Validation of GreenH\textsubscript{2}armony\textsuperscript{®} as a Tool for the Computation of Harmonised Life-Cycle Indicators of Hydrogen

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Abstract: The Life Cycle Assessment (LCA) methodology is often used to check the environmental suitability of hydrogen energy systems, usually involving comparative studies. However, these comparative studies are typically affected by inconsistent methodological choices between the case studies under comparison. In this regard, protocols for the harmonisation of methodological choices in LCA of hydrogen are available. The step-by-step application of these protocols to a large number of case studies has already resulted in libraries of harmonised carbon, energy, and acidification footprints of hydrogen. In order to foster the applicability of these harmonisation protocols, a web-based software for the calculation of harmonised life-cycle indicators of hydrogen has recently been developed. This work addresses—for the first time—the validation of such a tool by checking the deviation between the available libraries of harmonised carbon, energy, and acidification footprints of hydrogen and the corresponding tool-based harmonised results. A high correlation ($R^2 > 0.999$) was found between the library- and tool-based harmonised life-cycle indicators of hydrogen, thereby successfully validating the software. Hence, this tool has the potential to effectively promote the use of harmonised life-cycle indicators for robust comparative LCA studies of hydrogen energy systems, significantly mitigating misinterpretation.

Keywords: acidification; carbon footprint; energy footprint; harmonisation; hydrogen; life cycle assessment

1. Introduction

The current level of fossil fuel use in the energy sector raises significant sustainability concerns, e.g., on environmental issues such as greenhouse gas emissions [1]. Within this context, hydrogen is expected to play a major role in the path towards a clean, decarbonised energy system [2]. When compared to other fuels, hydrogen contains a high amount of energy per mass unit and avoids direct emissions of pollutants in the use phase. Thus, hydrogen is seen as a strategic energy carrier with potential uses across different sectors. However, it is not directly available in free form, but it needs to be separated from another feedstock (e.g., hydrocarbons, biomass, and water) through energy-intensive processes. To that end, a large variety of hydrogen production methods can be applied [3], which leads to the need for comparative analyses that check the suitability of a given hydrogen energy system from a life-cycle perspective. In this regard, the life cycle assessment (LCA) methodology [4,5] is widely applied to evaluate and compare the environmental performance of (generic) product systems, though LCA comparative studies are often affected by methodological differences that may hamper robust comparisons between case studies [6–10].

When focusing on hydrogen energy systems, numerous LCA studies are available in the current literature, with significant methodological differences between each other [11]. In fact,
a harmonisation initiative specific to hydrogen energy systems has been undertaken [12–15], leading to LCA harmonisation protocols for three relevant life-cycle indicators: carbon footprint (global warming, GWP) [12], non-renewable energy footprint (cumulative fossil and nuclear energy demand CED) [13], and acidification (AP) [14]. These harmonisation protocols have already been applied to a wide range of LCA case studies of hydrogen, resulting in libraries of harmonised life-cycle indicators of hydrogen for robust comparisons. Furthermore, in order to facilitate the harmonisation procedure —whose step-by-step application may result time-consuming—the web-based tool GreenH2armony® has recently been developed [16,17] for the computation of harmonised carbon, energy, and acidification footprints of hydrogen according to the available protocols [12–14]. This work addresses—for the first time—the validation of such a tool for the computation of harmonised life-cycle indicators of hydrogen, thus enhancing the usefulness of comparative LCA to support decision-making processes [18,19]. In this sense, the ultimate value of this work is to enable robust decision-making on hydrogen production systems by making the use of current LCA harmonisation protocols practical.

2. Materials and Methods

The harmonisation protocols defined and applied in Valente et al. [12–14] allow the mitigation of the misinterpretation risk associated with inconsistent methodological choices in comparative LCA studies of hydrogen energy systems. The current libraries of harmonised life-cycle indicators of hydrogen report the harmonised carbon, energy, and acidification footprints of conventional (fossil) hydrogen from steam methane reforming (SMR) as well as of a large number of renewable hydrogen alternatives. The goal of this article is to validate the software GreenH2armony® through its one-to-one application to the case studies included in the available libraries [12–14], subsequently checking the deviation and correlation between the library- and the tool-based harmonised results. In this sense, low deviation and high correlation are required to pass the validation process.

2.1. Methodological Background: Main Features of the Harmonisation Framework

The protocols currently available focus on the harmonisation of methodological choices on the life cycle impact assessment method, system boundaries, functional unit, multifunctionality approach, inclusion of capital goods, and final hydrogen conditions (pressure, temperature, and purity) [12–14]. As regards the life cycle impact assessment method, the harmonisation of carbon footprints requires the inclusion of at least CO₂, N₂O, and CH₄ emissions and the use of IPCC-based characterisation factors with a 100-year horizon [20]. In case of harmonising energy footprints, they have to be quantified as the sum of fossil and nuclear energy demand, while the harmonisation of acidification requires the use of a CML-based method [21].

Figure 1 shows the main subsystems involved in a harmonised hydrogen energy system [12–14]. The life-cycle impacts are harmonised to a common functional unit of 1 kg of hydrogen at 20 MPa and 25 °C, following a cradle-to-gate approach that covers up to the compression stage. Hence, all the steps needed up to the compressed pure hydrogen, regarding both the foreground and the background level, are embedded in the harmonised energy system. Capital goods are included in the harmonised system’s boundaries. In case of systems presenting multifunctionality (i.e., performing other functions in addition to hydrogen production, e.g., electricity production), it is addressed according to a common scheme at the subsystem level: system expansion is applied when the hydrogen-related product represents the main function of the multifunctional subsystem, whereas an allocation approach based on economic values is followed when the hydrogen-related product represents a secondary function.

To date, the LCA harmonisation initiative has led to libraries of harmonised carbon, non-renewable energy, and acidification footprints of hydrogen for a large number of case studies. The purpose of these libraries is to provide values for robust comparison of hydrogen alternatives, mitigating the misinterpretation risk linked to LCA methodological inconsistencies. These libraries are expected to be enlarged by LCA practitioners willing to consistently compare their original hydrogen energy system(s) with those available in the literature. In this regard, the calculation of harmonised life-cycle impact assessment
indicators from a new LCA study may require a relatively large number of operations, and therefore the step-by-step (“by-hand”) application of the protocols may result time-consuming. Moreover, human factors such as wrong calculations, cumulated approximations and typos can compromise the estimation of harmonised life-cycle impacts.

Figure 1. Harmonised hydrogen energy system.

2.2. Main Features of the Software GreenH2armony® and Validation Procedure

The web-based tool GreenH2armony® has been developed to facilitate an appropriate application of the protocols for the computation of life-cycle indicators of hydrogen [16]. This tool is characterised by a user-friendly interface and it does not require additional calculations by the user, but only specific qualitative and quantitative information available in the original LCA study. According to the user’s guide of the software [17], its application involves the following requirements:

- The user must have an LCA study of a hydrogen energy system whose carbon, energy and/or acidification footprint the user is willing to harmonise.
- Such a study must be based on an attributional modelling approach and include a hydrogen production stage in the system’s boundaries.
- The user must be able to identify the hydrogen production technology involved, the hydrogen carrier and the driving energy.
- The user must know the functional unit used in the original study.
- The user must know the stages involved in the system’s life cycle.
- The user must know the original results for the indicators to be harmonised.
- When the system includes stages beyond hydrogen purification, the user must know the impacts specific to these additional stages.
- The user must be able to identify multifunctional subsystems and quantitatively define the multifunctionality approach originally followed.
- Regarding carbon footprint, the impact assessment method used in the original study must involve IPCC-based characterisation factors (100-year horizon; at least for CO₂, N₂O, and CH₄; kg CO₂ eq units).
- Regarding energy footprint, the impact assessment method must be based on the quantification (in MJ) of the sum of fossil and nuclear energy demand from a life-cycle perspective.
- Concerning acidification, the impact assessment method used in the original study must be CML-based and expressed in kg SO₂ eq.
The tool is targeted at LCA practitioners in the field of hydrogen energy, not necessarily being proficient LCA users. Overall, the guided procedure for the harmonisation of an LCA case study of hydrogen involves six sequential steps:

- In the first step, information regarding the core technology (reforming, electrolysis, etc.), the type of inputs (heat, electricity, and feedstock) and the reference year and region is requested.
- In the second step, the tool requires the original functional unit, the stages considered, and the impacts per functional unit.
- In the third stage, information regarding multifunctionality (if present) is requested for those subsystems in which multifunctionality takes place. If, according to the protocols, modifications to the original multifunctionality approach are needed, the tool asks for additional information about the amount of co-products, the original allocation factors (if applied), and the original impacts associated with the subsystem.
- In the fourth step, if needed, the tool requests quantitative information on the impacts associated with the life-cycle stages after hydrogen production (i.e., compression/liquefaction, storage, distribution, and use).
- In the fifth step, regarding the conditioning stage, information about the initial pressure (if known) and the type of electricity used for compression is collected. It should be noted that, to increase the applicability of the harmonisation procedure in the event of data scarcity, the tool offers the possibility of using default values at some specific points (e.g., infrastructure, electricity and feedstock impacts).
- Finally, in the sixth step, the tool requests qualitative information about capital goods and quantitative data for relevant inputs (amount of hydrogen carrier and/or driving energy).

Once the abovementioned procedure is completed, the software provides the user with the harmonised values for the life-cycle indicators addressed. When GWP and CED are not both addressed, the software also offers the possibility of estimating one indicator from the other using a correlation equation between both indicators [15]. Additionally, a report benchmarking the results against those of conventional hydrogen from SMR is generated. This report also provides the ranking of the new harmonised case study within the set of case studies archived in the database of the software.

With the aim of validating the software, all the case studies currently included in the libraries available in [12–14] were individually processed using GreenH2armony®. In this regard, Table A1 in the Appendix A presents the list of case studies of hydrogen included in the libraries [22–57], and therefore used herein to check the validity of the software. For the sake of full traceability of the validation exercise, Supplementary Information reporting all of the information fed to the software is provided on a case-by-case basis.

The procedure for the validation of the software consists in determining the deviation and correlation between the library-based harmonised indicators and the tool-based ones. If low deviation and high correlation (i.e., $R^2 > 0.99$) are found, then the software is considered to pass the validation exercise. Otherwise, the software is not deemed valid. Furthermore, if the software is successfully validated, a secondary outcome of the study refers to the identification of incorrect values within the published libraries of harmonised life-cycle indicators of hydrogen due to human factors affecting the “by-hand” application of the harmonisation protocols.

3. Results and Discussion

As a result of the validation exercise, Tables 1–3 present the harmonised impacts computed through the software GreenH2armony® (tool use illustrated in [58]), also including the harmonised impacts of hydrogen already available in the current libraries [12–14]. For those case studies where a significant difference between tool- and library-based harmonised values was found, the reasons behind such a deviation were explored by revisiting the calculations performed in the original harmonisation studies [12–14] (column “Comment” in Tables 1–3). As the computational structure of
GreenH₂armony® was programmed in a Microsoft IIS 8.5 environment on Windows Server 2012 R2 using MsSQL databases and classic Active Server Pages language and following the decision trees readily available in the original protocols [12–14], the reasons behind potential deviations should be associated with human errors affecting the literature-based results. Otherwise, computational bugs would lead to the invalidation of the proposed software solution.

Regarding the harmonised carbon footprints of hydrogen (Table 1), the values with: (i) a relative difference above 5% between the tool-based GWP and the library-based one and (ii) an absolute difference higher than 0.6 kg CO₂ eq per functional unit (i.e., 5% of the harmonised carbon footprint of the reference case, SMR2) were revisited in order to identify the origin of such a significant deviation. This led to the finding that all the case studies with a significant deviation (viz., HE4, HE1, PVE1, CSE2, WPE8, BMG6, WPE8, and BMG3) were affected by human errors that occurred when applying the protocols by hand, i.e. without using GreenH₂armony®.

Similarly, Table 2 presents the harmonised non-renewable energy footprints of hydrogen from both the available library [13] and the software used. As previously done for the carbon footprint, the cases with a significant deviation (relative difference above 5% and absolute difference higher than 10 MJ per functional unit [5% of the harmonised non-renewable energy footprint of the reference case, SMR2]) were revisited. Under this life-cycle indicator, only the case study SBR10 was found to be affected by a significant deviation, which is linked to a human error (viz., double counting of capital goods in the library-based value).

Table 3 reports the harmonised acidification impacts. As done for the previous life-cycle indicators, the case studies with a significant deviation (relative difference above 5% and absolute difference higher than 0.001 kg SO₂ eq per functional unit [5% of the harmonised acidification of the reference case, SMR2]) were revisited. This led to identify SBR4 and SBR5 as the case studies with a significant deviation. Once revisited, human factors were also identified as the source of deviation, in particular the misreading of numbers in bar diagrams when harmonising these case studies without GreenH₂armony®.

According to the results in Tables 1–3, the remaining harmonised carbon, energy, and acidification footprints (i.e., more than 90% of the harmonised values) were found to be associated with negligible deviations. These insignificant deviations were found to be closely linked to digit approximations cumulated through the harmonisation process. Overall, a very low deviation between the tool-based harmonised life-cycle indicators and the library-based ones was therefore concluded.

Besides the relevance of the difference between the library- and tool-based harmonised values, the importance of harmonisation to avoid misinterpretation is actually shown by the difference between the original and harmonised values. Previous harmonisation works reported mitigation of misinterpretation risk for the three life-cycle indicators under study [12–14]. Concerning carbon footprints, examples of misinterpretation were reported when ranking renewable options, e.g., PVE5 < WPE19 < CSE1 < BME1 according to the original values while BME1 < CSE1 < WPE19 < PVE5 based on the harmonised ones [12]. Regarding energy footprints, misinterpretation risk was illustrated when checking the achievement of a 40% energy-saving target with respect to conventional hydrogen: target attained by BMG8 but not by BMF5 according to the harmonised values, while target attained by BMF5 but not by BMG8 according to the original values [13]. Finally, regarding acidification, misinterpretation risk was found when comparing e.g., the cases SBR9, BMG1, BMG4, BMG5, and BMG7 with conventional SMR2 [14].
Table 1. Library- and tool-based harmonised carbon footprints of hydrogen (kg CO\textsubscript{2} eq per functional unit).

| Code    | Library-Based GWP | Tool-Based GWP | Error [Absolute] (Relative) | Comment          |
|---------|-------------------|----------------|-----------------------------|------------------|
| SMR1    | 12.95             | 12.85          | [−0.10] (−0.8%)             | Negligible error |
| SMR2    | 11.43             | 11.32          | [−0.11] (−1.0%)             | Negligible error |
| BMG4    | 0.18              | 0.05           | [−0.13] (−260%)             | Negligible error |
| HE2     | 0.77              | 0.77           | [0.00] (0.0%)               | Negligible error |
| BMG1    | 2.09              | 2.10           | [0.01] (0.5%)               | Negligible error |
| BMG2    | 4.40              | 4.36           | [−0.04] (−0.9%)             | Negligible error |
| SBR8    | 6.98              | 6.86           | [−0.12] (−1.7%)             | Negligible error |
| SBR9    | 7.22              | 7.14           | [−0.08] (−1.1%)             | Negligible error |
| WPE15   | 0.74              | 0.75           | [0.01] (1.3%)               | Negligible error |
| PVE7    | 3.22              | 3.23           | [0.01] (0.3%)               | Negligible error |
| WPE7    | 1.15              | 1.16           | [0.01] (0.6%)               | Negligible error |
| PVE2    | 2.59              | 2.61           | [0.02] (0.8%)               | Negligible error |
| WPE16   | 0.63              | 0.64           | [0.01] (0.9%)               | Negligible error |
| WPE18   | 0.81              | 0.81           | [0.00] (0.0%)               | Negligible error |
| WPE19   | 2.29              | 2.31           | [0.02] (0.9%)               | Negligible error |
| WPE13   | 0.85              | 0.85           | [0.00] (0.0%)               | Negligible error |
| RNE2    | 3.52              | 3.52           | [0.00] (0.0%)               | Negligible error |
| BMF1    | 4.51              | 4.52           | [0.01] (0.2%)               | Negligible error |
| BMF2    | 2.39              | 2.39           | [0.00] (0.0%)               | Negligible error |
| BMF3    | 4.96              | 5.02           | [0.06] (1.2%)               | Negligible error |
| WPE17   | 0.84              | 0.84           | [0.00] (0.0%)               | Negligible error |
| CSE1    | 2.20              | 2.20           | [0.00] (0.0%)               | Negligible error |
| PVE8    | 5.04              | 5.04           | [0.00] (0.0%)               | Negligible error |
| HE3     | 1.99              | 1.99           | [0.00] (0.0%)               | Negligible error |
| BME1    | 1.72              | 1.58           | [−0.14] (−8.9%)             | Negligible error |
| BMG8    | 10.47             | 10.49          | [0.02] (0.2%)               | Negligible error |
| BMF4    | 5.01              | 4.83           | [−0.18] (−3.7%)             | Negligible error |
| BMF5    | 7.36              | 7.19           | [−0.17] (−2.4%)             | Negligible error |
| BMF6    | 4.89              | 4.62           | [−0.27] (−5.8%)             | Negligible error |
| WPE1    | 1.08              | 1.06           | [−0.02] (−1.9%)             | Negligible error |
| WPE3    | 5.75              | 5.73           | [−0.02] (−0.3%)             | Negligible error |
| WPE2    | 0.97              | 0.99           | [0.02] (2.0%)               | Negligible error |
| WPE3    | 0.96              | 0.96           | [0.00] (0.0%)               | Negligible error |
| WPE4    | 0.96              | 0.99           | [0.03] (3%)                 | Negligible error |
| PVE5    | 2.37              | 2.38           | [0.01] (0.4%)               | Negligible error |
| WPE5    | 0.51              | 0.64           | [0.13] (20.3%)              | Negligible error |
| WPE6    | 2.02              | 2.29           | [0.27] (11.8%)              | Negligible error |
| WPE9    | 0.73              | 0.71           | [−0.02] (−2.8%)             | Negligible error |
| WPE10   | 0.68              | 0.66           | [−0.02] (−3.0%)             | Negligible error |
| WPE11   | 0.68              | 0.66           | [−0.02] (−3.0%)             | Negligible error |
| WPE12   | 0.16              | 0.16           | [0.00] (0.0%)               | Negligible error |
| PVE6    | 0.69              | 0.69           | [0.00] (0.0%)               | Negligible error |
| PVE9    | 7.54              | 7.30           | [−0.24] (−3.3%)             | Negligible error |
| RNE1    | 6.11              | 6.07           | [−0.04] (−0.7%)             | Negligible error |
| TCC1    | 6.81              | 6.39           | [−0.42] (−6.6%)             | Negligible error |
| TCC2    | 6.69              | 6.36           | [−0.33] (−5.2%)             | Negligible error |
| SBR1    | 10.36             | 10.25          | [−0.11] (−1.1%)             | Negligible error |
| SBR6    | 9.94              | 9.83           | [−0.11] (−1.1%)             | Negligible error |
| SBR7    | 5.79              | 5.67           | [−0.12] (−2.1%)             | Negligible error |
| SBR11   | 5.80              | 5.69           | [−0.11] (−1.9%)             | Negligible error |
| POX1    | 5.88              | 5.78           | [−0.10] (−1.7%)             | Negligible error |
| SBR10   | 7.34              | 7.24           | [−0.10] (−1.4%)             | Negligible error |
| SBR2    | 7.35              | 7.24           | [−0.11] (−1.5%)             | Negligible error |
| SBR3    | 5.25              | 5.14           | [−0.11] (−2.1%)             | Negligible error |
Table 1. Cont.

| Code   | Library-Based GWP | Tool-Based GWP | Error [Absolute] (Relative) | Comment                  |
|--------|-------------------|----------------|----------------------------|--------------------------|
| SBR4   | 5.04              | 4.92           | [−0.12] (−2.4%)            | Negligible error         |
| SBR5   | 11.78             | 11.67          | [−0.11] (−0.9%)            | Negligible error         |
| SBR12  | 5.82              | 5.75           | [−0.07] (−1.2%)            | Negligible error         |
| SBR13  | 7.42              | 7.30           | [−0.12] (−1.6%)            | Negligible error         |
| BMG5   | 4.16              | 4.53           | [0.37] (8.2%)              | Negligible error         |
| BMG7   | −0.13             | −0.17          | [−0.04] (23.5%)            | Negligible error         |
| SBR14  | 5.24              | 5.15           | [−0.09] (−1.7%)            | Negligible error         |
| BMG9   | −24.19            | −23.10         | [1.09] (−4.7%)             | Negligible error         |
| MAF1   | 51.70             | 51.60          | [−0.10] (−0.2%)            | Negligible error         |
| MAF3   | 1707.60           | 1707.50        | [−0.10] (0.0%)             | Negligible error         |
| HE4    | 11.54             | 9.20           | [−2.34] (−25.4%)           | Human factor: misreading of the original impact |
| HE1    | 1.02              | 1.82           | [0.80] (44.0%)             | Human factor: wrong functional unit conversion |
| PVE1   | 2.18              | 3.98           | [1.80] (45.2%)             | Human factor: wrong functional unit conversion |
| CSE2   | 1.72              | 3.30           | [1.58] (47.9%)             | Human factor: wrong functional unit conversion |
| WPE8   | 1.20              | 2.10           | [0.90] (42.9%)             | Human factor: misreading of the original impact |
| BMG6   | 8.00              | 18.52          | [10.52] (56.8%)            | Human factor: misreading of the original impact |
| PVE4   | 3.98              | 2.29           | [−1.69] (−73.8%)           | Human factor: incorrect harmonisation of compression |
| BMG3   | 1.62              | 3.18           | [1.56] (49.1%)             | Human factor: incorrect harmonisation of compression |

Table 2. Library- and tool-based harmonised non-renewable energy footprints of hydrogen (MJ per functional unit).

| Code   | Library-Based CED | Tool-Based CED | Error [Absolute] (Relative) | Comment                  |
|--------|-------------------|----------------|----------------------------|--------------------------|
| SMR2   | 200.95            | 200.39         | [−0.56] (−0.3%)            | Negligible error         |
| BMG4   | 25.36             | 24.79          | [−0.57] (−2.3%)            | Negligible error         |
| HE2    | 8.71              | 8.70           | [−0.01] (−0.1%)            | Negligible error         |
| BMG1   | 41.86             | 41.96          | [0.10] (0.2%)              | Negligible error         |
| WPE16  | 8.07              | 8.06           | [−0.01] (−0.1%)            | Negligible error         |
| WPE18  | 11.46             | 11.41          | [−0.05] (−0.4%)            | Negligible error         |
| WPE19  | 17.57             | 17.55          | [−0.02] (−0.1%)            | Negligible error         |
| BME1   | 35.50             | 36.80          | [1.30] (3.5%)              | Negligible error         |
| BMF4   | 91.12             | 89.32          | [−1.80] (−2.0%)            | Negligible error         |
| BMF5   | 183.72            | 185.58         | [1.86] (1.0%)              | Negligible error         |
| BMF6   | 87.74             | 87.73          | [−0.01] (0.0%)             | Negligible error         |
| HE1    | 23.90             | 24.00          | [0.10] (0.4%)              | Negligible error         |
| PVE1   | 59.37             | 56.50          | [−2.87] (−5.1%)            | Negligible error         |
| CSE1   | 44.28             | 40.09          | [−4.19] (−10.5%)           | Negligible error         |
| WPE8   | 29.93             | 29.87          | [−0.06] (−0.2%)            | Negligible error         |
| SBR12  | 111.22            | 111.93         | [0.71] (0.6%)              | Negligible error         |
| SBR13  | 113.98            | 112.71         | [−1.27] (−1.1%)            | Negligible error         |
| MBG7   | 3.00              | 4.90           | [1.90] (38.8%)             | Negligible error         |
| SBR14  | 114.66            | 114.56         | [−0.10] (−0.1%)            | Negligible error         |
| BMG8   | 20.40             | 20.20          | [−0.20] (−1.0%)            | Negligible error         |
| SBR10  | 98.19             | 42.11          | [−56.08] (−133.2%)         | Human factor: wrong consideration of capital goods |
Furthermore, in order to assess the correlation between the two families of harmonised values, the library-based values not affected by human factors and the corresponding tool-based ones were jointly plotted in Figures 2–4. These linear regression studies show a very high correlation between library- and tool-based impacts, accounting for $R^2$ values above 0.999 for GWP, CED, and AP. The initial hypothesis of high correlation and low deviation between library- and tool-based harmonised values was therefore accepted. Hence, since occasional deviations were found to be exclusively associated with human errors and numerical approximations (and not to an incorrect performance of the software), the harmonisation tool GreenH$_2$armony® was deemed valid. In fact, the tool-based values provide a more reliable quantification of the harmonised indicators than those available in previous libraries. In other words, the tool-based harmonised values in Tables 1–3 should be considered an update of those reported in the original libraries [12–14]. Finally, it should be noted that the use of GreenH$_2$armony® was found to drastically reduce the time needed to perform the harmonisation of a single case study. This time reduction was estimated to be of 90% of the time typically required for a “by-hand” harmonisation, being the average time needed for a harmonisation study using GreenH$_2$armony® of around 15 minutes.

### Table 3. Library- and tool-based harmonised acidification of hydrogen (kg SO$_2$ eq per functional unit).

| Code | Library-Based AP | Tool-Based AP | Error [Absolute] (Relative) | Comment |
|------|------------------|---------------|-----------------------------|---------|
| SMR2 | 1.86×10^-2      | 1.85×10^-2    | [−1.00×10^-4] (-0.5%)       | Negligible error |
| BMG4 | 1.45×10^-2      | 1.43×10^-2    | [−2.00×10^-4] (-1.4%)       | Negligible error |
| HE2  | 2.23×10^-3      | 2.15×10^-3    | [−8.00×10^-5] (-3.7%)       | Negligible error |
| BMG1 | 1.72×10^-2      | 1.72×10^-2    | [2.00×10^-5] (0.1%)         | Negligible error |
| BMG2 | 1.49×10^-2      | 1.44×10^-2    | [−5.00×10^-4] (-3.5%)       | Negligible error |
| SBR8 | 9.29×10^-2      | 9.37×10^-2    | [8.00×10^-4] (0.9%)         | Negligible error |
| SBR9 | 1.24×10^-1      | 1.20×10^-1    | [−4.14×10^-3] (-3.5%)       | Negligible error |
| WPE16| 2.40×10^-3      | 2.40×10^-3    | [0.00] (0.0%)               | Negligible error |
| WPE18| 3.70×10^-3      | 3.73×10^-3    | [3.00×10^-5] (0.8%)         | Negligible error |
| WPE9 | 4.15×10^-3      | 4.30×10^-3    | [1.50×10^-4] (3.5%)         | Negligible error |
| WPE10| 3.05×10^-3      | 3.10×10^-3    | [5.00×10^-5] (1.6%)         | Negligible error |
| WPE11| 3.05×10^-3      | 3.10×10^-3    | [5.00×10^-5] (1.6%)         | Negligible error |
| SBR1 | 5.61×10^-2      | 5.62×10^-2    | [1.00×10^-4] (0.2%)         | Negligible error |
| SBR6 | 5.38×10^-2      | 5.38×10^-2    | [0.00] (0.0%)               | Negligible error |
| SBR7 | −3.81×10^-2     | −3.81×10^-2   | [0.00] (0.0%)               | Negligible error |
| SBR11| −4.40×10^-2     | −4.40×10^-2   | [0.00] (0.0%)               | Negligible error |
| POX1 | −3.71×10^-2     | −3.71×10^-2   | [0.00] (0.0%)               | Negligible error |
| BMG5 | 1.63×10^-2      | 1.64×10^-2    | [1.00×10^-4] (0.6%)         | Negligible error |
| BMG7 | 9.62×10^-3      | 9.65×10^-3    | [3.00×10^-5] (0.3%)         | Negligible error |
| SBR14| 7.27×10^-2      | 7.27×10^-2    | [0.00] (0.0%)               | Negligible error |
| BMG9 | 2.01×10^-2      | 2.02×10^-2    | [1.00×10^-4] (0.5%)         | Negligible error |
| SBR4 | 7.02×10^-3      | 1.23×10^-2    | [5.28×10^-3] (42.9%)        | Human factor: wrong functional unit conversion |
| SBR5 | 2.56×10^-2      | 2.75×10^-3    | [−2.29×10^-2] (−830.9%)     | Human factor: wrong functional unit conversion |
Figure 2. Linear regression between library- and tool-based harmonised carbon footprints.

Figure 3. Linear regression between library- and tool-based harmonised energy footprints.
4. Conclusions

The software GreenH2armony® for the computation of life-cycle indicators of hydrogen was successfully validated. A high correlation ($R^2 > 0.999$) between library- and tool-based harmonised life-cycle indicators of hydrogen was found for the three indicators addressed (carbon, non-renewable energy, and acidification footprints). In more than 90% of the cases, the deviation between the tool-based values and the library-based ones was found to be negligible. In the remaining cases, the deviation was found to be associated with human errors in the step-by-step application of the protocols without using GreenH2armony®. This favourable validation is expected to effectively pave the way for an extended, practical use of harmonised life-cycle indicators for robust comparative LCA studies of hydrogen energy systems. Since the harmonised indicators of the tool-based libraries were found to be more reliable than those from the by-hand application of the protocols, the new values provided in this work constitute an updated version of the current libraries of harmonised carbon, energy, and acidification footprints of hydrogen. Future work in this field should focus on overcoming the limitations of the tool regarding the reduced number of sustainability indicators and dimensions addressed to date. Other limitations such as the focus on hydrogen production remain out of the scope of the proposed computational solution.

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## Appendix A

### Table A1. Case studies in the libraries of harmonised carbon [12], energy [13], and acidification [14] footprints.

| Reference | Code | Hydrogen Production Process | Harmonised Indicators |
|-----------|------|-----------------------------|-----------------------|
| [22] | SMR1 | Steam reforming of natural gas | GWP |
| [23] | SMR2 | Steam reforming of natural gas | GWP, CED, AP |
| [24] | TCC1 | NiFeO thermochemical 2-step cycle (heat from solar reactor) | GWP |
| [25] | TCC2 | ZnO thermochemical 2-step cycle (heat from solar reactor) | GWP |
| [26] | SBR1 | Bioethanol reforming (wheat grains) | GWP, AP |
| [27] | SBR2 | Bio-oil reforming (rapeseed oil) | GWP |
| [28] | SBR3 | Bio-oil reforming (palm oil) | GWP |
| [29] | SBR4 | Bioethanol (56%) + CH₄ (44%) reforming (cassava) | GWP, AP |
| [30] | SBR5 | Bioethanol reforming (from cassava) | GWP, AP |
| [31] | SBR6 | Autothermal reforming of bioethanol (wheat grains) | GWP, AP |
| [32] | SBR7 | Autothermal reforming of biomethane (cattle manure) | GWP, AP |
| [33] | SBR8 | Biomethane reforming (non-food biowaste) | GWP, AP |
| [34] | SBR9 | Biomethane reforming (German substrate mix) | GWP, AP |
| [35] | SBR10 | Biogas reforming (farm waste) | GWP, CED |
| [36] | SBR11 | Biomethane reforming (cattle manure) | GWP, AP |
| [37] | SBR12 | Bio-oil reforming (fast pyrolysis of wood chips) | GWP, CED, AP |
| [38] | SBR13 | Bio-oil reforming (fast pyrolysis of willow) | GWP, CED, AP |
| [39] | SBR14 | Bio-oil reforming (fast pyrolysis of poplar) | GWP, CED, AP |
| [40] | POX1 | Partial oxidation of biomethane (cattle manure) | GWP, AP |
| [41] | BMG1 | Biomass gasification (short-rotation poplar) | GWP, CED, AP |
| [42] | BMG2 | Biomass gasification (willow) | GWP, AP |
| [43] | BMG3 | Biomass gasification (wood chips) | GWP |
| [44] | BMG4 | Biomass gasification (poplar) | GWP, CED, AP |
| [45] | BMG5 | Biomass gasification (woody biomass) | GWP, AP |
| [46] | BMG6 | Biomass gasification (woody biomass) | GWP, AP |
| [47] | BMG7 | Biomass gasification (vine pruning waste) | GWP, AP |
| [48] | BMG8 | Biomass gasification (woody biomass) | GWP, CED |
| [49] | BMG9 | Biomass gasification with CO₂ capture (short-rotation poplar) | GWP, AP |
| [50] | WPE1 | Water electrolysis (wind power) | GWP |
| [51] | WPE2 | Water electrolysis (wind power) | GWP |
| [52] | WPE3 | Water electrolysis (wind power) | GWP |
| [53] | WPE4 | Water electrolysis (wind power) | GWP |
| [54] | WPE5 | Water electrolysis (wind power) | GWP |
| [55] | WPE6 | Water electrolysis (wind power) | GWP |
| [56] | WPE7 | Water electrolysis (wind power) | GWP |
| [57] | WPE8 | Water electrolysis (wind power) | GWP |
| [58] | WPE9 | Water electrolysis (asbestos membrane) (wind power) | GWP, AP |
| [59] | WPE10 | Water electrolysis (advanced membrane) (wind power) | GWP, AP |
| [60] | WPE11 | Water electrolysis (advanced membrane; optimised system) (wind power) | GWP, AP |
| [61] | WPE12 | Alkaline water electrolysis (Na-Cl cell) (wind power) | GWP |
| [62] | WPE13 | Alkaline water electrolysis (wind power) | GWP |
| [63] | WPE14 | PEM water electrolysis (wind power) | GWP |
| [64] | WPE15 | PEM water electrolysis (wind power) | GWP |
| [65] | WPE16 | High-temperature water electrolysis (wind power) | GWP, CED, AP |
| [66] | WPE17 | Alkaline water electrolysis (wind power) | GWP |
| [67] | WPE18 | High-temperature electrolysis (wind power) | GWP, AP |
| [68] | WPE19 | High-temperature electrolysis (wind + biogas back-up) | GWP, CED, AP |
| [69] | PVE1 | Alkaline water electrolysis (PV power) | GWP, CED |
| [70] | PVE2 | Alkaline water electrolysis (PV power) | GWP |
| [71] | PVE3 | Water electrolysis (PV power) | GWP |
| [72] | PVE4 | Water electrolysis (PV power) | GWP |
| [73] | PVE5 | Water electrolysis (PV power) | GWP |
| [74] | PVE6 | Alkaline water electrolysis (Na-Cl cell) (PV power) | GWP |
| [75] | PVE7 | PEM water electrolysis (PV power) | GWP |
| [76] | PVE8 | Alkaline water electrolysis (PV power) | GWP |
| [77] | PVE9 | Alkaline water electrolysis (PV power) | GWP |
| [78] | CSE1 | Alkaline water electrolysis (thermal solar power) | GWP |
| [79] | CSE2 | Alkaline water electrolysis (thermal solar power) | GWP, CED |
| [80] | CSE3 | Alkaline water electrolysis (hydropower) | GWP, CED |
Table A1. Cont.

| Reference | Code | Hydrogen Production Process | Harmonised Indicators |
|-----------|------|-----------------------------|-----------------------|
| [53]      | HE2  | Alkaline water electrolysis (hydropower) | GWP, CED, AP |
| [51]      | HE3  | Alkaline water electrolysis (hydropower) | GWP |
| [52]      | HE4  | Alkaline water electrolysis (hydropower) | GWP |
| [51]      | BME1 | Alkaline water electrolysis (biomass gasification electricity) | GWP |
| [54]      | RNE1 | Alkaline water electrolysis (undefined renewable power) | GWP |
| [33]      | RNE2 | Alkaline water electrolysis (undefined renewable power) | GWP |
| [55]      | BMF1 | Two-stage fermentation (wheat straw) | GWP |
| [55]      | BMF2 | Two-stage fermentation (potatoes peels) | GWP |
| [55]      | BMF3 | Two-stage fermentation (sweet stalk) | GWP |
| [56]      | BMF4 | Photo-fermentation (sugarcane) | GWP, CED |
| [56]      | BMF5 | Dark fermentation (sugarcane) | GWP, CED |
| [56]      | BMF6 | Two-stage fermentation (sugarcane) | GWP, CED |
| [57]      | MAF1 | Dark fermentation (microalgal sugar) | GWP |
| [57]      | MAF2 | Dark fermentation (microalgal sugar) | GWP |

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