chain to facilitate comparison.

3.3.1 *Package designs*

A comparison of package designs reveals that the Supervent box has the lowest carbon footprint for all cold chain scenarios, although the Standard box follows quite closely. The reasons for this superior performance, compared to the Standard box, are the following:

- For the precooling unit operation, the energy coefficient is higher (EC = 0.41 vs 0.40 kJ heat removed/kJ of electricity consumed, Table S2) so that Supervent boxes exhibit a lower carbon footprint (57 instead of 58 g CO_2-eq/kg of fruit).

- For cooling down products in a refrigerated container, the energy coefficient is also higher for Supervent (Table S2). This cooling down is assumed to occur during lorry transport, whereas fruit are assumed to arrive already cooled in the ship. The differences in energy consumption between the packages, however, originate only during the initial phase of cooling in the container. After the fruit are cooled to the SECT, maintaining a constant interior temperature leads to equal energy consumption for all the packages. The reason is that this energy consumption depends mainly on the heat lost through the container's

exterior walls, which does not depend on the package design. As such, the carbon footprints for lorry cooling are very similar (23/72/57 g CO_2-eq/kg for Supervent vs. 23/73/59 g CO_2-eq/kg for Standard for forced-airflow precooling/ambient loading/ambient cooling).

The Opentop package has a much larger environmental impact compared to Standard (and Supervent) packages for the following reasons:

- For the precooling unit operation, the energy coefficient is lower (EC = 0.36 vs 0.40 kJ heat removed/kJ of electricity consumed) and therefore Opentop boxes exhibit a higher energy consumption for precooling (65 vs. 58 g CO_2-eq/kg).

- For cooling down products in a refrigerated container, which is assumed to happen during lorry transport, the energy coefficient is also lower for Opentop (EC = 0.27 vs. 0.40 kJ heat removed/kJ of electricity consumed).

- For cooling during transport (both ship and lorry), the fruit packing density is lower in Opentop packages (and thus in a pallet), due to the free open space that is present above the fruit in each package. As such, the amount of fruit per pallet is 3900 (60 x 65) instead of 5120 (64 x 80), which is 24% lower. Thus, the environmental impact for cooling is higher as more refrigerated containers (on ships) and lorries are needed to transport the same amount of fruit (205/28 vs. 156/23 g CO_2-eq/kg for ship cooling/lorry cooling).

- For transport, more trucks with Opentop boxes are needed in order to transport the same amount of orange fruit than with standard boxes, leading to a higher carbon footprint (91 vs. 80 g CO_2-eq/kg). For ships, however, we assume that their load factor is limited by weight and not by volume, because of which the same value is used for all packages.

- With respect to the packaging material, Opentop boxes contain 40% less carton and are thus lighter than Standard boxes, per kg of fruit. This is a beneficial effect and actually reduces the carbon footprint of the package part (47 vs. 76 g CO_2-eq/kg) which however is more than offset by the additional cooling and transport energy requirements (see above).

Significant differences between ventilated carton designs are found, with a maximal difference of 63 g CO_2-eq/kg, namely between Supervent and Opentop for the ambient loading scenario. As such, the Supervent package provides a reduction of 10% in the total carbon footprint of the supply chain (relative to that of Opentop), which is substantial.
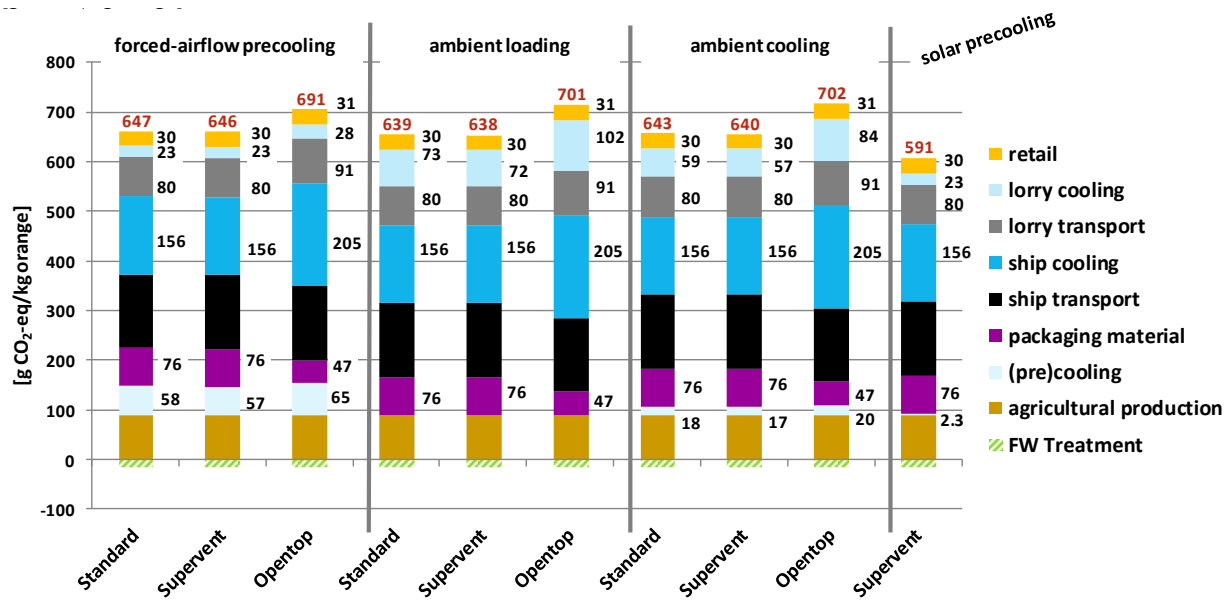
Figure 5. The calculated environmental impact (grams CO_2 equivalent per kg of fruit) of all package designs (Standard, Supervent, Opentop) and cold chain scenarios, split up into the different processes of the supply chain and food waste (FW) treatments. The impacts include the present amounts of food waste generated between agricultural production and retail, but exclude household food waste.

| g CO$_2$-eq/kg | forced-air precooling | | | ambient loading | | | ambient cooling | | | solar precooling |
Cold chain: Box type:	standard	supervent	opentop	standard	supervent	opentop	standard	supervent	opentop	supervent
(pre)cooling	0	-1	6	-58	-58	-58	-40	-41	-38	-55.8
packaging material	0	0	-29	0	0	-29	0	0	-29	0
ship cooling	0	0	49	0	0	49	0	0	49	0
lorry transport	0	0	11	0	0	11	0	0	11	0
lorry cooling	0	0	5	50	49	79	36	34	61	0
retail	0	0	2	0	0	2	0	0	2	0
total	0	-2	44	-8	-9	54	-4	-7	55	-56

(baseline: forced-air precooling)

Figure 6. Differences of environmental impact (grams CO$_2$ equivalent per kg of fruit) of the cold chain and package scenarios with the baseline scenario (forced-air precooling with standard box) for different operations in the supply chain.
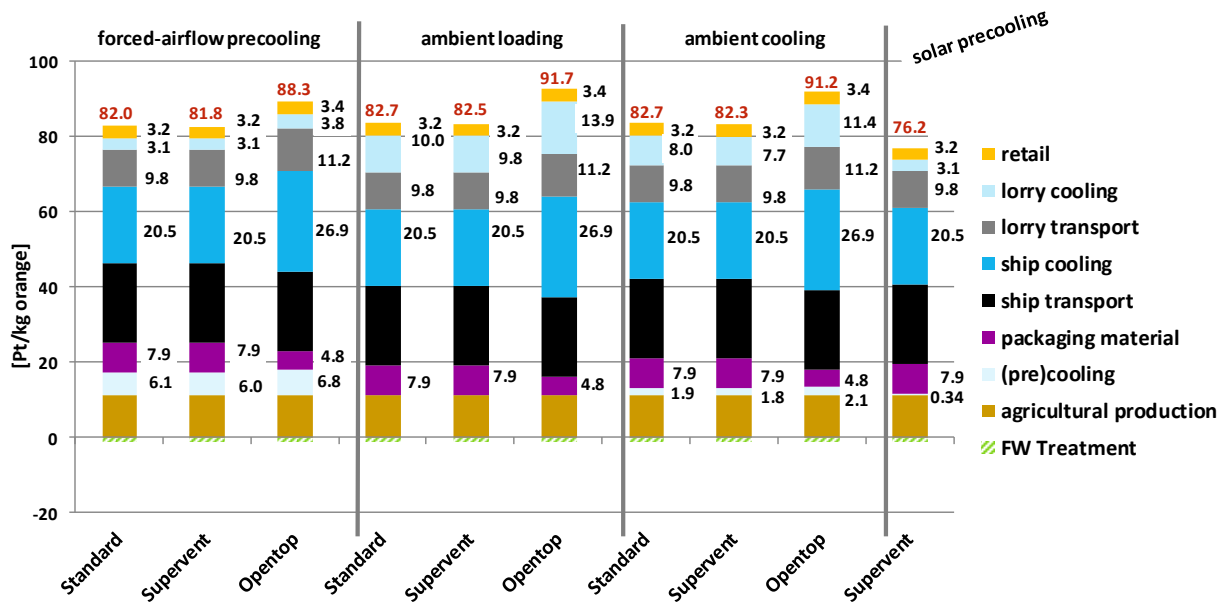
3.3.2 Cold chain scenarios

When comparing the three cold chain scenarios, ambient loading has the lowest environmental impact, except for Opentop packages. This lower impact is caused by the simple fact that for ambient loading, lorry cooling (during which the fruit are cooled down entirely in the container) caused lower greenhouse gas emissions than precooling plus lorry cooling for the forced-air precooling scenario. The reason is that South Africa's electricity mix, used for the precooling facility, is more carbon intensive than electricity generated for container cooling, which is done with an 18kW diesel-electric generating set (genset) that cools the refrigerated container. The South African electricity mix has a particularly high dependence on coal. The shorter cold chain for ambient loading also contributes to a reduction of the impact, but this effect is much smaller. The higher environmental impact of ambient loading for Opentop packages, however, is due to the lower energy coefficient in the container, compared to the precooling facility.

The environmental performance of ambient cooling lies between the other cold chain scenarios for Standard and Supervent, because the cooling is partly driven by electricity and partly by the diesel-electric generating set as fruit are partially cooled in the cold store and partially in the container. As the fruit are loaded warmer in the refrigerated container (on the lorry) than for

forced-air precooling, the lorry cooling also consumes more energy during ambient cooling. However, the quality loss is much higher in the ambient cooling scenario, as identified via the virtual cold chain method (Figure 4). Significant differences between cold chain scenarios are found (Figure 5 and 6), with a maximal difference of 11 g CO_2-eq/kg, namely between forced-air precooling and ambient cooling for the Opentop package. As such, Opentop packaging provide a reduction of 1.6% in the total carbon footprint of the supply chain (relative to that of forced air precooling), which is rather limited.

Figure 7 shows the environmental impacts calculated with the aggregated life cycle impact assessment method ReCiPe, which considers 17 environmental mechanisms (Goedkoop, 2013). Besides global warming, the method includes environmental mechanisms as water and land use, freshwater eutrophication as well as toxicity, which are relevant mechanisms in most agricultural systems. ReCiPe also includes stratospheric ozone depletion, which is relevant in refrigeration systems (for results see Figure S4 in Supplementary Material). Figure S4 shows that the general pattern for aggregated ReCiPe impacts is the same as for climate change and stratospheric ozone depletion, meaning that our conclusions with respect to packages type and cold chain scenario are valid for different environmental impact mechanisms.

With these results, we have made a step forward compared to the previous state of the art on combining fruit quality, energy use and global warming impact of food cold chains [40]. The previous study only analyzed the impact category "climate change", but we also show results for "stratospheric ozone depletion" (Figure S4) and aggregated environmental impacts according to the impact assessment method "ReCiPe".

Figure 7. Environmental impact (ReCiPe Pt per kg of fruit [49]) of all package designs and cold chain scenarios, split up into the different processes of the supply chain.

3.3.3 Agricultural production

The results of Table 4 indicate that the climate impacts of agricultural production account for between 10% and 15% of the climate impacts of the final product. This is not typical for food product LCAs, where agricultural production is the largest contributor to greenhouse gases for many types of products [27,51]. For example, roughly 75% of the impacts of German food consumption are caused by agricultural production and land use changes, whereas the rest is caused by processing, transport, storage, and packaging. However, in the case of fruit production in areas with relatively low fertilizer and pesticide inputs, the impacts of agricultural production are much lower (80 g CO_2-eq/kg of orange from South Africa) than for average products from

more intensive crops and animal production (2900 g CO_2-eq/kg average product consumed in Switzerland according to [26]). The impacts of the cold chain from Africa (especially transport and cooling) are much higher than for local products and products that do not need cooling. These results demonstrate that cold chains can be the most important contributor to the climate footprint of food products.

3.3.4 *Environmental impact of different sourcing regions*

The possible climate change impact of different fruit sourcing regions, namely South Africa to Switzerland versus Spain to Switzerland, is compared in Figure 8 for the Supervent box. The South African cold chain clearly has a larger environmental impact due to the additional contributions of ship transport and the associated cooling, even though the lorry transport contribution is a little lower. However, the total difference between South Africa and Spain is surprisingly small. This is attributed to the much higher impact of agricultural production in Spain. The contribution of agricultural production in both countries includes carbon footprints associated with fuel consumption of tractors, infrastructure, irrigation, planting, harvesting, etc. In this case, particularly the use of fertilizers and pesticides in Spain (see also Table S3), as well as the increased irrigation explain the differences. Further verification is needed to identify to what extent these differences are representative for exports from Spain and South Africa to Switzerland, or if they only relate to agricultural practices of domestic production. Nevertheless, it indicates the need for promoting agriculture with a lower environmental impact in this region. Furthermore, there is a need for more detailed and regionalized data on agricultural production of orange fruit in both Spain and South Africa.
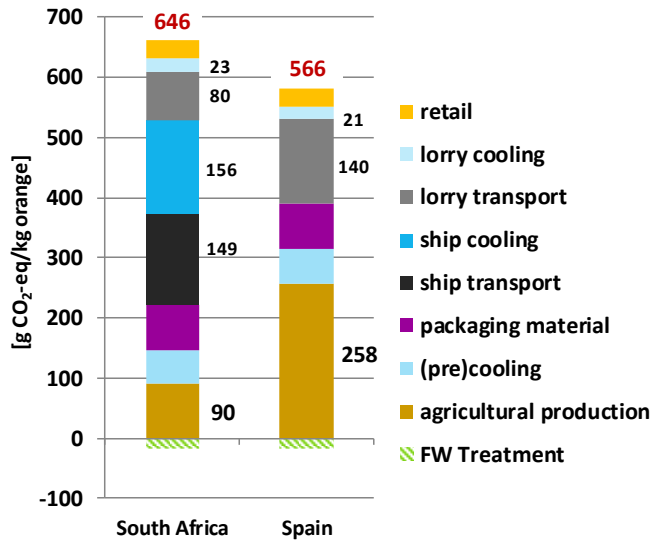
Figure 8. Climate impact (grams CO_2 equivalent per kg of fruit) of Supervent packages for two different fruit sourcing regions (Spain and South Africa), split up into the different processes of the food supply chain and food waste (FW) treatments.

3.4 **Optimal combination of packages and cold chain scenario**

By combining the information generated from the VCC simulations, on fruit cooling and quality, with that of LCA on environmental impacts, the best combination of package design with the cold chain scenario is identified. Using the present energy mixes, ambient loading of citrus with the Supervent box showed the best performance. Despite its large potential to provide good final fruit quality as well as a low environmental impact, this combination is only explored sporadically in the South African citrus export industry. As ambient loading does not require additional hardware investments, but just altered logistics, it can be implemented very swiftly in existing cold chains. This flexibility makes ambient loading (with Supervent boxes) a very attractive commercial option for the citrus industry.

Since the relatively high environmental impacts of cooling are related to the type of energy used, the use of solar energy to drive precooling is explored for the Supervent box (Figure 5) as an extra alternative. The results show that using solar energy provides an extra 55 g CO_2-eq/kg benefit, since the energy needed for precooling becomes almost climate neutral. Forced-airflow precooling also provides slightly better fruit quality than ambient loading (Figure 4). The implementation of solar panels to run the precooling facility requires additional hardware investments, but in the long term it can offset investment costs since electricity costs can be saved throughout the years. With this measure, by far the lowest environmental impacts can be achieved, notably even with the lowest fruit quality losses. The differences between forced air precooling with and without solar energy are thus significant (Figure 5 and 6), namely 55 g CO_2eq/kg for the Supervent package. As such, a reduction of 8.5% in the total carbon footprint of the supply chain can be achieved (relative to that of forced air precooling).

Finally, one needs to note that currently, the food quality information from the VCC method is not directly applied in the life cycle assessment calculation yet, but the authors are working towards this goal. However, the present results can already be linked to food losses. As an example, one could quantify by how much food losses need to be reduced in a specific cold chain to compensate for the higher climate impacts, as compared to another cold chain. For instance, the food losses in retail for forced-airflow precooling need to be reduced by at least 36% (i.e., 3.2% instead of 5.0% of purchases) for it to have the same carbon footprint as ambient loading. The quality benefits from cooling down the products more quickly at the start of the cold chain by using precooling is unlikely to reduce retail losses by more than 36%. Therefore, ambient loading is probably a more environment friendly option so far. However, if we assume that the differences in the remaining fruit quality for the different cold chains do not influence

only retail losses, but also losses at the household level, a reduction by 4-5% of household food losses (24.6% vs. 25.7% of purchases) is enough to compensate for the additional environmental impacts of precooling, compared to direct container loading with a diesel-electric generating set. However, the best overall option is clearly precooling powered by solar energy, since not only environmental impacts, but also quality losses are minimized.

4 CONCLUSIONS and OUTLOOK

The combination of life cycle assessment with virtual cold chains enabled, in a unique way, the identification and quantification of trade-offs between maintaining fruit quality and reducing environmental impact. This is essential information of which importers, exporters, container manufacturers and retailers can benefit, since these stakeholders often have different and conflicting interests. Retailers prefer to receive fruit with a maximal quality and shelf life. Container manufacturers, on the other hand, focus more on making their containers more energy efficient during transit [52]. This can be achieved by reducing internal air circulation, which however could negatively impact fruit quality in some cases. Such trade-offs have not been quantifiable so far by a lack of a more holistic approach combining environmental science with food engineering and mechanical engineering

As an example of a typical trade-off, ambient cooling showed a lower environmental footprint than forced-airflow precooling, but exhibited a much larger quality loss. By relying only on the life cycle assessment results without considering fruit quality, retailers and policy makers would be advised to opt for ambient cooling. This would however have significant impacts on fruit quality losses and the amounts of food waste as well as a reduced shelf life for the consumers. The combination of information of both methods will result in an improved decision making process based on a more holistic view of all the factors relevant to the fruit cold chain. In the

same way, this approach enables even more to push promising cold chain protocols forward, for example ambient loading. By quantifying remaining quality as well as energy consumption, diferent stakeholders can be better convinced to put these strategies into practice.

Apart from identifying trade-offs, our pioneering method enriched life cycle assessment with more customized input data from multiphysics modeling, and at the same time assessed food quality evolution. As illustrated in the present study, the holistic assessment could help different stakeholders in the perishables supply chain to choose the most optimal package and cold chain scenario to make their food supply chains more sustainable, and to optimize logistics. Significant differences between ventilated carton designs (63 g CO_2eq/kg) and cold chain scenarios (11 g CO_2eq/kg) were identified, or 10% and 1.6% of the total environmental impact of the supply chain, respectively. If solar electricity is used for precooling, the environmental impact was lowered by 55 g CO_2eq/kg of fruit (or 8.5%), while still providing similar fruit quality retention of the fruit.

As a future outlook, the virtual cold chain method should be extended to quantify the actual food losses in the cold chain on the basis of the thermal history of the products. The relation between the thermal history and the food quality evolution toward food loss amounts could be determined empirically for this purpose. By using the virtual cold chain-based input of food losses in life cycle assessment, both methods could be coupled more closely to evaluate the overall environmental performance of different cold chains.

Generally, different impact assessment methods in life cycle assessment (climate impacts, acidification, eutrophication, aggregated environmental indicators, etc.) can lead to diverging conclusions, depending on how different impact categories are weighted [53]. We showed additional results of aggregated environmental impacts in Supplementary Material. However, in

future studies, different impact categories should be analyzed separately (e.g. eutrophication, water scarcity, land use impacts, aquatic ecotoxicity). Furthermore, continued efforts are required to close the data gaps in life cycle assessment. As illustrated with the comparison of fertilizer and pesticide application in Spain and South Africa (Figure 8), agricultural practices can have a large influence on environmental impacts. Individual case studies are therefore not necessarily representative for the comparison between different countries. Larger datasets in various parts of the country are needed that differentiate agricultural production for domestic consumption and for export. Another point would be to evaluate the environmental impacts of reusable plastic boxes instead of recyclable corrugated cardboard boxes. In a recent study [54], it is mentioned that a durable reusable box is often a better choice compared to a recycled box. However, under specific circumstances, a recycled product can also be a good option, if a profitable and effective recycling system is implemented. In their case study, a recyclable corrugated cardboard box system was a more eco-friendly option than a reusable plastic crate system for bread deliveries.

The current study was performed for citrus fruit, which is quite a resilient species with a rather long shelf life. The differences in fruit quality loss between different cold chain scenarios and package designs are expected to become even more pronounced for more perishable species, such as berries or mango fruit. The increasing globalization of supply chains makes interdisciplinary approaches such as the one presented here even more timely.

The methods provided in this paper can also be applied for related application areas, optimizing the logistics of agricultural products and lowering food waste and environmental impacts. A typical example is the use of LCA for evaluating conversion of food waste into biogas by recycling [28,29]. Here, mechanistic modeling could help optimizing different unit

operations, such as the dehydration process for example, and thereby enrich LCA input data. Such work can also be linked to optimization of supply chains for bioenergy feedstock [55,56]. Furthermore, the applied methodology could also be applied for use of renewable energy in distribution networks of perishables [30].

ASSOCIATED CONTENT

The following files are available free of charge.

- Supplementary Material on the virtual cold chain method, life cycle assessment and additional figures. (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. The CRediT roles are specified:

T.D. conceptualized the study and did project administration; T.D. and S.H. wrote the project proposal and secured the funding; W.W., T.D. and C.B. developed the methodology; W.W. performed the CFD simulations and C.B. performed the LCA simulations; W.W., T.D. and C.B. performed data collection, analysis, interpretation and visualization of the results; P.C. collected data on the citrus cold chain; T.D., S.H. and P.C. performed supervision of W.W and C.B; T.D.

wrote the original draft with key input from W.W. and C.B; S.H. and P.C. critically reviewed and edited the manuscript, and T.D.,W.W. and C.B revised the manuscript on the basis of these suggestions.

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