The emerging field of printed electronics uses large amounts of printing and coating solvents during fabrication, which commonly are deposited and evaporated within spaces available to workers. It is in this context unfortunate that many of the currently employed solvents are non-desirable from health, safety, or environmental perspectives. Here, we address this issue through the development of a tool for the straightforward identification of functional and “green” replacement solvents. In short, the tool organizes a large set of solvents according to their Hansen solubility parameters, ink properties, and sustainability descriptors, and through systematic iteration delivers suggestions for green alternative solvents with similar dissolution capacity as the current non-sustainable solvent. We exemplify the merit of the tool in a case study on a multi-solute ink for high-performance light-emitting electrochemical cells, where a non-desired solvent was successfully replaced by two benign alternatives. The green-solvent selection tool is freely available at: www.opec-umu.se/green-solvent-tool.
Printed electronics enables for cost-efficient solution-based fabrication of functional and novel electronic and photonic devices, and, as such, it promises to develop into a multibillion industry in the near future. Large research and development efforts within academia and industry are currently dedicated to the design and development of improved printing and coating inks, because the functionality and performance of the resulting printed-electronic devices are strongly dependent on a variety of ink properties, such as solute solubility, viscosity, surface wettability, film-forming capacity, vapor pressure and shelf life.

However, an up-to-now often overlooked issue within the printed-electronics field is related to the sustainability of the employed solvents. Many of the currently employed solvents are not desirable for scale-up and commercial introduction since they present serious issues for health, safety, and/or environmental reasons. This is particularly problematic since it is anticipated that the fabrication of printed electronic devices, i.e. the deposition of the inks and the evaporation of the constituent solvents, often will be executed in open environments where workers will be exposed to the solvent vapors. Thus, it is easy to motivate why alternative "green" solvents, which deliver an ink performance on par with currently used non-sustainable solvents, should be identified.

In order to support and facilitate this transition to more sustainable solvents, we have developed a free online tool, which allows the user to identify greener functional replacement solvents for his/her particular application in a straightforward manner. More specifically, we first present the rationale for the organization and ranking of the solvent functionality with the aid of the Hansen solubility parameters and a number of key ink properties, then introduce the sustainability descriptors and ranking, and finally exemplify the utilization of the tool in a case study where two functional green replacement solvents for the solution-based fabrication of high-performance light-emitting electrochemical cells are identified.

Results

Green solvent definition. So, what is the definition of a green solvent? This is not a trivial question to answer in a succinct manner. More specifically, we first present the rationale for the organization and ranking of the solvent functionality with the aid of the Hansen solubility parameters and a number of key ink properties, then introduce the sustainability descriptors and ranking, and finally exemplify the utilization of the tool in a case study where two functional green replacement solvents for the solution-based fabrication of high-performance light-emitting electrochemical cells are identified.

Table 1 The solvent sustainability categories and subcategories in the GSK solvent sustainability guide and the method for the calculation of the composite score.

| Category         | Subcategory                  | Category Score | Composite Score |
|------------------|------------------------------|----------------|-----------------|
| Health           | Health Hazard                | \( H = \sqrt{HH \times EP} \) | \( G = \sqrt{H \times S \times E \times W} \) |
|                  | Exposure Potential           |                 |                 |
| Safety           | Flammability & Explosion     | \( S = \sqrt{F & E \times R & S} \) |                 |
|                  | Reactivity & Stability       |                 |                 |
| Environment      | Air Impact                   | \( E = \sqrt{Air \times Aqua} \) |                 |
| Waste Disposal   | Incineration                 | \( W = \sqrt{I \times R \times BT \times VOC} \) |                 |
|                  | Recycling                    |                 |                 |
|                  | Bio Treatment                |                 |                 |
|                  | Volatile Organic Compounds   |                 |                 |

The factor four in front of the dispersion term signals the relative importance of the dispersion parameter, and its origin is discussed in detail in references.
Figure 1 presents the location of 132 solvents in the 3D Hansen solubility space, with some of the more well-known solvents being identified by their chemical name. At this point, we recommend the reader to consult with the web tool at www.opeg-umu.se/green-solvent-tool, where the same graph is presented with higher detail and in addition can be rotated for clearer visualization. Moreover by clicking on a desired solvent in the Hansen space graph or in the solvent ranking table in the web tool, information is disclosed regarding its chemical structure, HSPs, CAS number, melting and boiling points, viscosity, surface tension, specific health, safety and environmental issues, and its scores in the different GSK sustainability categories (as identified in Table 1).

The composite sustainability score, $G$, is practical in that it provides a summary evaluation of the sustainability of a solvent. In order to directly visualize this overall sustainability, the sphere that represents a solvent in Fig. 1 (and in the web tool) is both size and traffic-light-color coded, according to the specification in the inset. In short, a large green circle represents a highly sustainable solvent (G ≥ 7), an intermediate-sized yellow/orange circle corresponds to a solvent with a limited number of sustainability issues (G = 5–6), while a small red circle is concomitant with a solvent that should be avoided (G ≤ 4). As an example, we note that ethylene glycol (G = 8.1) and n-butyl acetate (G = 7.5) are preferable green solvents from most aspects, whereas benzene (G = 3.7) and 1–4 dioxane (G = 4.1) should be avoided.

The closer two solvents are positioned in the 3D Hansen solubility space in Fig. 1, i.e., the smaller the $R_a$, the more similar is their solubility capacity (and cohesive energy). In other words, if we know that a certain solvent can dissolve a specific solute, the probability that a second solvent also can dissolve the same solute is increasing with decreasing $R_a$. The web tool provides a facile route to rank all of the solvents as regards to their $R_a$ value with respect to a known functional solvent. The procedure is a follows: (i) mark the “known functional solvent(s) of your solute” option in the upper left corner, (ii) select the functional solvent in the selection bar below, and (iii) click the update button. The outcome is that all solvents will be ranked from low to high $R_a$ with respect to the selected functional solvent. For instance, if we select toluene as the functional solvent for a specific solute, we are quickly informed that cumene ($R_a$ = 0.8) and benzene ($R_a$ = 1.6) are likely to be functional solvents for the same solute, but not acetonitrile ($R_a$ = 17.9) or ethylene glycol ($R_a$ = 25.9).

A refinement of this solvent ranking procedure is available if two or more solvents are known to dissolve the desired solute(s). All of the identified functional solvents are in this scenario included into the selection bar in step (ii); after the update button is pushed, the program calculates the mean HSP values for these known functional solvents and ranks the remaining solvents as regards to their $R_a$ distance from this mean of the HSPs of the functional solvents. A further improvement of the solvent ranking is possible when the HSPs of the solute are known. Then the solvent-ranking procedure follows the path: (i) mark the “known HSP of your solute”, (ii) include the values for $\delta_D$, $\delta_P$, and $\delta_H$, and (iii) click the update button.

The functionality of an ink solvent is further determined by its ability to enable a desired ink-substrate wetting, ink-film formation, and solute-film drying during the printing/coating procedure. Key solvent properties that determine if a specified solvent is suitable for a certain printing/coating process and substrate include its boiling point (which is related to the vapor pressure), viscosity and surface tension, and the web tool presents the tabulated values for each of these properties for all the solvents. It also allows the user to define the functional range of each of these properties for a specific application by clicking on the “Refinement options” tab and by dragging the corresponding sliders. By pressing the update button, the tool excludes solvents with properties outside the selected ranges, and also updates the solvent ranking table and the Hansen space plot accordingly. We note that for some solute combinations and applications, such as organic solar cells, it is common to utilize multi-solvent inks in order to attain the desired ink and dry-film properties. In this context, we mention that the tool is capable of identifying green replacements for each of the different solvents in a multi-solvent ink by simply repeating the above procedure for each solvent.

Case study with green ink formulation for high-performance LEC device. With this tool at hand, a viable and straightforward method to identify potential replacement solvents that are both functional (low $R_a$ value and appropriate physical properties) and sustainable (high $G$ value) is in place. We exemplify how such a solvent-replacement procedure can be executed in a case study on the identification of a sustainable solvent for the fabrication of a high-performance light-emitting electrochemical cell (LEC). A bright and record-efficient LEC, featuring a solution-processed single-layer active material sandwiched between two air-stable electrodes, was recently reported, but a drawback was that the active-material ink comprised chlorobenzene as the solvent. This is a concern since chlorobenzene is harmful to the skin (GHS hazard statements H312 and H315), the eyes (H319) and when inhaled (H332), as well as toxic to aquatic life (H411) and flammable (H226); see Table 2. Consequently, chlorobenzene scores low in both the health category ($H = 4.0$) and in the environmental category ($E = 3.7$), and the composite sustainability score is a modest $G = 5.4$. A further challenge with this LEC ink is that it comprises four different solutes (two host compounds, one guest emitter, and one electrolyte) that should be dissolved in its solvent in a high total solute concentration of ~30 g L⁻¹ (see the “Methods” section for details) in order to allow for the formation of a pinhole-free thin film following the ink deposition and drying. Since chlorobenzene is an established functional solvent for this particular multi-component solute, we start by selecting chlorobenzene as the “known functional solvent(s)” in the selection bar in the web tool and by clicking the update button.
This updates the solvent ranking table in the web tool and sorts all 132 solvents from lowest-to-highest $R_a$. The result in the solvent ranking table can be refined to only include sustainable solvents by first clicking on the "Refinement options" tab, thereafter dragging the slider under "Set lower limit for $G$" until it shows $G > 7$, and by finally clicking the update button. Table 2 reveals that the seven closest sustainable solvents (with $G \geq 7$), ranked from lowest to highest $R_a$, are: ethoxybenzene, anisole, cyclohexanone, methyl oleate, 2-ethylhexyl acetate, pentyl acetate, and n-butyl acetate.

We start our experimental investigation with the closest neighbor in the Hansen solubility space, ethoxybenzene ($R_a = 2.3$), and establish that it is capable of dissolving the multicomponent solute in the desired concentration. Since ethoxybenzene features a significantly higher composite sustainability score ($G = 7.2$) than chlorobenzene ($G = 5.4$), it is clearly a more preferred solvent from a sustainability perspective. In fact, Table 2 discloses that ethoxybenzene scores higher than chlorobenzene in all four category scores. The second-nearest neighbor, anisole ($R_a = 5.5$), also passes the solubility test, and since it scores slightly higher in the composite sustainability score ($G = 7.4$) it could be an even better option. It should however be noted that ethoxybenzene scored a modest 4.9 in the health category because of inadequate information, and that it is thus likely that a complete evaluation would have resulted in a higher composite sustainability score. We mention that a third solvent, cyclohexanone, also passed the solubility test, but that it was not considered further since it did not present an improvement in the composite sustainability and since it is labeled with a larger number of hazard statements; see Table 2. The three acetates—2-ethylhexyl acetate, pentyl acetate, and n-butyl acetate—as well as methyl oleate are interesting from a sustainability viewpoint, but could not dissolve the multicomponent solute in the desired high concentration. This indicates that the $R_a$ boundary for dissolution for this particular multi-solute system has been crossed at $R_a > 6$, although it should be emphasized that this boundary is not expected to be distinct.

With ethoxybenzene and anisole identified as potential green replacement solvents to chlorobenzene for the LEC ink, we turn to device fabrication and characterization in order to investigate the practical functionality of the different inks in LEC devices. It was first established that the chlorobenzene, ethoxybenzene and anisole based inks all could be used for repeatable spin-coating fabrication of uniform and pinhole-free active-material thin films (with a thickness of 120 nm), as evidenced by the low surface roughness in AFM and the spatial uniformity of the UV-activated photoluminescence (see Fig. S1). Note that the differences in viscosity and boiling point for the ink solvents (see Table 2) were compensated for by the selections for solute concentration and spin speed, as detailed in the Methods section. We fabricated six indium-tin-oxide/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)/active-material/Al devices with each fresh ink (prepared the same day), and Fig. 2a present the luminance and voltage transients as a function of ink-solvent selection for a typical device in each category when driven by a constant current density of $j = 77 \text{ A m}^{-2}$.

The transients are highly independent on the ink-solvent selection, and all devices feature the LEC-characteristic increasing luminance and decreasing voltage during the turn-on phase when a p–n junction doping structure forms in the active material. All three devices also emit vibrant green light ($\lambda_{\text{peak}} = 525 \text{ nm}$) with essentially identical electroluminescence spectrum (see Figure S2), which implies that the guest emitter is molecularly dispersed in the host matrix and not forming emission-shifting aggregates. Table S1 presents a summary of key performance.
metrics of the best-performing device in each category. It reveals that the turn-on time to a high luminance of >1000 cd m\(^{-2}\) is very fast (<2 s) and effectively identical for all devices, that the peak luminance and efficiency is a bit higher for the LECs fabricated from the chlorobenzene ink (3100 vs. ~2540 cd m\(^{-2}\) and 39.9 vs. ~32.9 cd A\(^{-1}\)), and that the operational lifetime is essentially the same for all devices.

We have also investigated the storage stability of the three inks based on chlorobenzene, ethoxybenzene, and anisole as the solvent, and Fig. 2b present the luminance and voltage transients for devices fabricated from such active-material inks stored for 30 days. Table S1 shows that the peak performance has dropped by 20% for the LEC fabricated from the chlorobenzene ink, while the device fabricated from the ethoxybenzene and anisole inks is more robust to long-term ink storage. This difference in long-term stability is attributed to that one component in the multi-solute ink (the n-type host OXD-7) is observed to fall out and crystallize in the chlorobenzene solvent, and that this tendency is inhibited with ethoxybenzene as the solvent, and absent with anisole as the solvent. This suggests that the HSPs of OXD-7 are closer to anisole than chlorobenzene.

We finally evaluated the merits of the three active-material inks for the fabrication of flexible LEC devices by bar coating, which is a much more scalable fabrication method than spin coating. We sequentially bar coated flexible ITO-coated poly(ethylene terephthalate) (PET) substrates with the poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, Clevios P VP AI 4083, Heraeus) ink was spin coated on top of the ITO at 4000 rpm for 60 s, and the required reflow of the distributed ink on the substrate before drying for the formation of a uniform film, it is critical to appropriately tune the ink viscosity for the required reflow, the ink surface tension for an appropriate wetting, and the ink boiling point for the desired drying time. Table 2 shows that the surface tension and the boiling point are fairly similar for the three investigated solvents, whereas the larger differences in solvent viscosity are effectively compensated for by the high viscosity introduced by the solute (primarily the high molecular weight host polymer PVK); see “Methods” section for details. Figure S3 shows that uniform thin films can be bar coated with all three active-material inks, and that these bar-coated films can be employed as the active material in LEC devices that deliver bright green and uniform light emission. Figure 2c and Table S2 provide quantitative information on the performance of these bar-coated LECs, and it is notable that the performance is very good and close to that of the spin-coated LEC. The slightly lower luminance and higher drive voltage of the bar-coated LECs in comparison to the spin-coated LECs can be attributed to that the thickness of the active material of the former is slightly larger than the optimum value of 120 nm. Most importantly, our results demonstrate that both anisole and ethoxybenzene can successfully replace the non-sustainable chlorobenzene for the solvent in a multi-solute LEC ink for both spin-coating and bar-coating fabrication, and that the new sustainable LEC ink can be utilized for an environmentally friendly and safe fabrication of bright, efficient and low-cost LEC devices.

**Discussion**

In summary, we introduce a tool for the rational and facile identification of functional and sustainable solvents for the field of printed electronics. Specifically, the tool orders a large number of solvents in accordance to their Hansen solubility and relevant physical parameters, as well as to descriptors and ranking values regarding health, environmental impact, safety, and overall sustainability. We demonstrate the functionality of the tool in a case study on a high-performance light-emitting electrochemical cell, where the currently employed non-sustainable ink solvent was successfully replaced by two alternatives that are more benign. It is our hope that this open-access green-solvent selection tool (freely available at www.opeg-umu.se/green-solvent-tool) will contribute to that dangerous, toxic, and non-sustainable solvents will be efficiently replaced in both research laboratories as well as in larger industrial settings.

**Methods**

**Ink preparation.** Master inks were prepared by separately dissolving the solutes poly(9-vinylcarbazole) (PVK, M\(_n\) = 1.1 x 10\(^6\) g mol\(^{-1}\), Sigma-Aldrich), 1,3-bis(2-(4-tert-butylphenyl)-1,3,4-oxadiazio-5-yl)benzene (OXD-7, Lumtec), tris(2-(3-substituent-phenyl)-pyridinato)iridium(III) (IrR-ppy), Merck) and tetraxylammonium tetrafluoroborate (THABF\(_4\), Sigma-Aldrich) in either chlorobenzene (anhydrous, Sigma-Aldrich), ethoxybenzene (Sigma-Aldrich) or anisole (anhydrous, Sigma-Aldrich) under stirring on a hot plate kept at 70 °C for 5 h. The master inks were blended together in a mass ratio of PVK:OXD-7:Ir(R-ppy):THABF\(_4\) = 32.3:32.3:29.0:6.4 and a total solute concentration as described below for each solvent. The resulting active-material ink was stirred on the hot plate at 70 °C for at least 1 h before further processing. The ink preparation was performed in a N\(_2\)-filled glovebox ([O\(_2\)], [H\(_2\)O] < 1 ppm).

**Device fabrication and characterization.** For the spin-coated LECs, indium-tin-oxide (ITO) coated glass substrates (20 Ω sq\(^{-1}\), Thin Film Devices) were carefully cleaned by subsequent sonication in detergent (Extran MA01, Merck), deionized water, acetone and isopropanol followed by drying at 120°C. Prior to film deposition the substrates were exposed to UV-ozone (model 42-220, Jelight) for 10 min. The poly(3,4-ethylenedioxythiophene)poly(styrene sulfonate) (PEDOT:PSS, Clevios P VP AI 4083, Heraeus) ink was spin coated on top of the ITO at 4000 rpm for 60 s, and dried on a hot plate at 120 °C for 30 min. The thickness of the dry PEDOT:PSS film

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**Fig. 2 LEC performance as a function of ink solvent, ink storage and ink deposition method.** The temporal evolution of the luminance and the voltage for LECs fabricated from inks based on chlorobenzene (solid red circles), ethoxybenzene (open blue squares) and anisole (open green diamonds). a The performance of LECs fabricated by spin coating “fresh” inks (prepared the same day). b The performance of LECs fabricated by spin coating “old” inks, which had been stored for 30 days following ink preparation. c The performance of LEC fabricated by scalable bar coating. All devices were driven by a constant current density of \(j = 77 \text{ A m}^{-2}\).
was 40 nm. The active-material ink was spin coated onto the PEDOT:PSS, and thereafter dried on a hotplate at 70 °C for 2 h. For the attainment of a 120 nm thick active-material film, the following deposition parameters were employed: chlorobenzene: solute concentration: 30 g l\(^{-1}\) - spin speed: 2000 rpm; ethoxybenzene: 46.5 g l\(^{-1}\) - 190 nm for the anisole ink. On top of the active material, a 100 nm thick Al electrode was deposited by thermal evaporation (p < 5 × 10\(^{-6}\) mbar, Leybold). The 1.5 × 8.5 mm\(^2\) emission area was defined by the overlap between the ITO anode and the Al cathode. The film thickness was measured with a stylus profilometer (DektakXT, Bruker). The surface uniformity of the spin-coated films was measured with atomic force microscopy (AFM, MultiMode SPM microscope, equipped with a Nanoscope IV Controller, Veeco Metrology) in tapping mode under ambient conditions.

For the bar-coated LECs, the PEDOT:PSS coated film was 50–60 nm thick. The active-material ink (solvent concentration = 35 g l\(^{-1}\)) was bar-coated onto the PEDOT:PSS layer, using a Mayer rod with 0.08 mm wire size and a coating speed of 20 mm s\(^{-1}\) at room temperature. The coated substrates were directly and gently transferred onto a hot plate at 120 °C and dried for 4 min. The dry thickness of the bar-coated PEDOT:PSS film was 50–60 nm. The active-material ink (solvent concentration = 35 g l\(^{-1}\)) was bar-coated onto the PEDOT:PSS layer, using a Mayer rod with 0.08 mm wire size and a coating speed of 20 mm s\(^{-1}\) at room temperature. The coated substrates were directly and gently transferred onto a hot plate at 120 °C and dried for 4 min. The dry thickness of the active-material film was 160–220 nm for the chlorobenzene ink, 160–170 nm for the ethoxybenzene ink, and 160–190 nm for the anisole ink. On top of the active material, a 100 nm thick Al electrode was deposited by thermal evaporation. The 2 × 2 mm\(^2\) emission area was defined by the overlap between the ITO anode and the Al cathode.

The non-encapsulated LEC devices were characterized in an N\(_2\)-filled glovebox ([O\(_2\)], [H\(_2\)O] < 1 ppm). The devices were driven by a constant current density of 10 mA cm\(^{-2}\) (U2722A, Agilent). For the bar-coated LECs, the luminance and voltage were measured with a calibrated photodiode (S9219-01, Hamamatsu Photonics) and the voltage was measured with a source measure unit (U2722A, Agilent). For the bar-coated LECs, the luminance and voltage were measured with an OLED lifetime tester (M6000 PMX, McScience). The devices were driven by a constant current density of 10 mA cm\(^{-2}\) and operated for 10 min to render its surface hydrophilic. A PEDOT:PSS ink (Clevios P VP AI 4083, Heraeus) was bar coated (AB3008, TCQ) on the PET/TTO substrate using a Mayer rod with 0.08 mm wire size and a coating speed of 20 mm s\(^{-1}\). The PEDOT:PSS coated substrates were directly and gently transferred onto a hot plate at 120 °C and dried for 4 min. The dry thickness of the bar-coated PEDOT:PSS film was 50–60 nm. The active-material ink (solvent concentration = 35 g l\(^{-1}\)) was bar-coated onto the PEDOT:PSS layer, using a Mayer rod with 0.08 mm wire size and a coating speed of 20 mm s\(^{-1}\) at room temperature. The coated substrates were directly and gently transferred onto a hot plate at 120 °C and dried for 4 min. The dry thickness of the active-material film was 160–220 nm for the chlorobenzene ink, 160–170 nm for the ethoxybenzene ink, and 160–190 nm for the anisole ink. On top of the active material, a 100 nm thick Al electrode was deposited by thermal evaporation. The 2 × 2 mm\(^2\) emission area was defined by the overlap between the ITO anode and the Al cathode. The film thickness was measured with a stylus profilometer (DektakXT, Bruker). The surface uniformity of the spin-coated films was measured with atomic force microscopy (AFM, MultiMode SPM microscope, equipped with a Nanoscope IV Controller, Veeco Metrology) in tapping mode under ambient conditions.

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Author contributions
C.L., P.L., and L.E. conceptualized the idea. S.T., P.L., C.L., E.M.L., J.W., and A.S. performed the experimental work and the data analysis. J.R.R. and C.L. designed and coded the green solvent tool. L.E., C.L., and P.L. wrote the paper. All authors reviewed the paper.

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Correspondence and requests for materials should be addressed to L.E.

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