

VOCs absorption from gas streams using deep eutectic solvents – a review

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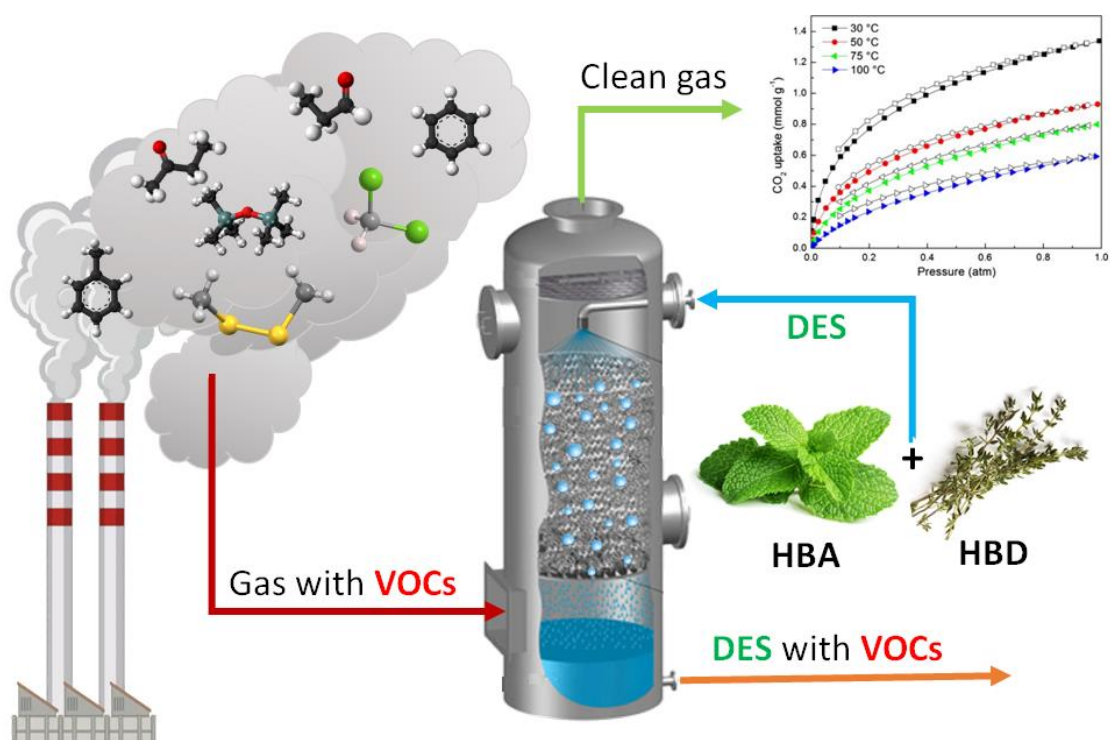
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Abstract: Volatile organic compounds (VOCs) are one of the most severe atmospheric pollutants. They are mainly emitted into the atmosphere from anthropogenic sources such as automobile exhaust, incomplete fuel combustion, and various industrial processes. VOCs not only cause hazards to human health or the environment but also adversely affect industrial installation components due to their specific properties, i.e., corrosive and reactivity. Therefore, much attention is being paid to developing new methods for capturing VOCs from gaseous streams, i.e., air, process streams, waste streams, or gaseous fuels. Among the available technologies, absorption based on deep eutectic solvents (DES) is widely studied as a green alternative to other commercial processes. This literature review presents a critical summary of the achievements in capturing individual VOCs using DES. The types of used DES and their physicochemical properties affecting absorption efficiency, available methods for evaluating the effectiveness of new technologies, and the possibility of regeneration of DES are described. In addition, critical comments on the new gas purification methods and future perspectives are included.

Keywords: deep eutectic solvents, volatile organic compounds, absorption, gas purification, air

33 **Graphical abstract:**



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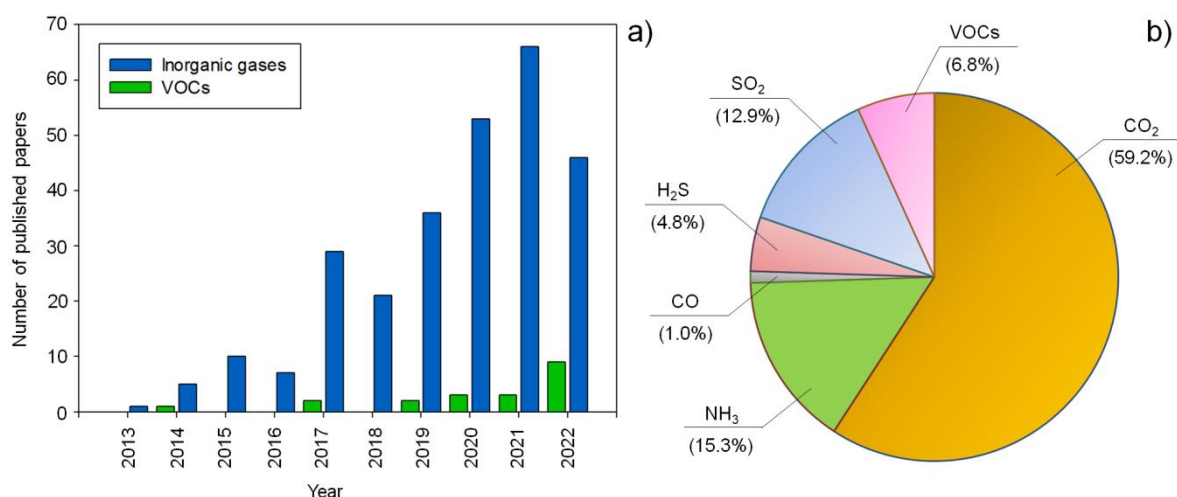
35 **1. Introduction**

36 Volatile organic compounds (VOCs) are considered one of the most important chemical
 37 compounds that cause severe air pollution. According to the definition introduced by the World
 38 Health Organization (WHO), VOCs are organic compounds whose boiling points (BP) range from 50 to
 39 260 °C. In addition, they are characterized by high vapor pressure at room temperature. VOCs can
 40 react with other substances in the atmosphere and affect the formation of photochemical fog, acid
 41 rain, ozone depletion, etc. In addition, some VOCs are harmful to the environment, human health,
 42 animals, and industrial installations. Particularly unfavorable VOCs include compounds from the
 43 groups of volatile oxygen-, sulfur-, halogen-, and silicon-organic compounds, as well as
 44 monoaromatic and aliphatic hydrocarbons. They can be emitted into the atmosphere from both
 45 natural and anthropogenic sources. However, the most severe risk is caused by VOCs that are
 46 released from automobile exhaust, incomplete combustion of domestic fuel, and industrial sources
 47 such as process vessels, venting, leaks from piping and other process equipment, wastewater
 48 streams, heat exchange systems, breathing and loading losses from storage tanks, and transport of
 49 raw materials and final products [1–7]. Therefore, it is essential to effectively capture VOCs from the
 50 air, fuels, and industrial gas streams.

51 There are various methods to treat gaseous streams, e.g., catalytic and thermal oxidation,
 52 adsorption, physical and chemical absorption, adsorption, pressure swing adsorption, membrane
 53 separation, and cryogenic and biological separation [8–15]. Among available methods, absorption is
 54 widely used due to its simplicity, high efficiency, low operation, investment cost, and ability to
 55 regenerate the absorbent. However, to achieve high absorption efficiency while meeting the
 56 principles of green chemistry and green engineering, it is essential to select the appropriate
 57 absorbent [16–20]. Commercially available and widely used absorbents include water, silicon,
 58 minerals, vegetable and petroleum oils, polyethylene glycols, amines and other traditional organic

59 solvents with high BP [21–24]. Most of these absorbents are characterized by low sorption capacity
 60 of VOCs (mainly hydrophobic ones), solvent degradation, and corrosion of installation equipment. In
 61 addition, some absorbents require a distillation process for regeneration. This process involves high
 62 energy requirements, and secondary contaminants can form during solvent regeneration [25,26].
 63 Therefore, new alternative absorbents are still being sought.

64 Until recently, ionic liquids (ILs) were considered ideal absorbents due to their non-volatile
 65 nature, non-flammability, non-explosivity, and the ability to tune other physicochemical properties
 66 based on the selection of appropriate cation and anion [27]. However, expensive and complicated
 67 synthesis, as well as proven toxicity and negligible biodegradability, have prevented them from being
 68 implemented in industrial processes [28,29]. To overcome the above limitations, in 2003 Abbott et al.
 69 introduced a new type of green solvent called deep eutectic solvents (DES) [30]. According to the
 70 definition, a DES is a mixture of two or more chemical compounds that bind to each other by specific
 71 non-covalent interactions. In the DES structure, one of the components plays the role of a hydrogen
 72 bond donor (HBD) and the second one an acceptor (HBA). Due to the HBA-HBD interaction (charge
 73 delocalization), a significant decrease in the melting point of DES compared to pure components is
 74 observed [31]. In addition, DES are characterized by similar unique physicochemical properties to ILs,
 75 but they are "generally" less toxic, more biodegradable, and more cost-effective. The DES's ability to
 76 interact with impurities molecules via hydrogen bonds or weaker electrostatic interactions, as well as
 77 the ability to regenerate multiple times, can transform well-known technological processes into more
 78 ecological ones. The favorable properties of DES have resulted in an increasing number of scientific
 79 articles on their industrial application. DESs have been tested as extraction solvents for fuel
 80 desulfurization and denitrification [32–35], separation of aromatic and aliphatic hydrocarbons [36],
 81 biocatalysis [37], recycling of Li-ion batteries [38], and extraction of bioactive compounds [39]. To
 82 date, the application of DES for the capture of inorganic harmful gaseous molecules, that is, carbon
 83 dioxide, water, ammonia, hydrogen sulfide, or sulfur dioxide from the air, gaseous fuels, biogas, and
 84 industrial gases, has been relatively well described in the literature [40–49]. However, there is still
 85 little information on the capture of VOCs. Figure 1 shows the number of published papers on the
 86 absorption of inorganic pollutants from gaseous streams compared to VOCs between 2013 and 2022.



87
 88 **Figure 1** a) Number of published papers between 2013 and 2022. The graphs were built using the Scopus database, from
 89 the topics deep eutectic solvents, absorption or capture, and volatile organic compounds; b) Percentage breakdown of the
 90 number of publications from 2013 to 2022 describing the use of DES to remove individual impurities from the gas phase.
 91 (Access date: 21.09.2022).

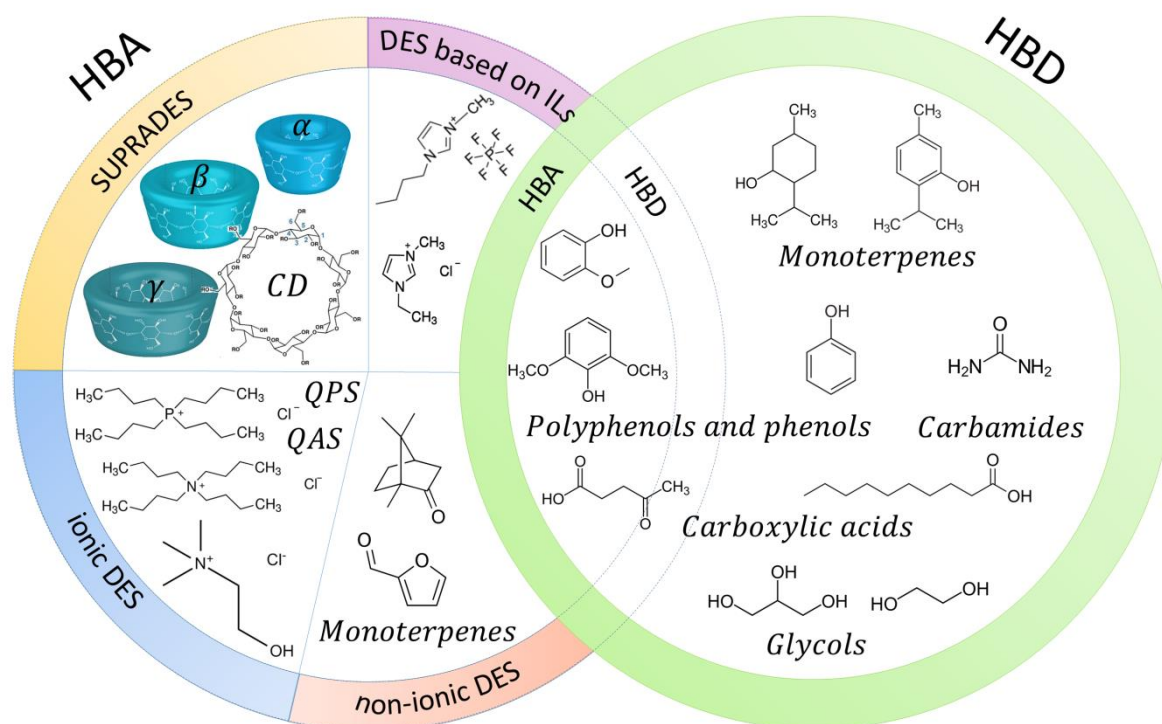
92 To the best of the author's knowledge, this is the first review article to critically summarize
93 and compare the achievements in the application of DES for the absorption of various groups of
94 VOCs from gaseous streams. In addition, the paper describes the influence of the physicochemical
95 and structural properties of DES on the efficiency of VOCs captured from gas streams and methods of
96 DES regeneration. Critical comments on the new gas treatment procedures and future perspectives
97 are also included.

98 **2. Characterization of deep eutectic solvents used for VOCs capture**

99 **2.1. Structures of DES**

100 DES are a new generation of solvents that have become an alternative to non-aqueous and
101 organic solvents because of their unique properties. DES are also known as tailor-made solvents
102 because of the ability to tune their parameters for specific applications by selecting appropriate HBAs
103 and HBDs. Currently, there are several classifications of DES [50–54]. However, we can distinguish
104 four types of DES tested as new absorbents. They can be classified according to the HBA component
105 in the structure of the eutectic mixture. The first group includes the most popular and well-known
106 ionic DES, which consists of quaternary ammonium salts (QAS) or phosphonium salts (QPS). The
107 second group includes non-ionic DES, in which organic compounds containing active oxygen atoms
108 ($=O$, $-O-$) in their structure, including monoterpenes, polyphenols, or carboxylic acids, are used for
109 preparation [55]. The third group is the recently discovered SUPRADES, which contain cyclodextrins
110 (CDs) in their structures [56,57]. The last group includes DES composed of ionic liquid and an organic
111 compound [58]. In all types of DES, the same substances, i.e., monoterpenes, phenols, polyphenols,
112 carbamides, sugars, carboxylic acids, and glycols, are present as the second component. Some HBDs
113 can also be used as HBAs due to several active groups or specific groups, i.e., a carboxyl group, which
114 can combine through hydrogen bonds with another acid and form a DES. Examples of DES

115 component structures are shown in Figure 2.



116

117 **Figure 2** Main structures of HBA and HBD in deep eutectic solvents used for VOCs absorption.

118 2.2. DES properties

119 The main parameter determining the effective absorption of VOCs is their solubility in DES.
120 However, in dynamic processes, other parameters of absorbents also define their usefulness in
121 purifying gas streams [59]. These physicochemical properties include melting/freezing point,
122 degradation temperature, rheological parameters (viscosity, density, and surface tension), and
123 polarity. To meet the requirements of green chemistry and green engineering, it is also necessary to
124 determine the toxicity, vapor pressure, and biodegradability of DES [60,61]. All physicochemical
125 parameters of DES used so far in the absorption of VOCs are summarized in Table 1.

126 2.2.1. Liquid range of DES

127 In the absorption process, the use of liquid sorption media is crucial. The liquid range of DES
128 depends on the lower (melting (MP), freezing (FP), and glass transition temperature (T_g)) and the
129 upper-temperature limit (degradation temperature (T_{deg})) [62]. Physical absorption is a highly
130 exothermic process. To achieve high VOCs absorption efficiency, it is necessary to use low
131 temperatures in the process. MP/FP is the parameter to classify eutectic complexes as solvents. The
132 main difference between conventional solvents, solvent mixtures, and DES is the sharp drop in the
133 MP/FP of the complex compared to the pure components [63]. The decrease in melting/freezing
134 point is related to the strength of the non-covalent interactions between HBA and HBD. The stronger
135 the interactions between DES components, the lower the MP/FP. The types and strength of
136 interactions depend on the chemicals and stoichiometry used to prepare DES. The melting point of

137 ionic DES used in the absorption of VOCs ranges from -68.9 to 12 °C, while non-ionic DES range from -
138 48 to 21.9 °C. For SUPRADES and IL-based DES, only glass transition temperature in the range from -
139 51 to -87 °C can be found. T_g is the temperature at which a hard glassy state of an amorphous
140 material is converted to a rubbery state, and it is much lower than MP or FP.

141 The degradation temperature affects the ability of DES to regenerate. Typically, regeneration
142 processes are conducted at elevated temperatures (60-125 °C) to ensure high desorption efficiency,
143 and to reduce process time. At high temperatures, the phase and significant changes in the deep
144 eutectic solvent structures and the breakdown of non-covalent interaction between HBA and HBD
145 may occur [64]. Therefore, DES used in the absorption/desorption process should exhibit high T_{deg} .
146 The degradation temperatures of ionic DES are relatively high. T_{deg} values range from 107.2 to 396 °C.
147 High degradation temperatures can also be observed for SUPRADES (117–137 °C).

148 This indicates that they could potentially be suitable absorbents for capturing VOCs and their
149 subsequent desorption. However, information on the degradation temperatures of the other two
150 DES groups is missed. However, due to the tendency of monoterpenes to sublime, the degradation
151 temperature of most non-ionic DES can be expected to be much lower [65]. Therefore, these
152 substances are not a suitable choice as absorption media. On the other hand, ILs-based DES should
153 probably have similar or higher degradation temperatures to ionic DES due to the high T_{deg} values of
154 pure ionic liquids [65,66]. Nevertheless, still more research is needed.

155

156 2.2.2. Rheological parameters

157 Viscosity

158 Dynamic viscosity (η) is a crucial rheological parameter that affects absorption efficiency. Most
159 DES are characterized by relatively high viscosity (>100 mPas) at room temperature (RT) and under
160 atmospheric pressure [53,67,68]. However, the high viscosity of the absorbents is unfavorable
161 because it hinders the mass transfer process. During absorption process the high dynamic viscosity of
162 DES can lead to slow gas diffusion through the gas-liquid interface. Large gas bubbles are formed in
163 the barbotage column instead of tiny gas bubbles, which decreases the gas-liquid contact area.
164 Consequently, at high DES viscosities, local hydrodynamics are modified by generating a more
165 stabilized DES liquid. In addition, applying low viscous solvents in gas purification can reduce energy
166 costs associated with pumping, heating, or desorption. The dynamic viscosity values of deep eutectic
167 solvents depend mainly on the substrates used to prepare DES and their stoichiometry.

168 Ionic DES composed of QAS or QPS and various HBDs have the highest viscosity. Their viscosities
169 range from 15 to 3451.35 mPas [69,70]. The high values of solvent dynamic viscosity are attributable
170 to the large ion size in the HBA structure and relatively small empty volumes (steric effect) [56]. In
171 addition, the length of the alkyl chain in HBA and HBD significantly affects the viscosity of DES. As the
172 chain length decreases, solvent viscosity decreases. Slightly lower viscosities have SUPRADES, whose
173 viscosities at 30 °C range from 212.9 to 1300 mPas [71]. The addition of cyclodextrins to the eutectic
174 mixtures increases the viscosity of DES. However, the increase is relatively small due to the low
175 amount of CD in DES structures [57]. IL-based DES also exhibits relatively high viscosities (150-470
176 mPas at 25 °C) [72]. Their high viscosity is mainly due to the very high η of ionic liquids [73]. The
177 lowest viscosities are characterized by non-ionic DES (4.2–31.1 mPas at 20 °C) [61,74].

178 DES viscosity is also affected by the strength and type of interactions between the HBA and
179 HBD. Stronger non-covalent interactions, i.e., hydrogen bonds, electrostatic bonds, van der Waals, or

180 π - π , reduce the mobility of free species within the eutectic molecule and cause an increase in
181 viscosity [75]. One possibility to lower the viscosity of DES is to add water. However, many DES are
182 highly hydrophobic. Therefore, the addition of water is only possible for selected absorbents. The
183 water addition can significantly modify other physicochemical properties, causing a reduction in the
184 solubility of VOCs in DES. The viscosity of DES can also be reduced by raising the temperature of the
185 absorption process, which is consistent with the Arrhenius or Vogel-Fulcher-Tammann model. This is
186 because an increase in temperature causes an increase in the average velocity of HBA and HBD in the
187 liquid phase, resulting in a decrease in intermolecular forces. This results in a reduction in fluid flow
188 resistance and a change in dynamic viscosity [20]. However, increasing the temperature of the
189 absorption process is unfavorable due to the exothermic nature of the absorption process [76].
190 Therefore, it is more advantageous to select DES, which has a lower viscosity at low temperatures.

191

192 **Density**

193 Another important rheological parameter of solvents is density (ρ). However, in the
194 absorption process, the importance of this parameter is minor. DES density values can determine the
195 selection of the DES regeneration process. Deep eutectic solvents, whose densities differ significantly
196 from conventional solvents, i.e., water, can be regenerated by extraction.

197 Most ionic DES have a higher density than water, within the range of 1000 to 1350 kg/m³
198 [51,68]. Similar to the viscosity parameter, the exact value of DES density strongly depends on the
199 type of ingredients used in DES preparation. IL-based DES have the highest density. Their density
200 values range from 1040 to 1689.3 kg/m³. This is due to the high density of ILs. SUPRADES have
201 viscosities in the range of 1184.5 to 1234.3 kg/m³. Non-ionic DES have the lowest viscosity (898 –
202 943.7 kg/m³). Only those non-ionic DES composed of levulinic acid and terpenes have a slightly
203 higher viscosity than water. The density of DES can be controlled by mixing water or another organic
204 solvent with DES, or by decreasing/ increasing the temperature [77]. The strong temperature
205 dependence on DES density is because of a change in the kinetic energy of the molecules. At a high
206 temperature, the increase in the movement of the deep eutectic solvent molecules occurs. This
207 evokes an increase in the molar volume of DES [78,79].

208 **Surface tension**

209 The surface tension (ST) of DES also affects the mass transfer process [80]. According to the
210 definition, surface tension is the solvent's tendency to obtain the minimum surface area. The value of
211 surface tension mainly depends on the type and hydrophilic or hydrophobic nature of HBA and HBD,
212 the strength of the intermolecular interactions between DES components, and temperature [81].
213 Typically, the surface tension of DES is in the range of 40–65 mN·m⁻¹ at room temperature [82,83],
214 and they are higher than surface tension of most ionic liquids [84]. However, only ionic DES are
215 characterized by high surface tension (45.8 – 75.3 mN/m). Values of surface tension in ILs and ionic
216 DES correspond to the strength of H-bonding between molecules [85]. Non-ionic DES have much
217 lower surface tension. The surface tension values are comparable to conventional organic solvents
218 (25-36.5 mN/m). However, there is no data on surface tension of SUPRADES and ILs-based DES. Low
219 surface tension values favorably affect the efficiency of VOCs absorption. To reduce the surface
220 tension of absorbents, HBA or HBD with a longer alkyl chain should be used to synthesize DES. This
221 contributes to lowering the charge density and decreasing the importance of electrostatic
222 interactions between DES components [61]. Likewise, the QAS or QPS content of the DES structure

223 can be increased, which destroys the hydrogen bond network [81]. In addition, increasing the
224 temperature of DES reduces surface tension. This is due to an increase in the movement of molecules
225 and the average kinetic energy, as well as a decrease in cohesion forces between DES components
226 [81,86,87].

227 2.2.3. Polarity

228 The polarity of the absorbents is an important parameter characterizing the ability of DES to
229 dissolve VOCs. This parameter corresponds to the intermolecular interactions between DES and
230 solutes that do not lead to chemical reactions [88]. The exact value of the DES polarity can be
231 determined using Betaine Dye 30 on a scale of $E_T(30)$, Betaine Dye 33 on a scale of $E_T(33)$, normalized
232 polarity scale (E_TN), and Nile red polarity scale ($E_T(NR)$). [89–91]. All these parameters allow for
233 determining the electronic transition energy in DES-VOC complexes. DES exact polarity values may
234 differ depending on the method used. Because of the numerous available ways, it isn't easy to
235 compare the polarity of the new DES with literature data. Published data indicate that most DES are
236 relatively polar. The polarity values of DES are similar to popular organic solvents i.e., methanol
237 ($E_T(33)=63.9$ kcal/mol; $E_T(30)=55.5$ kcal/mol; $E_TN=0.76$), ethanol ($E_T(33)=60.5$ kcal/mol; $E_T(30)=52.1$
238 kcal/mol; $E_TN=0.66$), acetonitrile ($E_T(33)=54.8$ kcal/mol; $E_T(30)=46.6$ kcal/mol; $E_TN=0.49$), and water
239 ($E_T(33)=71.5$ kcal/mol; $E_T(30)=63.1$ kcal/mol; $E_TN=1.0$), and dimethylsulfoxide ($E_T(33)=53.9$ kcal/mol;
240 $E_T(30)=45.5$ kcal/mol; $E_TN=0.46$) [92,93]. The $E_T(33)$, $E_T(30)$, $E_T(NR)$ and E_TN values of the ionic DES
241 and DES-based ILs are slightly lower than water and higher than the popular organic solvents. The
242 ranges of $E_T(33)$, $E_T(30)$, $E_T(NR)$, and E_TN values for DES are 65.7-67.59, 47.75-57.3, 45.5, and 44.88-
243 58.07 kcal/mol, and 0.81-0.87, respectively. This means that ionic DES are medium-polar. To reduce
244 the polarity of DES, HBDs containing longer hydrocarbon chains should be used for their synthesis
245 [93]. There is no data about the polarity of SUPRADES and non-ionic DES. Some published papers also
246 describe the Kamlet-Taft parameters of deep eutectic solvents [94]. The π^* , α , and β values for ionic
247 DES are in ranges of 0.85 – 1.14, 0.421 – 1.85, and 0.27 – 1.05, respectively. Data are missing for
248 most non-ionic DES, SUPRADES, and DES based on ILs. Kamlet-Taft parameters were calculated only
249 for Th:DA (1:1) ($\alpha=1.11$; $\beta=0.05$; $\pi^*=0.71$). DES have similar dipolarity values to water ($\pi^*=1.09$),
250 while they are higher than the rest of conventional solvents (0-58). In addition, the π^* value
251 increases with decreasing hydrocarbon chain length in HBDs structures. The occurrence of
252 permanent dipoles, delocalized bonds, and DES structures can explain this phenomenon [90,95].
253 Most DES show a higher donation capacity of protons in DES-to-solute hydrogen bonding than pure
254 alcohols and ILs. On the other hand, DES show only slightly lower α values compared to water and
255 pure acids [95–98]. Therefore, these compounds may be more suitable for the absorption of
256 chlorinated or oxygenated organic compounds due to their potential ability to form hydrogen bonds
257 such as $-OH\cdots Cl$, $-COOH\cdots Cl$, $-NH_2\cdots Cl$, $-OH\cdots O=$, $-COOH\cdots O=$, $-NH_2\cdots O=$. On the other hand, most DES
258 are characterized by β values lower than 0.7. This means that the DES commonly used for VOC
259 capture are weak hydrogen bond acceptors and show less alkaline properties [95]. Only TBPCI: Lev
260 (1:2), TBABr:DA (1:2), and ChCl:Res (1:2) show a higher potential to be a hydrogen bond acceptor
261 than conventional solvents. Therefore, they can absorb more efficiently VOCs such as alcohols and
262 amines. The polarity can be modified by increasing or decreasing the DES temperature. The
263 decreasing of the temperature generally increases the DES polarity. This is due to an increase in the
264 absorbent's hydrogen-bond donating acidity and the formation of stronger interaction between HBA
265 and HBD [99]. However, most absorption processes are carried out at room temperature or lower.

266 Therefore, the selection of DES for removing specific VOC groups should be driven by the structures
267 of HBA and HBD.

268 **2.2.4. DES volatility**

269 From an industrial and environmental point of view, the volatility of DES is a crucial
270 parameter. The application of absorbents characterized by high vapor pressure can cause the
271 evaporation of DES components during the absorption and desorption processes. This can lead to the
272 loss of DES and the need for frequent replacement, which significantly increases the cost of the gas
273 purification process. It can also result in the contamination of treated gas and atmospheric air. [100].
274 Therefore, only non-volatile sorption media should be used in the absorption process. Fortunately,
275 deep eutectic solvents are considered non-volatile. However, compared to the numerous reports on
276 the volatility of ILs, only limited information on the volatilization of DES can be found in the literature
277 [101–104]. The vapor pressure (V_p) of DES depends strongly on the type of HBA and HBD. Ionic DES
278 are mostly non-volatile. The total vapor pressures of the most popular DES, i.e., ChCl:U (1:2) and
279 ChCl:Gly (1:2), are 2.94 and 46.16 Pa at 393K, respectively [105,106]. The vapors pressure of ChCl:EG
280 (1:2) is about two orders of magnitude higher than that of ChCl:Gly (1:2). This is because the vapor
281 pressure of pure EG is much higher (6148.6 Pa at 393 K) compared to Gly (97.9 Pa at 393 K). This
282 indicates that the type of HBD and its stoichiometry plays a crucial role in the volatility of DES
283 because the vapor pressure of most quaternary ammonium and phosphonium salts is negligibly low.
284 Non-ionic DES, which consist of monoterpenes (i.e., menthol, thymol, lidocaine), has a higher vapor
285 pressure. Vapor pressure values at 40 °C typically range from 0.44 to 15.84 Pa. This is due to the
286 ability of monoterpenes to sublime [65]. Replacing one of the monoterpenes with a carboxylic acid
287 significantly reduces the vapor pressure of DES [107]. Currently, there is no information on the
288 volatility of SUPRADES and DES based on ILs. However, a comparison of available literature values of
289 the volatility of DES with conventional organic solvents and ILs, indicates that DES have much lower
290 vapor pressures than organic solvents and higher than ionic liquids [108]. In addition, the vapor
291 pressure of all absorbent groups increases with increasing temperature [109]. At elevated
292 temperatures, in the first stage, the interactions between HBA and HBD are weakened, and
293 evaporation of the individual components occurs in the next step [106,110]. However, due to the
294 limited volatility data of DES, before the selection of the most suitable absorbent, it is necessary to
295 examine the vapor pressure of DES in both absorption and desorption process temperatures.

296 **2.2.5. Toxicity**

297 Apart from the physicochemical properties of DES that have a decisive impact on the
298 absorption efficiency of VOCs, it is essential to determine their ecological footprint. Most published
299 articles describe DES as non-toxic and environmentally friendly solvents. However, there are still few
300 papers that confirm this assumption. The literature provides information on the toxicity of the
301 individual components of DES (HBA and HBD) [111,112]. For instance, Morrison et al. examined the
302 toxicity profiles of ChCl and U for rats and mice (oral LD 50), which were 3400 and 3900 mg/kg for
303 ChCl and 8471 and 11000 mg/kg for U. The results suggest that these substances are
304 pharmaceutically acceptable [113]. Since DES are composed of at least two components, a synergy
305 effect between HBA and HBD may occur. This effect may increase or decrease the toxicity of eutectic
306 complexes compared to pure DES components [114]. However, only a few DES have been studied in
307 detail. Hayyan et al. demonstrated, based on toxicity tests on two Gram-positive bacteria (*Bacillus*

308 *subtilis* and *Staphylococcus aureus*) and two Gram-negative bacteria (*Escherichia coli* and
309 *Pseudomonas aeruginosa*), as well as brine shrimp (*Artemia salina*), that four DES (ChCl:Gly, ChCl:EG,
310 ChCl:TEG i ChCl:U) do not have toxic effects. On the other hand, they found that the cytotoxicity of all
311 tested DES was higher than their individual components [114]. The same conclusions were also
312 suggested in other works [115,116]. In contrast, in another study, the authors obtained contradictory
313 findings. The authors observed that DES showed an antagonistic effect against *Allivibrio fischeri*. This
314 indicates that DES are less toxic than separate substances [117]. Based on in vitro toxicity data on cell
315 lines (fish COO and human MCF cancer cell lines), the authors indicated that ChCl:Gly (1:2) has low
316 cytotoxicity (EC 50 > 2000 mg/L), and the ChCl:Ox (1:2) has moderate cytotoxicity (EC 50=218.7 and
317 559 mg/L, respectively for COO and MCF cells). On the other hand, phytotoxicity results indicate that
318 both DES are non-toxic at seed germination (EC50 > 5000 mg/L) [118]. Similar results were obtained
319 for HEK293T, MCF-7, and HeLa 7 cell lines [70]. This suggests that DES containing organic acids in
320 their structure have toxic properties and cannot be considered green absorbents. Other studies have
321 shown that DES toxicity increases with alkyl chain length in QAS and QPS [119]. In addition, it has also
322 been shown that the molar ratio of HBA components: HBD can positively or negatively affect the
323 toxicity of DES [120]. Current studies suggest that the toxicity of DES depends on several variables.
324 Some of the DES are not toxic, so they can successfully be used as green solvents. However, only a
325 few DES have been studied. For most DES, information on their toxicities is lacking. However, further
326 cytotoxicity studies are crucial to enable their large-scale utilization.

327 **2.2.6. Biodegradability**

328 New absorbents should be biodegradable to meet the requirements of green chemistry and
329 engineering. This is a significant parameter because it allows an understanding of the environmental
330 impact of solvents. According to the most common biodegradability test (No. 301) of the OECD
331 Guidelines, solvents are considered readily biodegradable if a minimum of 70% of dissolved organic
332 carbon is removed [121]. In addition, 60% of CO₂ should be generated by the respirometry method
333 within ten days in a 28-d period of the aerobic test. Most of the DES-based absorbents used to date
334 are readily biodegradable. After 28 days of experiments, the biodegradability of DES composed of
335 QAS is above 67.2% [70,118,122,123]. This means that DES are more biodegradable than ILs [124].
336 Only TBABr:DA (1:2) does not exhibit biodegradability after a 14-day process, which suggests that
337 DES does not meet the requirements for green solvents. This is because the process of degradation
338 of hydrophobic DES is very slow. Therefore, a significant amount of water should be added to dilute
339 the absorbents and accelerate their biodegradation. This significantly increases the cost of DES
340 degradation. Moreover, substances with long alkyl chains in structures are less susceptible to
341 transmembrane transport, resulting in relatively low biodegradation. In theory, DES that contains the
342 hydroxyl, carboxyl, and amino groups may be readily biodegradable due to potential sites for enzyme
343 reactions [125]. However, there is still very little data on the biodegradability of DES. There is a lack
344 of biodegradability results for SUPRADES, non-ionic DES, and DES based on ILs. In addition, the direct
345 mechanisms of DES biodegradation are also poorly understood. The chemical mechanism may be
346 related to the ability of HBA and HBD to cross cell walls or to degrade on their own [126,127].
347 However, further studies are needed.

348 **2.2.7. Price of DES**

350 From an economical and industrial point of view, the price of DES is crucial. Since DES are not
351 commercially available, their cost can only be estimated by the cost of individual HBAs and HBDs on a
352 laboratory scale. Therefore, their prices are relatively high. Prices per kilogram of ionic, non-ionic,
353 DES based on ILs and SUPRADES range from 29.9–1165.7 €, 18.0–533.0 €, 55.9–246.9 €; 2521.4–
354 15482.0 €, respectively (Table 1). The costs of the selected DES were estimated from the original
355 articles. These values, in unchanged form, are included in the manuscript. For the remaining DES,
356 prices were determined by averaging the market prices (per kilogram) of popular chemical sellers.
357 The cost of DES was subsequently calculated based on the stoichiometry of the individual
358 components. The presented prices were estimated for high-purity (above 98%) DES components. The
359 results indicate that the average cost of non-ionic DES is lower than other DES. This is due to the
360 relatively low price of monoterpenes, carboxylic acids, and polyphenols, which are usually used to
361 prepare DES. The high costs of the other groups of DES are influenced by the relatively high prices of
362 QASs, QPSs, cyclodextrins, and ionic liquids [128]. Nevertheless, the price is lower for large-scale
363 sales. It is generally considered that the cost of absorbents on an industrial scale is 10% of the price
364 of absorbents sold on a laboratory scale [129]. In addition, it is possible to reduce costs by decreasing
365 the purity of HBA and HBD. However, additional impurities can affect the efficiency of removing
366 VOCs from the gas and influence the physicochemical properties of DES. Therefore, further research
367 is needed before using this approach. The unit price per kilogram of DES is lower than ILs but higher
368 than conventional organic solvents [130]. However, due to the low volatility of DES, they can
369 potentially be regenerated many times without significant loss (this is a common occurrence with the
370 use of organic solvents). This means that despite the higher purchase price of DES compared to
371 conventional absorbents, the total cost of the VOCs absorption process can be comparable to or
372 lower than the price for other gas treatment methods [131].

373 Table 1 Physicochemical properties of DES used for VOCs absorption

DES (HBA:HBD molar ratio)	Abbreviation	Viscosity at 20°C (mPa·s)	Density at 20°C (g/cm ³)	Surface tension at 20 °C [mN/m]	MP/FP /TG [°C]	Decomposition temperature [°C]	Vp [Pa]	Polarity	Toxicity EC ₅₀	Biodegradability [%]	Hydrophobicity	Price per 1 kg	Literature
Ionic DES													
Choline chloride : urea (1:2)	ChCl:U (1:2)	1618.3	1.17	63.76	12	134.5	0.336 ^{e)} ; 2.941 ^{f)} ; 6.6 ^{h)}	E _T (33)=65.4 kcal/mol; E _T (30)=57.0 kcal/mol; E _T (RN)=49.25 kcal/mol E _T N=0.81 α=1.42; β=0.5; π*=1.14	4500 mg/L (<i>Allivibro fischeri</i>) 7.098 % v/v (<i>Allivibro fischeri</i>) >2000 mg/L (<i>Hela cells</i>) >2000 mg/L (<i>HEK293T cells</i>) >2000 mg/L (<i>MCF-7 cells</i>) 8532 mg/L (<i>R. subcapitata</i>) 1099 mg/L (<i>D. magna</i>) 26346 mg/L (<i>A. fischeri</i>) 295.9 mM <i>E. coli</i>	97.1 % (28 days) 64.6 (28 days)	No	44.6 €	[70,88, 122,12 3,126,1 32– 136]
Choline chloride : resorcinol (1:2)	ChCl:Res (1:2)	326 2218	1.1869	61.56	-43.9	n.d.	n.d.	α _{pred} =1.85; β _{pred} =1.0	n.d.	n.d.	No	71 €	[69,13 5,137,1 38]
Choline chloride : levulinic acid (1:2)	ChCl:Lev (1:2)	206 ^{c)}	1.1371 ^{b)} 1.1345 ^{c)}	n.d.	-11.9 ⁱ⁾	159	n.d.	E _T (NR)=49.79 kcal/mol; E _T (33)=50.26 kcal/mol; E _T N=0.35 α=0.51; β=0.57; π*=1.0	n.d.	67.2 (28 days)	No	42.5 €	[72,13 2,135,1 39,140]
Choline chloride : lactic acid (1:2)	ChCl:Lac (1:2)	305 ^{c)}	1.176 ^{c)}	47.4	-63.9	196.83	n.d.	E _T (30)=48.02 kcal/mol; E _T (33)=67.59; kcal/mol; E _T (NR)=44.88 kcal/mol; E _T N=0.87; α=1.34; β=0.5; π*=0.85	33.6 (<i>A. fischeri</i>)	n.d.	No	35.5 €	[81,14 1]
Choline chloride : Oxalic acid (1:2)	ChCl:OxA (1:2)	5363 ^{c)}	n.d.	75.30 ^{b)}	-21 ⁱ⁾	134.81	n.d.	E _T (30)=47.78 kcal/mol; E _T (33)=66.19 kcal/mol;	330.9 mg/L (<i>Hela cells</i>) > 2000 mg/L (<i>HEK293T cells</i>) 558.98 mg/L (<i>MCF-7</i>)	73.4 (28 days)	No	50.3 €	[70,13 5,139]



								$E_T N=0.828$; $\alpha_{pred}=1.19$; $\beta_{pred}=0.27$	cells)				
Choline Chloride : phenol (1:2)	ChCl:Ph (1:2)	122.51 ^{b)}	1.0989 ^{b)}	35.46 ^{b)}	-68.9	n.d.	n.d.	n.d.	1.54 % v/v (<i>Allivibro fischeri</i>)	79 (14 days)	No	50.6 €	[134,142]
Choline chloride : ethylene glycol (1:2)	ChCl:EG (1:2)	207.14	1.1168	49.4; 48.91	-66	107.2	790 ^{e)} ; 5600 ^{g)}	$E_T(NR)=57.3$ kcal/mol $E_T(33)=65.7$ kcal/mol; $E_T(30)=57.3$ kcal/mol; $E_T N=0.82$ $\alpha=1.47$; $\beta=0.57$; $\pi^*=1.07$	4000 mg/L (<i>Allivibro fischeri</i>) 9196 mg/L (<i>R. subcapitata</i>) 1868 mg/L (<i>D. magna</i>) 108526 mg/L (<i>A. fischeri</i>) 532.0 E. Coli	81.9 (28 days) 73.4 (28 days)	No	37.5 €	[64,81,82,88,122,123,135,136]
Choline chloride : diethylene glycol (1:2)	ChCl:DEG (1:2)	52.5	1.1143 ^{b)} 1.1216	48.49	-13	n.d.	n.d.	ENR=50.8 kcal/mol	n.d.	n.d.	No	56.9 €	[132,143,144]
Choline chloride : glycerol (1:2)	ChCl:Gly (1:2)	237; 136.1 ^{c)}	1.1920	57.24; 57.8	-36	184	2.141 ^{e)} ; 46.16 ^{g)}	$E_T(NR)=58.1$ kcal/mol; $E_T(33)=66.4$ kcal/mol; $E_T(30)=58.0$ kcal/mol; $E_T N=0.84$ $\alpha=1.49$; $\beta=0.52$; $\pi^*=1.11$	9000 mg/L (<i>Allivibro fischeri</i>) 7080 mg/L (<i>R. subcapitata</i>) 2528 mg/L (<i>D. magna</i>) 86726 mg/L (<i>A. fischeri</i>) 434.4 mM (E.Coli)	95.9 (28 days)	No	40.1 €	[64,81,82,88,122,123,135,136]
Choline chloride : 1,2-propanediol : water (1:1:1)	ChCl:PC:H ₂ O (1:1:1)	114.5	1.08	n.d.	n.d.	134.0	n.d.	$E_T(NR)=50.2$ kcal/mol; $\alpha=0.421$; $\beta=0.531$; $\pi^*=1.05$	1.541 % v/v (<i>Allivibro fischeri</i>)	n.d.	No	38.9 €	[70]
Choline chloride : glucose : water (5:2:5)	ChCl:Glu:H ₂ O (5:2:5)	3451.3	1.41	n.d.	n.d.	154.4	n.d.	$E_T(NR)=53.3$ kcal/mol	8.448 % v/v (<i>Allivibro fischeri</i>) 182 mM (<i>HeLa S3</i>) 139 mM (<i>CoOV3</i>), 186 mM (<i>MCF-7</i>) 211 mM (<i>B16F10</i>)	92 (28 days) 83 (14 days)	No	38.2 €	[70,134]
Choline chloride : xylose : water (2:1:3)	ChCl:Xyl:H ₂ O (2:1:3)	400.3	1.17	n.d.	n.d.	207.9	n.d.	$E_T(NR)=50.4$ kcal/mol	n.d.	89.7 (28 days) 80 (14 days)	No	29.9 €	[70,134]
Tetraethylammonium chloride : phenol (1:2)	TEACl:Ph (1:2)	15	1.04	n.d.	1.9	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	406.8 €	[69]
Tetraabutylammonium bromide: decanoic acid (2:1)	TBABr:DA (2:1)	1784 ^{d)}	0.9158 ^{d)}	n.d.	-60	185	n.d.	n.d.	n.d.	n.d.	Yes	706 €	[145]
Tetraabutylammonium bromide: decanoic acid (1:1)	TBABr:DA (1:1)	1100 ^{d)}	0.9108 ^{d)}	n.d.	-63	197	n.d.	n.d.	n.d.	n.d.	Yes	534 €	[145]



Tetrabutylammonium bromide: decanoic acid (1:1.5)	TBABr:DA (1:1.5)	600 ^{d)}	0.9071 ^{d)}	n.d.	-72	205	n.d.	n.d.	n.d.	n.d.	Yes	432 €	[145]
Tetrabutylammonium bromide: decanoic acid (1:2)	TBABr:DA (1:2)	548	0.9045 ^{d)}	n.d.	-9	204	n.d.	$E_T(NR)=51.4$ kcal/mol $\alpha=0.95$; $\beta=1.05$; $\pi^*=0.71$	0.123 %, v/v (<i>Allivibro fischeri</i>)	0% (14 days)	Yes	361.9 €	[95,13 5,145]
Tetrabutylammonium bromide: decanoic acid (1:3)	TBABr:DA (1:3)	102.4 ^{d)}	0.8964 ^{d)}	n.d.	-4	197	n.d.	n.d.	n.d.	n.d.	Yes	281 €	[145]
Tetrabutylphosphonium chloride : Levulinic acid (1:1)	TBPCL:Lev (1:1)	480	0.99	n.d.	n.d.	367	n.d.	n.d.	n.d.	n.d.	No	1165.7 €	[146]
Tetrabutylphosphonium chloride : Levulinic acid (1:2)	TBPCL:Lev (1:2)	229.3	1.0228	n.d.	n.d.	369	n.d.	$\beta=0.82$; $\pi^*=1.06$	n.d.	n.d.	No	777.9 €	[146]
Tetrabutylphosphonium chloride : Levulinic acid (1:3)	TBPCL:Lev (1:3)	175	1.04	n.d.	n.d.	369	n.d.	n.d.	n.d.	n.d.	No	595 €	[146]
Tetrabutylphosphonium chloride : Levulinic acid (1:4)	TBPCL:Lev (1:4)	138	1.06	n.d.	n.d.	369	n.d.	n.d.	n.d.	n.d.	No	481.5 €	[146]
Tetrabutylphosphonium bromide : Levulinic acid (1:6)	TBPBr:Lev (1:6)	118.2	1.09	n.d.	n.d.	118.95	n.d.	$E_T(NR)=49.2$ kcal/mol	0.12 %, v/v (<i>Allivibro fischeri</i>)	n.d.	No	76.1 €	[147,1 48]
Tetrabutylphosphonium bromide : decanoic acid (1:2)	TBABr:DA (1:2)	85	0.95	n.d.	n.d.	126.7	n.d.	$E_T(NR)=51.4$ kcal/mol	n.d.	n.d.	Yes	183 €	[137]
Tetrabutylphosphonium bromide: glycerol (1:1)	TBPB:Gly (1:1)	n.d.	n.d.	45.77	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	No	199.9 €	[147,1 48]
Tetrapropylammonium bromide: Tetraethylene glycol (1:2)	TPABr:TEG (1:3)	84.6 ^{c)}	1.552	n.d.	-48	n.d.	n.d.	n.d.	n.d.	n.d.	No	66.1 €	[149]
Trimethylallyl ammonium chloride : phenol (1:2)	TAACl:Ph (1:2)	45	1.06	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	No	414 €	[25]
Triethyl benzyl ammonium chloride : levulinic acid (1:2)	TEBACl:Lev (1:2)	1850	1.11	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	No	77.1 €	[25]
Triethyl benzyl ammonium chloride : Ethylene glycol	TEBACl:EG (1:2)	420	1.10	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	No	91.7 €	[25]
Triethyl benzyl ammonium chloride : phenol (1:2)	TEBACl:Ph (1:2)	2150	1.088	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	83.4 €	[25]
Triethyl benzyl ammonium chloride : phenol (1:3)	TEBACl:Ph (1:3)	325	1.085	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	No	64.5 €	[25]
Tetraethylammonium chloride : phenol (1:2)	TEACl:Ph (1:2)	170	1.05	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	No	406.8 €	[25]
Tetraethylammonium chloride :oleic acid (1:3)	TEACl:OA (1:3)	203	0.91	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	304.7 €	[150]
Non-ionic DES													
Guaiacol : Camphor: Levulinic acid (1:1:3)	Gu:C:Lev (1:1:3)	n.d.	1.1123 ^{b)}	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	33 €	[132]
Camphor: Guaiacol (1:1)	C:Gu (1:1)	10.9	1.0284	n.d.	-48	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	32.78 €	[74]
Syringol : Levulinic acid (1:1)	Syr:Lev (1:1)	31.1	1.1675	n.d.	-23	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	263.5 €	[74]
Guaiacol : Levulinic acid (1:1)	Gu:Lev (1:1)	30	1.1382	n.d.	-43	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	36.01 €	[74]



Camphor : Levulinic acid (1:2)	C:Lev (1:2)	26.2	1.0597	n.d.	8	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	24.02 €	[74]
Eucalyptol : octanoic acid (1:1)	E:OA (1:1)	5.25	0.9157	30	-18.9	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	43 €	[61,15 1]
Camphor : octanoic acid (1:1)	C:OA (1:1)	9.42	0.9397	30.9	-1.4	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	29.9 €	[61,15 1]
Camphor : decanoic acid (1:1)	C:DA (1:1)	11.2	0.9437	28.1	17.0	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	35.5 €	[61,15 1]
Camphor : undecanoic acid (1:1)	C:UDA (1:1)	13.4	0.8985	n.d.	18.1	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	95.1 €	[61,15 1]
Camphor : dodecanoic acid (3:1)	C:DDA (3:1)	4.2 ^{b)}	0.925	n.d.	-18.8	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	44 €	[61,15 2]
Carvone : levulinic acid (1:1)	Car:Lev (1:1)	7	1.04	36.5	-25	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	62.4 €	[61]
Carvone : octanoic acid (1:1)	Car:OA (1:1)	4.4	0.95	31	-22	n.d.	n.d.	E _r (NR)=52.5 kcal/mol	n.d.	n.d.	Yes	58.4 €	[61] 131]
Carvone : nonanoic acid	Car:NA (1:1)	4.6	0.94	29.8	-21	n.d.	n.d.	E _r (NR)=52.5 kcal/mol	n.d.	n.d.	Yes	61.1 €	[61] 131]
Carvone : decanoic acid (1:1)	Car:DA (1:1)	5	0.93	29	-19	n.d.	n.d.	E _r (NR)=52.7 kcal/mol	n.d.	n.d.	Yes	63.8 €	[151] [61] 131]
Thymol : decanoic acid (1:1)	Th:DA (1:1)	11.8	0.9281	30.02	11.5	n.d.	n.d.	$\alpha=1.11$; $\beta=0.05$; $\pi^*=0.71$	n.d.	n.d.	Yes	41 €	[61,13 5,151,1 53,154]
Lidocaine : decanoic acid (1:2)	Lid:DA (1:2)	55 ^{a)}	0.96	n.d.	n.d.	192	55 ^{f)}	n.d.	n.d.	n.d.	Yes	533 €	[137]
Dodecanoic acid : decanoic acid (1:2)	DDA:DA (1:2)	4.7 ^{c)} ; 12.9	0.898	n.d.	21.9; 18	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	27 €	[153,1 55,156]
Dodecanoic acid : decanoic acid (1:3)	DDA:DA (1:3)	10.8	0.9	n.d.	21.2	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	33.1 €	[137]
Octanoic acid : dodecanoic acid (3:1)	OA:DDA (3:1)	6 ^{c)} ; 8.2	0.91	25	9	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	25.1 €	[155,1 56]
Octanoic acid : decanoic acid (2:1)	OA:DA (2:1)	4.9	0.904	n.d.	< RT	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	18 €	[137]
Nonanoic acid : dodecanoic acid (3:1)	NA:DDA (3:1)	7.5 ^{c)} ; 10.1	0.901	n.d.	9	n.d.	n.d.	n.d.	n.d.	n.d.	Yes	29.1 €	[155,1 56]
SUPRADES													
1ifobutylether-β-CD : Levulinic acid (1:44)	Captisol:Lev (1:44)	1300 ^{c)}	1.2343 ^{c)}	n.d.	-67.8 ⁱ⁾	127.6	n.d.	n.d.	n.d.	n.d.	n.d.	55.93 €	[71]
2w methylated-β-CD : Levulinic acid (1:25)	CRYSMEB:Lev (1:25)	480 ^{c)}	1.2075 ^{c)}	n.d.	-73.5 ⁱ⁾	137.7	n.d.	n.d.	n.d.	n.d.	n.d.	246.94 €	[71]
3ydroxypropyl-β-CD : Levulinic acid (1:32)	HP- β -CD:Lev (1:32)	600 ^{c)}	1.2047 ^{c)}	n.d.	-73.3 ⁱ⁾	130.4	n.d.	n.d.	n.d.	n.d.	n.d.	130.4 €	[71]
4andomly methylated-β-CD : Levulinic acid (1:27)	RAMEB:Lev (1:27)	212.9 ^{c)}	1.1845 ^{c)}	n.d.	-74.3 ⁱ⁾	117.6	n.d.	n.d.	n.d.	n.d.	n.d.	71.4 €	[71]
DES based on ILS													



Hexylcholinium levulinate	Chol-C6-Lev	300 ^{b)}	1.08 ^{b)}	n.d.	-79.8 ⁱ⁾	n.d.	n.d.	E _r (NR)=52.9 kcal/mol	n.d.	n.d.	n.d.	n.d.	[72]
Octylcholinium levulinate	Chol-C8-Lev	470 ^{b)}	1.06 ^{b)}	n.d.	-82.6 ⁱ⁾	n.d.	n.d.	E _r (NR)=53.9 kcal/mol	n.d.	n.d.	n.d.	n.d.	
Hexylcholinium lactate	Chol-C6-Lac	150 ^{b)}	1.10 ^{b)}	n.d.	-73.1 ⁱ⁾	n.d.	n.d.	E _r (NR)=52.5 kcal/mol	n.d.	n.d.	n.d.	n.d.	
Octylcholinium lactate	Chol-C8-Lac	460 ^{b)}	1.04 ^{b)}	n.d.	-68.7 ⁱ⁾	n.d.	n.d.	E _r (NR)=53.0 kcal/mol	n.d.	n.d.	n.d.	n.d.	
Tetrabutylammonium perfluorobutanesulfonate : perfluoropentanoic acid (1:1)	[N ₄₄₄₄][C ₄ F ₉ SO ₃]:C ₄ F ₉ CO ₂ H (1:1)	n.d.	1.3517	n.d.	-61 ⁱ⁾	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	15482 €	[157]
Tetrabutylammonium perfluorobutanesulfonate : perfluoropentanoic acid (2:1)	[N ₄₄₄₄][C ₄ F ₉ SO ₃]:C ₄ F ₉ CO ₂ H (2:1)	n.d.	1.3066	n.d.	-51 ⁱ⁾	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	19874 €	
Tetrabutylammonium perfluorobutanesulfonate : perfluoropentanoic acid (1:2)	[N ₄₄₄₄][C ₄ F ₉ SO ₃]:C ₄ F ₉ CO ₂ H (1:2)	n.d.	1.4191	n.d.	-82 ⁱ⁾	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	11089 €	
(2-Hydroxyethyl)trimethylammonium perfluorobutanesulfonate : perfluoropentanoic acid (1:1)	[N ₁₁₁₂ (OH)][C ₄ F ₉ SO ₃]:C ₄ F ₉ CO ₂ H (1:1)	n.d.	1.6336	n.d.	-79 ⁱ⁾ (MP=0° C)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3826 €	
(2-Hydroxyethyl)-trimethylammonium perfluoropentanoate : perfluoropentanoic acid (1:1)	[N ₁₁₁₂ (OH)][C ₄ F ₉ CO ₂]:C ₄ F ₉ SO ₃ H (1:1)	n.d.	1.6291	n.d.	-75 ⁱ⁾	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3671 €	
(2-Hydroxyethyl)-trimethylammonium perfluorooctanesulfonate : perfluoropentanoic acid (1:1)	[N ₁₁₁₂ (OH)][C ₈ F ₁₇ SO ₃]:C ₄ F ₉ CO ₂ H (1:1)	n.d.	1.6841	n.d.	-87 ⁱ⁾	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3496 €	
1-Ethyl-3-methylimidazolium perfluorooctanesulfonate : perfluoropentanoic acid (1:1)	[C ₂ C ₁ Im][C ₈ F ₁₇ SO ₃]:C ₄ F ₉ CO ₂ H (1:1)	n.d.	1.6721	n.d.	-84 ⁱ⁾ (MP=-4°C)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2790 €	
1-Ethyl-3-methylimidazolium perfluorooctanesulfonate : perfluoropentanoic acid (2:1)	[C ₂ C ₁ Im][C ₈ F ₁₇ SO ₃]:C ₄ F ₉ CO ₂ H (2:1)	n.d.	1.6875	n.d.	-84 ⁱ⁾	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3058.6 €	
1-Ethyl-3-methylimidazolium perfluorooctanesulfonate : perfluoropentanoic acid (1:2)	[C ₂ C ₁ Im][C ₈ F ₁₇ SO ₃]:C ₄ F ₉ CO ₂ H (1:2)	n.d.	1.6893	n.d.	-80 ⁱ⁾	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2521.4 €	

374 a) measurements at 0°C; b) measurements at 25°C; c) measurements at 30°C; d) measurements at 40°C; e) measurements at 70°C; f) measurements at 100°C; g) measurements at 120°C; h) measurements at 160°C; i) solid-liquid transition
 375 temperatures.

376 3. VOCs absorption using DES

377 In the published works, the authors mainly focused on the absorption of VOCs, which have
378 particularly adverse effects on the environment, human health, and industrial installations.
379 Therefore, the following section presents the achievements in the absorption of particular groups of
380 pollutants, including volatile aliphatic and monoaromatic hydrocarbons, volatile organosulfur-,
381 oxygen-, sulfur-, halogen-, and silicon-organic compounds using DES. The detailed processes
382 condition and obtained results are compared in Table 2.

383 2.1. Volatile hydrocarbons

384 Volatile aliphatic, cyclic, and monoaromatic hydrocarbons are one of the most commonly studied
385 contaminants of air, gaseous fuels, biofuels, and industrial, natural, and waste gasses [158–160].
386 They come from anthropogenic sources such as coal combustion, automobile engines, petroleum
387 refineries, and architectural materials. The group of monoaromatic hydrocarbons includes benzene,
388 ethylbenzene, toluene, xylenes (BTEX), and styrene. The group of aliphatic hydrocarbons consists of
389 linear hydrocarbons from C₂ to C₁₀. From the ecological, public health, and industrial perspectives,
390 the presence of hydrocarbons in air and waste gas streams is unfavorable [161,162]. High toxicity
391 and proven carcinogenic effects on organisms characterize them. They undergo photochemical
392 reactions with ozone [163–167]. In addition, the presence of selected aromatic hydrocarbons in
393 biogas and natural gas streams during fuel combustion can affect corrosion, contamination, and
394 clogging of engine systems [168,169].

395 Several works have demonstrated the high DES absorption efficiency of monoaromatic and
396 aliphatic hydrocarbons from biogas and exhaust gas. In most published works, researchers have
397 focused on removing only toluene as a representative of the monoaromatic hydrocarbons. In these
398 studies, the authors determined vapor-liquid partition coefficients (K) using the static headspace
399 technique to evaluate the effectiveness of the new DES [147,170,171]. According to the definition, K
400 is a parameter that describes the tendency of VOCs to distribute between two phases, liquid, and
401 gas, at equilibrium. It can be calculated as the ratio of the VOCs concentration in the vapor phase in
402 equilibrium to the VOCs concentration in the liquid DES in the equilibrium state [172,173]. In
403 practice, the Headspace procedure involves placing 0.5 - 2 ml of DES in a 20 ml vial and adding a
404 small amount of carefully weighed VOCs. The vial is then sealed with a cap with a membrane and
405 shaken at the specified temperature. After 24 hours, the headspace is drawn and injected into a gas
406 chromatograph. The smaller peaks obtained on the chromatogram indicate lower concentrations of
407 VOCs in the gas phase and their potentially better absorption in DES. In one of the first studies,
408 Moura et al. tested several popular ionic eutectic mixtures based on quaternary ammonium and
409 phosphonium salts such as ChCl:U (1:2), ChCl:EG (1:2), ChCl:Gly (1:2), ChCl:Lev (1:2), TBPB:Gly (1:1),
410 TBPB:Lev (1:6), and TBAB:DA (1:2). They found that the higher ability to absorb volatile
411 monoaromatic hydrocarbons has DES, consisting of tetrabutylammonium bromide and decanoic acid
412 in a 1:2 molar ratio. The K value of toluene in DES was similar to other common solvents, i.e., ionic
413 liquids or silicone oil (K=0.001) and 251-fold at 30 °C and 654-fold lower at 60 °C than water
414 [147,170,171]. In addition, they observed that there is no difference in vapor-liquid partition
415 coefficients at different temperatures. This can be an advantage due to the instability of process
416 gases' temperature. Thus, the lack of difference in absorption capacity may benefit the economics of
417 the process, as it may prevent additional energy expenses used to control the temperature of the gas
418 stream before the purification process [147]. Similar results were obtained by Moufawad et al. They

419 determined vapor-liquid partition coefficients for toluene, n-heptane, and 1-decene in the same ionic
420 DES. This work confirmed that the highest affinity for both monoaromatic and aliphatic hydrocarbons
421 has TBAB:DA (1:2) [148]. The K value for toluene in TBAB:DA (1:2) was comparable to common
422 organic solvents and silicone oils [174,175]. The experimental toluene absorption results were in line
423 with theoretical studies determined using the conductor-like screening model segment activity
424 coefficient (COSMO-SAC). Calculation studies indicated that as the length of the alkyl chain in the salt
425 increases, the absorption capacity of DES increases [150]. In addition, the significant effect of alkyl
426 chain length in carboxylic acids (HBD) on absorption efficiency was shown. The K-value results
427 indicated the same trend as for the HBA component of DES. The longer the alkyl chain of the acid,
428 the better hydrocarbon solubility in DES. The same conclusions were obtained in another
429 experimental study [137]. The lowest value of the vapor-liquid partition coefficient was obtained for
430 1-decene. The K value was, 60000-fold lower than water. The aliphatic hydrocarbons such as n-
431 heptane and 1-decene indicate a similar affinity profile as monoaromatic hydrocarbons for the
432 studied ionic DES. As the authors observed, the absorption capacity of aliphatic hydrocarbons in DES
433 decreases as the number of hydroxyl groups in HBD increases. However, the authors did not explain
434 the reason for this phenomenon [148]. In a study of the removal of styrene from the gas, the authors
435 observed that the crucial parameter determining solubility, and thus absorption capacity, is the
436 general principle of chemistry. This indicates that polar substances dissolve more easily in polar
437 solvents and vice versa. The DES absorption capacity of styrene was ranked as follows: TBABr:DA
438 (1:2) >> TBPBr:Lev (1:6) > ChCl:Xyl:H₂O (2:1:3) > ChCl:PCH:H₂O (1:1:1) > ChCl:Urea (1:2) >
439 ChCl:Glu:H₂O (5:2:5) [134]. Significantly lower values of partition coefficients of toluene were
440 obtained for DES containing cyclodextrins (CD) in their composition. The K values were 0.003, 0.002,
441 0.002, and 0.001 for Captisol:Lev (1:44), CRYSMEB:Lev (1:25), HP-β-CD:Lev (1:32), and RAMEB:Lev
442 (1:27), respectively. The authors demonstrated the affinity DES to toluene is due to the formation of
443 the CD/toluene complex during the absorption process [176–178]. In another paper, the authors
444 proved by 1D ¹H and ROESY spectra with diffusion and relaxation NMR experiments that the water
445 addition to SUPRADES can improve the inclusion of toluene [179]. However, there is a lack of
446 experimental studies to confirm this theory. The results are similar to those obtained for non-ionic
447 DES composed of guaiacol, camphor, and levulinic acid in a 1:1:3 molar ratio [132]. In this system, the
448 driving force of the toluene removal process is most likely π-π and electrostatic interactions. Also,
449 Chen et al. showed high efficiency in removing low concentrations of BTEX from the gas phase with
450 non-ionic DES. The lowest value of Henry's constant was obtained for DDA: DA (1:3). This
451 corresponds with the equilibrium capacity of BTEX absorption equal to 9.94 mg/g at an inlet
452 concentration of 800 ppm v/v [137]. On the other hand, based on COSMO-RS modeling, among 39
453 new absorbents, DES composed of monoterpenes and carboxylic acids such as C:OA (1:1), C:DA (1:1),
454 Car:OA (1:1), Car:NA (1:1), Car:DA (1:1), Car:Lev (1:1), and Eu:OA (1:1) was shown to have the highest
455 affinity for BTEX. The authors suggested that the coexistence of two types of interactions between
456 DES and monoaromatic hydrocarbons (π-π and van der Waals interactions) cause this phenomenon
457 [61]. The structure of monoaromatic hydrocarbons also affects absorption efficiency. The study
458 showed that the absorption capacity of all DES increases in the following order, benzene < toluene <
459 xylene < ethylbenzene. This indicates that the alkyl groups form stronger non-covalent bonds with
460 DES compared to π-π interactions [180]. All obtained data suggest that non-ionic DES or SUPRADES
461 can be better media for volatile hydrocarbon removal from the gas phase. The lowest affinity of
462 hydrocarbons for ionic DES is due to the presence in DES of a strong non-bonded interaction

463 between cation and anion, which is stronger than the electrostatic interactions formed between
464 absorbents and hydrocarbons [181].

465 To fully assess the suitability of DES as absorbents for removing VOCs from gaseous streams, it is
466 necessary to consider both the affinity of DES to pollutants and other absorption process
467 parameters. For this purpose, it is essential to conduct research in a dynamic system, which is a
468 miniaturized version of industrial installations. In such studies, the effects of gas mixture
469 composition, temperature, water addition to DES, gas flow, and pressure on absorption efficiency are
470 commonly investigated. In addition, it is possible to determine the absorption capacity of individual
471 DES [182–184]. So far, several papers have presented the results of BTEX removal using DES in a
472 dynamic system [61,155,185]. All the papers reached similar conclusions about the effect of process
473 parameters on the efficiency of hydrocarbon capture from gas mixtures. The authors showed that
474 the absorption capacity of hydrocarbons gradually decreases with the increase of the DES
475 temperature. This is in line with typical absorbents' behavior (ILs, glycols, and oils) due to the
476 exothermic character of the physical absorption process [183]. Theoretically, an increase in DES
477 temperature decreases viscosity, which can positively affect mass transfer between phases.
478 However, this can't counterbalance the decrease in affinity between hydrocarbons and DES.
479 Therefore, in all works, it was shown that the optimal temperature for the absorption of
480 hydrocarbons was between 10 and 30 °C [25,61,72,132,134,137,147,148,153,155,186–188]. The
481 water addition to DES is advantageous from an industrial point of view because it reduces the cost of
482 the absorbents. However, published work has shown a decrease in the absorption capacity of both
483 linear and aromatic hydrocarbons. This is due to their limited solubility in water. In addition, too
484 much water in DES structures breaks the hydrogen bonds between HBA and HBD [189,190]. From an
485 industrial point of view, the gas flow rate should be relatively high to purify a significant amount of
486 gas within a short period. However, too high a flow rate of contaminated gas reduces the absorption
487 efficiency of DES. This is because as the contaminated gas flow rate increase, the contact time of the
488 gas phase with liquid DES decreases [191]. Another crucial physical absorption parameter is the
489 composition of inlet gas. Most research has focused only on the VOCs removal from an inert gas (N₂).
490 This method only enables the study of interactions between specific contaminants and DES. Nitrogen
491 as an inert gas should not affect the efficiency of absorption. Therefore, it is possible to identify in
492 detail the types and strengths of interactions between DES and hydrocarbons. However, industrial
493 gases, fuels, air, or waste gases contain a wide range of organic and inorganic contaminants. The
494 presence of other impurities can negatively affect the process of hydrocarbon absorption by blocking
495 active groups to which hydrocarbons can potentially attach [61]. However, in some cases, there may
496 be a temporary improvement in the absorption capacity of DES due to incorporating other impurities
497 into DES structures that contain active groups capable of attaching hydrocarbons. Such a
498 phenomenon was observed during biogas purification. Absorbed carbon dioxide created additional
499 active sites that formed hydrogen bonds with -CH...O= hydrocarbons. However, due to the relatively
500 high CO₂ content of the biogas, which causes rapid supersaturation of DES, enhancement of
501 absorption efficiency is observed only for a short period. Then a rapid decrease in process efficiency
502 is observed [61]. Therefore, before using DES in the biogas treatment process, carbon dioxide should
503 be preliminarily removed by other methods [192,193]. The concentration of the absorbed substance
504 is also a crucial parameter. Most of the presented works concern the removal of volatile
505 hydrocarbons at low concentrations from the gas phase. Xu et al. proved the utility of DES in the
506 separation of gaseous hydrocarbons, i.e., C₂H₄/C₂H₆. Due to the similar boiling points of ethane and
507 ethylene, the use of the traditional method of distillation is inefficient and energy-consuming. The

508 authors in the study demonstrated that by selecting the proper molar ratio of TBACl: DA, it is possible
509 to tune the selective solubility of hydrocarbons in DES. They also indicated significantly higher
510 solubility of C₂H₄ in DES than in ionic liquids [145].

511 Among the DES tested to date, the highest absorption efficiency for monoaromatic hydrocarbons
512 was observed for ionic DES composed of quaternary ammonium salts and phenol. The solubility of
513 toluene in DES ranged from 227 to 304 mg/g. The dominant role in the absorption efficiency
514 belonged to phenol (HBD), as well as the benzyl group in HBA that formed π - π , O-H $\cdots\pi$,
515 π \cdots H \cdots Cl⁻ and C-H $\cdots\pi$ interactions with toluene. The results showed that DES that contained more
516 phenol molecules in the structure better-absorbed toluene from the gas. The absorption efficiency
517 for TEBAc:Ph (1:3) was significantly higher than that of the ionic liquid [EMIM][Tf₂N] (223 mg/g) and
518 TEG (163 mg/g) under the same process conditions [25]. Slightly lower efficiencies were found for
519 TEBAc:LA (1:2), TEBAc:LA (1:2), and non-ionic DES in the form of E:OA (1:1). Absorption capacities
520 were 218, 78, and 56 mg/g for toluene and sum of BTEX, respectively [61]. For other DES, the
521 efficiency values ranged from 0.23 to 4.04 mg/g. Absorption efficiencies for the rest group of
522 hydrocarbons were much lower. The highest absorption capacities for cyclohexane (0.058 mg/g),
523 ethylene (0.066 mol/kg), ethane (0.074 mol/kg), n-hexane (0.75 mg/g), n-heptane (2.37 mg/g), 1-
524 decene (0.51 mg/g), and limonene (2.6 mg/g) were observed for ChCl:U (1:1), TBACl:DA (1:1.5),
525 TBACl:DA (1:3), Th:DA (1:1), TBABr:DA (1:2), TBABr:DA (1:2), and NA:DDA (3:1), respectively
526 [25,145,147,148,155]. However, a solid comparison of DES is difficult due to the different conditions
527 of the dynamic absorption process and different methods of evaluating DES effectiveness. In
528 published works, absorption efficiency factors are expressed as Henry's constants, activity
529 coefficients, or solubility of hydrocarbons in DES.

530 All the described studies were performed on a laboratory scale. Only one paper has presented
531 results obtained on an enlarged 20-fold scale. The authors showed that it is possible to scale up the
532 process and receive similar absorption efficiencies to the laboratory process. [61]. However,
533 industrial or semipreparative scale tests are needed to prove the applicability of DES. In addition, no
534 tests have been reported on real waste gas streams or gaseous fuels. All tests that have been
535 performed have been on model streams.

536

537 **2.2. Volatile organosulfur compounds**

538 Volatile organosulfur compounds (VOSCs) are common contaminants in the off-gases of oil
539 refineries, pulp mills, manure and sewer systems, rayon production, and wastewater treatment
540 plants [194–196]. In addition, these compounds can be found in landfill, natural and petroleum gas,
541 and biogas produced from agri-food waste [191,197,198]. Their concentrations strictly depend on
542 the biogas production process. They can vary from a few ppb levels in agricultural emissions to ppm
543 levels in industrial gases [199]. The VOSCs group includes thiols, sulfides, disulfides, and thiophenes.
544 They are characterized by unpleasant smells, even at low concentrations. Typically, the odor
545 threshold values of VOSCs are between 0.07 and 5.9 ppb (v/v). At higher concentration levels (0.5 to
546 20 ppm, v/v), some VOSCs compounds can evoke human health problems such as eye irritation,
547 dizziness, vomiting, and headaches [200–202].

548

549

550

551 In addition, VOSCs can cause many technological problems due to their corrosive and
552 reactive activity. For example, during biogas or natural gas combustion, VOSCs can be converted into
553 SO_x. In the next stage, SO_x can react with water or oxygen and form highly corrosive H₂SO₄. As a
554 result, corrosion of installation equipment, including combustion chamber components, occurs [191].
555 Therefore, the absorption of VOSCs from industrial or waste gas streams is crucial [191].

556 A comparison of three DES composed of choline chloride as HBA and three different HBDs
557 such as phenol (Ph), ethylene glycol (EG), and levulinic acid in a molar ratio of 1:2 showed that it is
558 possible to efficiently capture dimethyldisulfide (DMDS) from a model biogas stream. The authors
559 observed that as the viscosity of DES increased, the absorption efficiency of DMDS decreased. This
560 was due to the hindrance of the mass transfer process. Investigations of the integration mechanisms
561 between DES and DMDS revealed the absence of strong bonds between the sulfur atom and the
562 active groups of DES, including S-H...π, O-H...S and C-H...S. Density functional analysis (DFT) indicated
563 that weaker van der Waals interactions were the main driving force for DMDS absorption. The
564 calculated theoretical DES-DMDS interaction energy showed the same trend as the experimental
565 study ChCl:Ph (1:2) > ChCl:EG (1:2) > ChCl:Lev (1:2) [191]. Other process parameters, such as
566 temperature, the flow rate of contaminated gas, initial concentration of contaminants, or pressure,
567 also affect absorption efficiency. The study showed typical correlations of the absorption process. It
568 has been proven that as the temperature increases, the process efficiency decreases. This is due to
569 the decrease in the solubility of DMDS in DES because the absorption processes are exothermic. The
570 increase in gas flow rate reduces absorption efficiency, which is caused by too short contact between
571 the impurities and the absorbent. In addition, increasing the volume of DES improves absorption due
572 to the prolongation of contact time between the gas and liquid phases, which increases the
573 supersaturation time [21,191,203–205]. Theoretically, the increase of DMDS concentration in inlet
574 gas should result in a linear decrease in the partition coefficients and an increase in the amount of
575 VOCs soluble in DES. However, no changes were observed in the study, which was most likely due to
576 the saturation of DES and reaching the solubility limit of DMDS in the absorbent [148,191]. Another
577 paper also demonstrated the possibility of absorbing DMDS from a model biogas mixture (75% v/v
578 CH₄ and 25% v/v CO₂) using two DES. The authors showed that higher absorption efficiency was
579 obtained with non-ionic DES (C:Gu 1:1) than with ionic ChCl:U (1:1). However, the authors did not
580 explain why such results were obtained, and other parameters affecting DMDS uptake were not
581 studied [187]. Panda et al. investigated the possibility of using supramolecular low-melting eutectic
582 mixtures for thiophene absorption through the static headspace technique. Based on the K-factor
583 results, they showed that DES have about 30 times better thiophene absorption capacity than water,
584 and twice better compared to conventional non-cyclodextrin DES i.e., ChCl:Lev (1:2). The vapor-liquid
585 partition coefficient was 0.139, 0.009, 0.006, 0.005, 0.006, 0.009 for water Captisol:Lev (1:44),
586 CRYSMEB:Lev (1:25), HP-β-CD:Lev (1:32), RAMEB:Lev (1:27), ChCl:Lev (1:2), respectively. However,
587 the authors did not explain what mechanisms determine the high uptake of thiophene. The paper
588 also does not present studies in a dynamic system [177].

590 2.2. Volatile methyl siloxanes

591 Volatile methyl siloxanes (VMS) named siloxanes are commonly used in personal care
592 products, household cleaning, and many branches of industries as solvents or lubricants. Therefore,
593 VMS can easily migrate into the atmosphere [206]. The group of volatile siloxanes that are most
594 commonly identified in air and gaseous fuels include cyclic VMS such as



595 hexamethylcyclotetrasiloxane (D3), octamethylcyclotetrasiloxane (D4),
596 decamethylcyclotetrasiloxane (D5), dodecamethylcyclotetrasiloxane (D6), and aliphatic siloxanes
597 including hexamethyldisiloxane (L2), octamethyltrisiloxane (L3), decamethyltetrasiloxane (L4) and
598 dodecamethylpentasiloxane (L5). The impact on human health and the environment is currently
599 quite controversial. Some studies show the possibility of VMS bioaccumulation in biota, endocrine-
600 disrupting effects of D4, and immunosuppressant and carcinogenic effects of D5. While other studies
601 prove the absence of negative impact on the environment and organisms [207–209]. The presence
602 of VMS in biogas has unfavorable effects on combustion equipment components. The combustion of
603 VMS-rich biogas in burners results in the formation of solid silicon dioxide (SiO₂) particles. SiO₂ can
604 deposit on turbine blades and heat exchangers, shortening their lifespan and increasing plant
605 maintenance costs, reducing energy efficiency, and causing numerous malfunctions. [210,211].

606 In one of the published papers, the authors performed calculations of the ability of selected
607 ionic DES to absorb 200 ppm v/v of VMS (L2, L3, and D4) from a model biogas mixture composed of
608 CH₄:CO₂:H₂S (68:31:1 % v/v). The authors divided the studied DES into three groups. The first group
609 included absorbents that effectively captured all impurities, including CO₂ siloxanes and H₂S. The
610 second group consisted of DES that selectively captured only siloxanes, while the third group
611 included other DES that showed no absorption efficiency for any pollutants. ChCl: U (1:2) and ChCl:
612 OA (1:2) were selected as the most effective DES. The study proved that the type of HBA has the
613 main effect on the efficiency of contaminant capture. The application of DES that contains ChCl is
614 favorable due to lower viscosity than DES composed of QAS and QPS with long alkyl chain lengths.
615 [68,131]. However, the paper did not include an experimental study. All conclusions were based on
616 theoretical studies using the COSMO-RS model. The COSMO-RS is a fully predictive model because no
617 experimental data, functional group parameters, or system-specific adjustments are needed to
618 predict thermodynamic properties and the calculation of interactions between molecules. This is an
619 advantage of the model, but it can also affect the lower accuracy compared to locally adapted
620 methods. [212]. Authors carried out more reliable experimental studies of VMS uptake for glycols
621 such as ethylene glycol (EG), glycerol (Gly), triethylene glycol (TriEG), tetraethylene glycol (TEG),
622 diethylene glycol (DEG), and DES composed of glycols and QAS. From an economic point of view,
623 pure glycols application is preferable due to the relatively high price of QAS. However, as the authors
624 observed, the solubility of VMS in DES is much higher than in glycols. In addition, VMS capture
625 efficiency decreases as the glycol content of the DES structure increases. The addition of TPABr
626 lowers the polarity of the DES molecule, which increasingly adopts a similar character to pure VMS.
627 Among the DES tested, TPABr:TEG (1:3) had the highest solubility due to the content of more -O-
628 active groups in the glycol structure, which can form weak hydrogen bonds with -CH₃ groups in VMS
629 structures. [149]. Nevertheless, a comparison of ionic and non-ionic DES showed higher efficiency of
630 VMS capture by eutectic complexes composed of non-ionic components [132]. In another work,
631 authors tested 90 non-ionic hydrophobic DES composed of terpenes, carboxylic acids, and
632 polyphenols as potential green absorbents for L2, L3, L4, D3, and D5 removal from model biogas,
633 using COSMO-RS model. The authors compared the logarithmic activity coefficients of individual
634 siloxanes in DES. The eutectic complexes composed of carvone and carboxylic acids, i.e., DA, UDA,
635 and DDA in a 1:1 molar ratio, showed the highest affinity for VMS. This is due to the simple structure
636 of DES components. Carvone has one carbonyl group, which acts as a hydrogen bond acceptor, while
637 DA, UDA, and DDA contain a carboxyl group, which is a hydrogen bond donor. The absence of active
638 groups, i.e., =O, -O-CH₃, -CH₃, increases the access of the oxygen atom from the Si-O-Si to the
639 carboxyl group to which it could attach. On the other hand, the presence of these groups reduces the

640 strength of interactions between siloxanes and DES and reduces their solubility in DES. The paper
641 demonstrated that cyclic siloxanes absorb significantly better in DES than linear siloxanes. This is due
642 to the higher number of Si-O bonds, which tend to form stronger bonds between siloxanes and DES.
643 The lowest absorption efficiency was observed for L2. The lowest absorption efficiency was observed
644 for L2 because of its high vapor pressure, and polarity. An additional advantage of Car:DA (1:1) is the
645 low solubility of methane. Methane was captured below 1% by DES. This value is within the
646 acceptable range [152].

647 **2.4. Oxygenated volatile organic compounds**

648 Oxygenated volatile organic compounds (O-VOCs) are a specific group of VOCs that contain
649 at least one atom of oxygen, i.e., aldehydes, ketones, alcohols, phenols, esters, ethers, and carboxylic
650 acids. They are characterized by relatively high toxicity and vapor pressure, mutagenic, and
651 malodorous properties. In addition, they are highly reactive. O-VOCs play an active role in
652 tropospheric ozone formation [7,213–215]. The source of O-VOCs in the environment is primarily
653 industrial installations due to the widespread use of solvents in paints [216,217], pharmaceuticals
654 [218], polymer processing, and the refinery industry [219]. They can harm the human respiratory
655 system, skin, and eyes. Therefore, it is crucial to eliminate O-VOCs emissions from various industrial
656 sources.

657 Due to the presence of active groups in O-VOC, such as -OH, =O, -O-, -COOH, they can be
658 absorbed by DES with high efficiency. During the absorption, active O-VOCs groups can form strong
659 hydrogen bonds with DES components. However, there are only a few reports in the literature on the
660 removal of O-VOCs from gas streams by absorption using DES. The authors mainly focused on
661 determining the vapor-liquid partition coefficients of O-VOCs between DES and the gas phase. In one
662 work, a preliminary preselection based on the determined K values showed that non-ionic DES could
663 slightly better capture propionaldehyde compared to ionic DES, i.e., ChCl:U (1:2), ChCl:Lev (1:2);
664 ChCl:DEG (1:2). The results were comparable to conventional absorbents based on n-hexadecane. On
665 the other hand, the results demonstrated that water is characterized by the higher absorption
666 efficiency of O-VOCs than most DES [132]. Similar results were obtained for SUPRADES. The authors
667 in their study showed that there is no significant difference in the absorption efficiency of
668 acetaldehyde and butanone between different SUPRADES and water. The high polarity of O-VOCs
669 hinders their incorporation into the hydrophobic cavity of the CD molecule in the SUPRADES
670 structure [177]. Methyl ethyl ketone (MEK) also shows a higher affinity for water than ionic DES. On
671 the other hand, the authors obtained lower K values of acetaldehyde in ionic DES (i.e., ChCl: U,
672 ChCl:EG, ChCl:G, ChCl:Lev, TBPBr:EG, TBPBr:G, TBPBr:Lev, and TBABr:DA) compared to water, which
673 contradicts the previous studies [132,185]. Indra et al. compared the acetaldehyde uptake potential
674 of two popular DES (ChCl:U (1:2) and ChCl:Lev (1:2)) using the molecular dynamics (MD) simulation
675 analysis. The authors demonstrated that acetaldehyde absorbs better in ChCl:U (1:2). This is due to
676 the occurrence of non-polar interactions at the liquid-vapor interface, which increases the absorption
677 of acetaldehyde. The authors obtained similar results of partition coefficients to those obtained by
678 the experimental method [147,220]. In addition, they showed that DES has a twofold higher
679 absorption capacity compared to ionic liquid in the form of [BMIM]PF₆ [221].

680 Chen et al. researched acetone removal using hydrophobic and hydrophilic DES. Acetone is
681 hydrophilic due to its high polarity (2.88 D). Therefore, it should dissolve better in hydrophilic DES
682 [222]. However, hydrophobic DES composed of Th and DA (1:1) showed a higher absorption capacity

683 than hydrophilic ChCl:Res (1:2). The absorption capacities were 2.66 and 0.76 mg/g for hydrophobic
684 and hydrophilic DES, respectively. The results indicate that the polar nature of DES does not
685 guarantee the high absorption efficiency of polar VOCs. The efficiency is more affected by the low
686 viscosity and the presence of specific active groups in the DES structures capable of H-bonding
687 formation [153].

688

689 **2.6. Volatile organochlorine compounds**

690 Volatile organochlorine compounds (VOX) are commonly present in waste gas streams from
691 paint manufacturing, plastic manufacturing, foam, and electronic industries [223–225]. In addition,
692 they can be identified in landfill gas or biogas produced from sewage sludge [226]. Some of these
693 compounds have been recognized by the United States Environmental Protection Agency (US EPA)
694 and the European Environment Agency (EEA) as extremely hazardous gaseous pollutants that
695 adversely affect both human health and the environment [227]. Uncontrolled emissions of VOX to
696 the atmosphere can cause the formation of photochemical smog and global warming. Exposure of
697 humans to organochlorine substances can cause irritation symptoms, cancer, allergies, asthma, and
698 neurological and liver toxicity. These compounds are also technologically disadvantageous due to
699 their highly corrosive properties, which can shorten the lifespan of many industrial installation
700 components. [228].

701 Most scientific papers describe the removal of only dichloromethane (DCM) from the gas
702 phase. Moura et al. observed that from seven ionic DES, the TBABr:DA and TBPBr:Lev were the most
703 effective absorbents for DCM removal. The absorption capacity was 28- and 23-fold higher than
704 water at 30 °C [147]. However, the paper does not explain which interactions contributed to the
705 selection of mentioned DES. In another study, the same DES were applied to the absorption of DCM.
706 The authors confirmed that DES, composed of alkylphosphonium or alkylammonium salts with long
707 alkyl chains, promotes the removal of DCM from the gas phase. This is because the longer alkyl
708 chains in HBA affect the increasing hydrophobic nature of DES [148]. Similar conclusions were
709 obtained for other absorbents, i.e., silicone oil or ionic liquids [174,229,230]. Mu et al. explained that
710 this is due to the steric hindrance in DES structures. This hindrance increases the distance between
711 the anion and the cation. The authors also showed that DES composed of non-cyclic cation HBAs
712 (QAS and QPS) have a higher absorption capacity than cyclic HBAs (i.e., pyridinium and imidazolium-
713 based) due to the low polarity of non-cyclic cations. Among the 28 DES examined, TBPCl:Lev (1:2)
714 showed the highest DCM absorption capacity (899 mg/g) comparable to traditional organic solvents.
715 This is due to the formation of relatively strong non-covalent interactions between DCM and Lev (C-
716 H...O) [146]. Another group of scientists tested four SUPRADES as green absorbents for DCM removal
717 from the gas phase. The absorption capacities ranged from 0.274 to 0.3 mg/g. This indicates that the
718 type of cyclodextrin has only a minor effect on the absorption capacity of DCM [177].

719 However, due to the strong influence of CD type on the physicochemical properties of
720 SUPRADES, they can play a crucial role in dynamic absorption processes. However, no such studies
721 have been performed to date. Inferior DCM partition coefficient results were obtained for G:C:Lev
722 (1:1:3) compared to SUPRADES and ionic DES. The lower absorption capacity of G:C:Lev (1:1:3) may
723 be due to the competitiveness of interactions formed with toluene, propionaldehyde, or
724 hexamethyldisiloxane [132]. In other studies, experiments were conducted on a single impurity
725 removal from an inert gas (nitrogen). Only one paper has described research on the application of
726 DES to the dynamic absorption of other VOX substances such as 1,1,2,2-tetrachloroethane (TCE),

727 2,2,2-trichloroethanol (TCeOH), chloroform (CF), and carbon tetrachloride (TCM) from model
728 gaseous fuels. In these studies, the authors tested four DES such as Syr : Lev (1:1), C: Gu (1 : 1), C: Lev
729 (1:2), and Gu: Lev (1:1). They showed that the highest absorption capacity was found in Syr:Lev (1:1)
730 due to the formation of van der Waals interactions between DES and most VOX. Only between DES
731 and TCeOH authors identified the coexistence of van der Waals interactions and hydrogen bonds
732 between the hydroxyl groups of Syr, Lev, and TCeOH (OH...HO). The bond lengths were 1.79 and 1.8
733 Å, respectively. The ability to form relatively strong non-covalent interactions is the reason for the
734 high absorption capacity of DES, which was 420 ± 22 , 360 ± 16 , 304 ± 11 , 292 ± 8 , and 661 ± 34 mg g-
735 1 for DCM, CF, TCM, TCE, and TCeOH, respectively. These values are 100 times higher than for ILs
736 and ionic DES composed of quaternary ammonium salts (i.e., ChCl, TBABr, or TPABr) [147,231]. Only
737 for the metal-organic framework (UiO-66) was it possible to achieve a higher absorption capacity
738 [232]. However, as the authors observed, the price of UiO-66 was about 300 times higher than DES.
739 On the other hand, such a high price makes it impossible to use the sorption medium in industrial
740 processes. The effect of the rest of the parameters on absorption efficiency was similar to the other
741 VOCs groups [226].

742 2.7. Volatile fluorinated gases

743 Volatile fluorinated gases (VFG) such as sulfur hexafluoride, perfluorocarbons, and
744 hydrofluorocarbons are dangerous greenhouse gaseous. They are mainly used in refrigeration and air
745 conditioning. VFGs are responsible for climate change and ozone depletion. The global warming
746 potential (GWP) of VFG is up to 23 000 times higher than that of CO₂. Many countries have
747 introduced legislation to limit VFG emissions into the atmosphere. However, capturing VFG from the
748 air remains a challenge [233–235].

749 Only one study has examined the possibility of using DES to capture VFG from the air. The
750 authors tested a DES composed of fluorinated ionic liquids and perfluorinated acids for
751 difluoromethane (R-32), pentafluoroethane (R-125), and 1,1,1,2-tetrafluoroethane (R-134a)
752 absorption. The authors selected two DES including [N₄₄₄₄][C₄F₉SO₃]/C₄F₉CO₂H and
753 [C₂C₁Im][C₈F₁₇SO₃]/C₄F₉CO₂H as the most efficient absorbents.

754 The authors observed that the cationic nature of fluorinated ionic liquids has a major effect
755 on the solubility of VFG in DES. On the other hand, the type of anion did not affect the absorption
756 process. In addition, they observed that DES has high selectivity toward R-134a in binary gas mixtures
757 at pressures above 0.2 MPa. Therefore, new absorbents can also be used to recover commercial
758 refrigerants for re-use. However, only one paper has been published so far. Therefore, it is
759 challenging to assess whether other types of DES would be preferable for VFG absorption [157].

760 **Table 2** Summary of studies on VOCs absorption from gas streams using DES.

VOC	DES (HBA:HBD molar ratio)	Gas matrix	Absorption capacity / solubility [mg/g]	Saturation time [min]	Vapor-liquid partition coefficients (K)/ Henry's Law Constant*	Absorption process conditions	Desorption process conditions	The absorption/desorption cycles	Ref.
VOLATILE HYDROCARBONS									
toluene	Captisol:Lev (1:44)	n.d.	0.282	n.d.	0.003	<u>Static headspace analysis:</u> Initial concentration: 59 g m ⁻³ Temperature: 30°C Time of equilibrium: 24 h DES volume: n.d.	Thermal desorption at 60°C, 48 h	5	[177]
	CRYSMEB:Lev (1:25)	n.d.	0.287	n.d.	0.002			5	
	HP-β-CD:Lev (1:32)	n.d.	0.285	n.d.	0.002			5	
	RAMEB:Lev (1:27)	n.d.	0.287	n.d.	0.001			5	
	ChCl: Lev (1:2)	n.d.	0.280	n.d.	0.004			5	
toluene	ChCl:DEG (1:2)	N ₂	n.d.	> 100	n.d.	<u>Dynamic absorption:</u> Initial concentration: 2000 ppm v/v Temperature: 30°C Flow rate: 25 mL/min Pressure: 10 kPa DES volume: 30 mL	n.d.	n.d.	[186]
	ChCl:U (1:2)	N ₂	n.d.	65	n.d.		n.d.	n.d.	
	ChCl:U:TEG (1:2)	N ₂	n.d.	> 100	n.d.		n.d.	n.d.	
toluene n-heptane 1-decene	ChCl:U (1:2)	Air	0.275 n.d. n.d.	n.d.	0.25 11 1.2	<u>Static headspace analysis:</u> Initial concentration: 59 g m ⁻³ Temperature: 30°C Time of equilibrium: 24 h <u>Dynamic absorption:</u> Initial concentration: n-heptane: 885 g/m ³ ; 1-decene: 4720 g/m ³ Flow rate: n.d. Pressure: n.d. DES mass: 30 g	Thermal desorption at 60°C, 48 h	5	[147,148]
	ChCl:EG (1:2)	Air	0.26 n.d. n.d.	n.d.	0.096 0.018 0.64			5	
	ChCl:Gly (1:2)	Air	0.23 n.d. n.d.	n.d.	0.022 0.018 1,1			5	
	ChCl:Lev (1:2)	Air	0.55 0.01 n.d.	n.d.	0.054 0.27 0.029			5	
	TBPB:Gly (1:1)	Air	0.28 n.d. n.d.	n.d.	0.005 0.11 0.0011			5	
	TBPB:Lev (1:6)	Air	1.42 0.1 n.d.	n.d.	0.0013 0.034 0.0015			5	



	TBAB:DA (1:2)	Air	0.32 2.37 0.51	n.d.	0.0009 0.0033 0.0004			5	
toluene	G:C:Lev (1:1:3)	N ₂	n.d.	1800	<0.001	<u>Static headspace analysis:</u> Initial concentration: 50 ppm v/v Temperature: 25°C Volume of absorbent: 1 mL Time of equilibrium: 48 h <u>Dynamic absorption:</u> Initial concentration: 50 ppm v/v Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL	Adsorption with activated carbon	5	[132]
	ChCl:U (1:2)	N ₂	n.d.	n.d.	0.026			5	
	ChCl:Lev (1:2)	N ₂	n.d.	n.d.	0.035			5	
	ChCl:DEG (1:2)	N ₂	n.d.	n.d.	0.076			5	
toluene	DDA:DA (1:2)	N ₂	3.54	n.d.	n.d.	<u>Dynamic absorption:</u> Initial concentration: 400 ppm v/v Temperature: 30°C Flow rate: 150 mL min ⁻¹ Pressure: 10 kPa DES mass: 3 g	thermal distillation 80 °C in continuously flowing nitrogen at a flow rate of 90 mL min ⁻¹ for 3 h	5	[137]
	DDA:DA (1:3)	N ₂	4.04	120	0.79 (Pa × m ³)/mol			5	
	DDA:DA (1:4)	N ₂	3.3	120	n.d.			5	
	Lid:DA (1:2)	N ₂	2.26	45	n.d.			5	
	Th:DA (1:1)	N ₂	2.2	60	n.d.			5	
	TBPBr:DA (1:2)	N ₂	0.8	70	n.d.			5	
	OA:DA (2:1)	N ₂	0.68	30	n.d.			5	
ChCl:Res (1:2)	N ₂	0.51	25	n.d.	5				
toluene	TEACl:Ph (1:2)	N ₂	227	n.d.	n.d.	<u>Dynamic absorption:</u> Initial concentration: 20000 ppm v/v Temperature: 25°C Flow rate: 100 mL min ⁻¹ Pressure: 101.3 kPa DES mass: 2 g	Vacuum dryer; pressure: 0.05 bar; Temperature: 60°C Time 2 h	10	[25]
	TAACl:Ph(1:2)	N ₂	198	n.d.	n.d.			10	
	TEBACl:LA (1:2)	N ₂	219	n.d.	n.d.			10	
	TEBACl:EG (1:2)	N ₂	78	n.d.	n.d.			10	
	TEBACl:Ph (1:2)	N ₂	257	n.d.	n.d.			10	
	TEBACl:Ph (1:3)	N ₂	304	n.d.	n.d.			10	
toluene	ChCl:Lev (1:2)	n.d.	n.d.	n.d.	0.0045	<u>Static headspace analysis:</u> Initial concentration: 3000 g/m ³ Temperature: 30 °C Mass of absorbent: 3.4 g Time of equilibrium: 48 h	Thermal desorption at 60°C, 48 h	n.d.	[72]
	ChCl:Lac (1:2)	n.d.	n.d.	n.d.	0.0101			n.d.	
	Chol-C6-Lev	n.d.	n.d.	n.d.	0.0021			n.d.	
	Chol-C8-Lev	n.d.	n.d.	n.d.	0.0016			n.d.	
	Chol-C6-Lac	n.d.	n.d.	n.d.	0.0028			n.d.	
	Chol-C8-Lac	n.d.	n.d.	n.d.	0.0020			n.d.	
toluene	ChCl:U (1:2)	n.d.	n.d.	n.d.	0.002	Molecular dynamics (MD) simulation technique (30°C)	n.d.	n.d.	[220]
	ChCl:Lev (1:2)	n.d.	n.d.	n.d.	0.002			n.d.	
benzene	E:OA (1:1)	CH ₄ :CO ₂ :N	Sum of	3118	0.027	<u>Dynamic absorption:</u>	Nitrogen barbotage at	10	[61]



ethylbenzene toluene xylene		2 (50%:30%: 20% v/v)	BTEX: 56	4319 5731 5859	0.018 0.013 0.013	Initial concentration: 2000 mg/m ³ Temperature: 10°C Flow rate: 500 mL/min Pressure: 1 atm DES volume: 1L	120°C for 3 h					
	benzene ethylbenzene toluene xylene	Cam:DA (1:1)	N ₂	n.d.	1536 3221 4452 4562			0.035 0.024 0.02 0.019	<u>Dynamic absorption:</u> Initial concentration: 2000 mg/m ³ Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL <u>Static headspace analysis:</u> Initial concentration: 1.5 µL/mL Temperature: RT Volume of absorbent: 0.5 mL Time of equilibrium: 24 h	Nitrogen barbotage at 120°C for 3 h	10	[61]
		Cam:OA (1:1)	N ₂	n.d.	2365 3251 4571 4579			0.034 0.027 0.021 0.02			10	
C-one:Lev (1:1)		N ₂	n.d.	2103 3065 4498 4507	0.038 0.028 0.026 0.025	10						
C-one:OA (1:1)		N ₂	n.d.	3108 3215 4165 4007	0.031 0.029 0.021 0.019	10						
C-one:NA (1:1)		N ₂	n.d.	2789 4256 4259 4264	0.033 0.023 0.022 0.02	10						
C-one:DA (1:1)		N ₂	n.d.	1997 4984 5845 5856	0.035 0.026 0.016 0.015	10						
E:OA (1:1)		N ₂	n.d.	2789 5898 5926 5946	0.027 0.018 0.013 0.013	10						
styrene	ChCl:U (1:2)	n.d.	n.d.	n.d.	0.12	<u>Static headspace analysis:</u> Initial concentration: 0.5 – 5 µL/mL Temperature: 28°C Volume of absorbent: 0.5 mL Time of equilibrium: 72 h	n.d.	n.d.	[134]			
	TBPBr:Lev (1:6)	n.d.	n.d.	n.d.	0.02			n.d.				
	TBABr:DA (1:2)	n.d.	n.d.	n.d.	0.001			n.d.				
	ChCl:PCH:H2O (1:1:1)	n.d.	n.d.	n.d.	0.06			n.d.				
	ChCl:Glu:H2O (5:2:5)	n.d.	n.d.	n.d.	0.13			n.d.				



	ChCl:Xyl:H2O (2:1:3)	n.d.	n.d.	n.d.	0.04			n.d.	
toluene limonene	OA:DDA (3:1)	N ₂	n.d.	n.d.	0.00048 0.00012	<u>Static headspace analysis:</u> Initial concentration: 59 mg/g Temperature: 30°C Mass of absorbent: 3.5 g Time of equilibrium: 24 h <u>Dynamic absorption:</u> Initial concentration: 59 -531 ppm Flow rate: 15µL/h Pressure: 10 kPa Temperature: 30°C DES mass: 20 g	Nitrogen barbotage (flow rate: 40 L/h) at 30°C for 5 h.	5	[155]
	NA:DDA (3:1)	N ₂	0.29 - 2.6	n.d.	0.00046 0.00012			5	
	DA:DDA (2:1)	N ₂	0.7	n.d.	0.00051 0.00011			5	
	Rameb:Lev (7:3)	N ₂	n.d.	n.d.	0.0018 0.00062			5	
	ChCl:Lev (2:1)	N ₂	n.d.	n.d.	0.0042 n.d.			5	
Benzene hexane	Th:DA (1:1)	N ₂	1.98 0.75	n.d.	n.d.	<u>Dynamic absorption:</u> Initial concentration: 1480 ppm v/v Temperature: 30°C Flow rate: 150 mL/min Pressure: 10 kPa DES mass: 3 g	Nitrogen barbotage (flow rate: 150 mL min ⁻¹) at 80°C.	5	[153]
C₂H₄ C₂H₆	TBACl:DA (1:3)	C ₂ H ₄ :C ₂ H ₆	0.0636 mol/kg 0.0741	n.d.	15.7 13.5 100kPa·kg/ mol	<u>Dynamic absorption:</u> Initial concentration: 50:50 %, v/v Temperature: 40°C Flow rate: n.d. Pressure: 1.0 bar DES mass: 2 g	Depressurizing method (pressure: 0.1 kPa, temperature: 313 K , time 1h.	10	[145]
C₂H₄ C₂H₆	TBACl:DA (1:2)	C ₂ H ₄ :C ₂ H ₆	0.0646 mol/kg 0.0719	n.d.	15.5 13.9 100kPa·kg/ mol			10	
C₂H₄ C₂H₆	TBACl:DA (1:1.5)	C ₂ H ₄ :C ₂ H ₆	0.0661 mol/kg 0.0665	n.d.	15.1 15.0 100kPa·kg/ mol			10	
C₂H₄ C₂H₆	TBACl:DA (1:1)	C ₂ H ₄ :C ₂ H ₆	0.0632 mol/kg 0.0568	n.d.	15.8 17.6 100kPa·kg/ mol			10	
C₂H₄ C₂H₆	TBACl:DA (2:1)	C ₂ H ₄ :C ₂ H ₆	0.0651 mol/kg 0.0497	n.d.	15.4 20.1 100kPa·kg/ mol			10	[145]
cyclohexane	ChCl:U (1:2)	CH ₄ :CO ₂	n.d.	n.d.	n.d.	<u>Dynamic absorption:</u>	n.d.	n.d.	[187]



		(75:25% v/v)				Initial concentration: 16 ppm Temperature: Flow rate: 300 cm ³ min ⁻¹ Pressure: n.d. DES volume: n.d.			
	C:Gu (1:1)	CH ₄ :CO ₂ (75:25% v/v)	n.d.	n.d.	n.d.		n.d.	n.d.	
cyclohexane	ChCl:EG (1:1)	N ₂	0.01465	n.d.	72.6 bar (30 °C)	<u>Dynamic absorption:</u> Initial concentration: 400 ppm Temperature: 30°C Flow rate: 15 ml min ⁻¹ Pressure: 1 bar DES volume: n.d.	Regenerayion in vacuum drying oven (0.1 kPa 2 h, 333.2 K)	10	[188]
	ChCl:EG (1:2)	N ₂	0.003820	n.d.	260.4*			10	
	ChCl:EG (1:3)	N ₂	0.00178	n.d.	562.2*			10	
	ChCl:U (1:1)	N ₂	0.05812	n.d.	14.4*			10	
	ChCl:U (1:2)	N ₂	0.04682	n.d.	15.0*			10	
	ChCl:U (1:3)	N ₂	0.04749	n.d.	19.3*			10	
VOLATILE ORGANOSULFUR COMPOUNDS									
Thiophene	Captisol:Lev (1:44)	n.d.	n.d.	n.d.	0.009	<u>Static headspace analysis:</u> Initial concentration: 59 g m ⁻³ Temperature: 30°C Time of equilibrium: 24 h DES volume: n.d.	Thermal desorption at 60°C, 48 h	5	[177]
	CRYSMEB:Lev (1:25)	n.d.	n.d.	n.d.	0.006			5	
	HP-β-CD:Lev (1:32)	n.d.	n.d.	n.d.	0.005			5	
	RAMEB:Lev (1:27)	n.d.	n.d.	n.d.	0.006			5	
	ChCl: Lev (1:2)	n.d.	n.d.	n.d.	0.009			5	
DMDS	ChCl:Ph (1:2)	N ₂	n.d.	1050	n.d.	<u>Dynamic absorption:</u> Initial concentration: 1 mg/Nm ³ Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL	Nitrogen barbotage at RT for 5 h.	5	[191]
	ChCl:Lev (1:2)	N ₂	n.d.	800	n.d.			5	
	ChCl:EG (1:2)	N ₂	n.d.	600	n.d.			5	
DMDS	C:Gu (1:1)	CH ₄ :CO ₂ (75:25% v/v)	n.d.	n.d.	n.d.	<u>Dynamic absorption:</u> Initial concentration: 16 ppm Temperature: RT Flow rate: 300 cm ³ min ⁻¹ Pressure: n.d. DES volume: n.d.	n.d.	n.d.	[187]
	ChCl:U (1:2)		n.d.	n.d.	n.d.		n.d.	n.d.	
Volatile organochlorine compounds									
DCM	ChCl:U (1:2)	Air	0.2	n.d.	0.055	<u>Static headspace analysis:</u> Initial concentration: 59 g m ⁻³ Temperature: 30°C Time of equilibrium: 24 h	Thermal desorption at 60°C, 48 h	5	[147][148]
	ChCl:EG (1:2)	Air	0.26	n.d.	0.034			5	
	ChCl:Gly (1:2)	Air	0.24	n.d.	0.037			5	
	ChCl:Lev (1:2)	Air	0.27	n.d.	0.008			5	
	TBPB:Gly (1:1)	Air	0.28	n.d.	0.0053			5	
	TBPB:Lev (1:6)	Air	0.29	n.d.	0.006			5	
	TBAB:DA (1:2)	Air	0.3	n.d.	0.005			5	
DCM	Captisol:Lev (1:44)	n.d.	0.27	n.d.	0.013	<u>Static headspace analysis:</u>	Thermal desorption at	5	[177]



	CRYSMEB:Lev (1:25)	n.d.	0.275	n.d.	0.010	Initial concentration: 59 g m ⁻³ Temperature: 30°C Time of equilibrium: 24 h DES volume: n.d.	60°C, 48 h	5	
	HP-β-CD:Lev (1:32)	n.d.	0.275	n.d.	0.010			5	
	RAMEB:Lev (1:27)	n.d.	0.279	n.d.	0.009			5	
	ChCl: Lev (1:2)	n.d.	0.274	n.d.	0.011			5	
DCM	TBPCI:Lev (1:2)	N ₂	899 mg/g (5.58 mol/mol)	40	n.d.	Dynamic absorption: initial DCM partial pressure 5.53 kPa Temperature: 30°C Flow rate: 100 mL/min DES mass: 2g	Heating under vacuum at 100 °C and 0.5 kPa for 8 h.	10	[146]
	TBPCI:Lev (1:1)	N ₂	988 mg/g	n.d.	n.d.			10	
	TBPCI:Lev (1:3)	N ₂	860 mg/g	n.d.	n.d.			10	
	TBPCI:Lev (1:4)	N ₂	843 mg/g	n.d.	n.d.			10	
	TEACl:Lev (1:2)	N ₂	800	n.d.	n.d.			10	
	TEACl:Ph (1:2)	N ₂	541	n.d.	n.d.			10	
	TEACl:EG (1:2)	N ₂	498	n.d.	n.d.			10	
	BPYCl:Lev (1:2)	N ₂	758	n.d.	n.d.			10	
	TBACl:Lev (1:2)	N ₂	867	n.d.	n.d.	10			
DCM	G:C:Lev (1:1:3)	N ₂	n.d.	1000	0.057	Static headspace analysis: Initial concentration: 50 ppm v/v Temperature: 25°C Volume of absorbent: 1 mL Time of equilibrium: 48 h Dynamic absorption: Initial concentration: 50 ppm v/v Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL	Adsorption with activated carbon	5	[132]
	ChCl:U (1:2)	N ₂	n.d.	n.d.	0.052			5	
	ChCl:Lev (1:2)	N ₂	n.d.	n.d.	0.075			5	
	ChCl:DEG (1:2)	N ₂	n.d.	n.d.	0.038			5	
DCM	ChCl:Lev (1:2)	n.d.	n.d.	n.d.	0.0105	Static headspace analysis: Initial concentration: 3000 g/m ³ Temperature: 30 °C Mass of absorbent: 3.4 g Time of equilibrium: 48 h	Thermal desorption at 60°C, 48 h	n.d.	[72]
	ChCl:Lac (1:2)	n.d.	n.d.	n.d.	0.0211			n.d.	
	Chol-C6-Lev	n.d.	n.d.	n.d.	0.009			n.d.	
	Chol-C8-Lev	n.d.	n.d.	n.d.	0.0071			n.d.	
	Chol-C6-Lac	n.d.	n.d.	n.d.	0.0141			n.d.	
	Chol-C8-Lac	n.d.	n.d.	n.d.	0.0105			n.d.	
DCM CF TCM TCE TCeOH	Syr:Lev (1:1)	CH ₄ :CO ₂ :H ₂ O:N ₂ (58:38:2:2 % v/v)	304 420 360 292	800 700 450 580	n.d.	Dynamic absorption: Initial concentration: 0.5 mg cm ⁻³ Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL	Nitrogen barbotage (flow rate: 70 mL min ⁻¹) at 100°C for 1 h.	10	[74]
	C:Gu (1:1)	CH ₄ :CO ₂ :H	215	500	n.d.			10	

		$_2\text{O}:\text{N}_2$ (58:38:2:2 % v/v)	561.5 320 262.4 275.3	620 400 600 1200					
	C:Lev (1:2)	$\text{CH}_4:\text{CO}_2:\text{H}_2\text{O}:\text{N}_2$ (58:38:2:2 % v/v)	181 401.5 143.5 248 198.15	275 220 225 280 650	n.d.			10	
	Gu:Lev (1:1)	$\text{CH}_4:\text{CO}_2:\text{H}_2\text{O}:\text{N}_2$ (58:38:2:2 % v/v)	130.7 399.5 115.8 154 161.2	200 550 250 350 800	n.d.			10	
OXYGENATED VOLATILE ORGANIC COMPOUNDS (O-VOCS)									
acetaldehyde butanone	Captisol:Lev (1:44)	n.d.	0.275 0.281	n.d.	0.007 0.005	<u>Static headspace analysis:</u> Initial concentration: 59 g m^{-3} Temperature: 30°C Time of equilibrium: 24 h DES volume: n.d.	Thermal desorption at 60°C , 48 h	5	[177]
	CRYSMEB:Lev (1:25)	n.d.	0.282 0.285	n.d.	0.005 0.003			5	
	HP- β -CD:Lev (1:32)	n.d.	0.278 0.282	n.d.	0.006 0.004			5	
	RAMEB:Lev (1:27)	n.d.	0.278 0.283	n.d.	0.007 0.005			5	
	ChCl: Lev (1:2)	n.d.	0.283 0.277	n.d.	0.001 0.007			5	
acetaldehyde	ChCl:U (1:2)	Air	0.28	n.d.	$0.14 \cdot 10^{-4}$	<u>Static headspace analysis:</u> Initial concentration: 59 g m^{-3} Temperature: 30°C Volume of absorbent: n.d. Time of equilibrium: 24 h	Thermal desorption at 60°C , 48 h	5	[147]
	ChCl:EG (1:2)	Air	0.28	n.d.	$0.97 \cdot 10^{-4}$			5	
	ChCl:Gly (1:2)	Air	0.28	n.d.	$0.39 \cdot 10^{-4}$			5	
	ChCl:Lev (1:2)	Air	0.28	n.d.	$4.66 \cdot 10^{-4}$			5	
	TBPBr:Gly (1:6)	Air	0.28	n.d.	$0.37 \cdot 10^{-4}$			5	
	TBPBr:Lev (1:1)	Air	0.28	n.d.	$1.65 \cdot 10^{-4}$			5	
	TBABr:DA (1:2)	Air	0.29	n.d.	$4.02 \cdot 10^{-4}$			5	
acetaldehyde	ChCl:U (1:2)	n.d.	n.d.	n.d.	0.06	Molecular dynamics (MD) simulation technique (30°C)	n.d.	n.d.	[220]
	ChCl:Lev (1:2)	n.d.	n.d.	n.d.	0.05		n.d.	n.d.	
propionaldehyde (PA)	G:C:Lev (1:1:3)	N_2	n.d.	1200	0.008	<u>Static headspace analysis:</u> Initial concentration: 50 ppm v/v Temperature: 25°C	Adsorption with activated carbon	5	[132]

	ChCl:U (1:2)	N ₂	n.d.	n.d.	0.01	Volume of absorbent: 1 mL Time of equilibrium: 48 h <u>Dynamic absorption:</u> Initial concentration: 50 ppm v/v Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL	5		
	ChCl:Lev (1:2)	N ₂	n.d.	n.d.	0.011		5		
	ChCl:DEG (1:2)	N ₂	n.d.	n.d.	0.01		5		
MEK Acetaldehyde (AA)	TBABr:DA (1:2)	Air	13.69 4.49	n.d.	0.004 0.0004	<u>Dynamic absorption:</u> Initial concentration: MEK: 3540 g/m ³ ; AA: 1180 g/m ³ Temperature: 30°C Flow rate: 73 µL.hr-1 Pressure: 10 kPa DES mass: 30 g	Thermal desorption at 60°C, 48 hr.	5	[185]
MEK	ChCl:Lev (1:2)	n.d.	n.d.	n.d.	0.0066	<u>Static headspace analysis:</u> Initial concentration: 3000 g/m ³ Temperature: 30 °C Mass of absorbent: 3.4 g Time of equilibrium: 48 h	Thermal desorption at 60°C, 48 h	n.d.	[72]
	ChCl:Lac (1:2)	n.d.	n.d.	n.d.	0.0098			n.d.	
	Chol-C6-Lev	n.d.	n.d.	n.d.	0.0055			n.d.	
	Chol-C8-Lev	n.d.	n.d.	n.d.	0.0051			n.d.	
	Chol-C6-Lac	n.d.	n.d.	n.d.	0.0067			n.d.	
	Chol-C8-Lac	n.d.	n.d.	n.d.	0.0062			n.d.	
acetone	Th:DA (1:1)	N ₂	2.66	60	8.58 kPa	<u>Dynamic absorption:</u> Initial concentration: 1480 ppm v/v Temperature: 30°C Flow rate: 150 mL/min Pressure: 10 kPa DES mass: 3 g	Nitrogen barbotage (flow rate: 150 mL min ⁻¹) at 80°C.	5	[153]
	ChCl:Re (1:2)	N ₂	0.76	40	n.d.			5	
	DDA:DA (1:2)	N ₂	0.49	18	n.d.			5	
VOLATILE METHYL SILOXANES									
L2 L3 L4 D3 D5	C:DA (1:1)	N ₂ :CH ₄ : CO ₂ (66.7:33.3 % v/v)	n.d.	5000	n.d.	<u>Dynamic absorption:</u> Initial concentration: 30 g/m ³ Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL	Nitrogen barbotage (flow rate: 70 mL min ⁻¹) at 150°C for 90 min.	5	[152]
				5500					
	C:UDA (1:1)	N ₂ :CH ₄ : CO ₂ (66.7:33.3 % v/v)	n.d.	500	n.d.			5	
1000				1100					
1100									
C:DDA (1:1)	N ₂ :CH ₄ :	n.d.	400	n.d.	5				



		CO ₂ (66.7:33.3 % v/v)		2000 1000 1000 1000					
L2	G:C:Lev (1:1:3)	N ₂	n.d.	2500	<0.001	<u>Static headspace analysis:</u> Initial concentration: 50 ppm v/v Temperature: 25°C Volume of absorbent: 1 mL Time of equilibrium: 48 h <u>Dynamic absorption:</u> Initial concentration: 50 ppm v/v Temperature: 25°C Flow rate: 50 mL/min Pressure: 10 kPa DES volume: 50 mL	Adsorption with activated carbon	5	[132]
	ChCl:U (1:2)	N ₂	n.d.	n.d.	0.012			5	
	ChCl:Lev (1:2)	N ₂	n.d.	n.d.	0.0085			5	
	ChCl:DEG (1:2)	N ₂	n.d.	n.d.	0.0096			5	
L2 L3 D4	ChCl:U (1:2)	CH ₄ :CO ₂ :H ₂ S (68:31:1 % v/v)	1.42 mol/L 0.99 1.09	1313.4 1245.6 1470.6	n.d.	<u>Dynamic absorption:</u> Initial concentration: 200 ppm v/v Temperature: 20°C Flow rate: 813 m ³ /h Pressure: 100 kPa DES volume: 2.35 m ³	Stripper column, temperature of 115–125 °C and air pressure of 140–170 kPa. inlet air stream (403 m ³ /h)	73	[131]
	ChCl:OA (1:2)	(68:31:1 % v/v)	0.24 mol/L 0.1 0.14	223.8 120.6 187.2	n.d.			60	
L2 L3 D4	TPABr:TEG (1:3)	N ₂ :CH ₄ (2:1% v/v)	380 g/L 230 g/L 5000 g/L	375 280 5300	n.d.	<u>Dynamic absorption:</u> Initial concentration: 50 mg/dm ³ Flow rate: 50 mL/min. Pressure: 10 kPa Temperature: 25°C DES volume: 50 mL	Nitrogen barbotage (flow rate: 50 mL min ⁻¹) at 90°C for 3 h.	10	[149]
D4	OA:DDA (3:1)	N ₂	n.d.	n.d.	0.000297	<u>Static headspace analysis:</u> Initial concentration: 59 mg/g Temperature: 30°C Mass of absorbent: 3.5 g Time of equilibrium: 24 h <u>Dynamic absorption:</u> Initial concentration: 59–531 ppm Flow rate: 15 μL/h Pressure: 10 kPa	Nitrogen barbotage (flow rate: 40 L/h) at 30°C for 5 h.	5	[155]
	NA:DDA (3:1)	N ₂	n.d.	n.d.	0.000292			5	
	DA:DDA (2:1)	N ₂	n.d.	n.d.	0.000314			5	
	Rameb:Lev (7:3)	N ₂	n.d.	n.d.	0.0136			5	
	ChCl:Lev (2:1)	N ₂	n.d.	n.d.	n.d.			5	

						Temperature: 30°C DES mass: 20 g			
VOLATILE FLUORINATED GASES									
R-32 R-125 R-134a	[N ₄₄₄₄][C ₄ F ₉ SO ₃]/C ₄ F ₉ CO ₂ H (1:1)	pure VFA	0.0951 0.1761 0.2659	n.d.	1.111 MPa 1.358 1.169	<u>Stainless steel volumetric system:</u> Temperature: 30°C Pressure: 0.6 MPa DES mass: 0.5 - 0.8 g	n.d.	n.d.	[157]
	[N ₄₄₄₄][C ₄ F ₉ SO ₃]/C ₄ F ₉ CO ₂ H (2:1)	pure VFA	0.0925 0.1861 0.2681	n.d.	1.040 0.916 0.975			n.d.	
	[N ₄₄₄₄][C ₄ F ₉ SO ₃]/C ₄ F ₉ CO ₂ H (1:2)	pure VFA	0.0673 0.2975 0.2567	n.d.	1.576 1.780 1.061			n.d.	
	[C ₂ C ₁ Im][C ₈ F ₁₇ SO ₃]/C ₄ F ₉ CO ₂ H (1:1)	pure VFA	0.0636 0.1449 0.2175	n.d.	1.707 2.177 1.271			n.d.	
	[C ₂ C ₁ Im][C ₈ F ₁₇ SO ₃]/C ₄ F ₉ CO ₂ H (2:1)	pure VFA	0.0637 0.1582 0.1736	n.d.	1.759 1.469 0.881			n.d.	
	[C ₂ C ₁ Im][C ₈ F ₁₇ SO ₃]/C ₄ F ₉ CO ₂ H (1:2)	pure VFA	0.0569 0.1280 0.2084	n.d.	2.4317 2.408 1.474			n.d.	

761 3. DES regeneration

762 The reversible use of absorbents for VOCs capture determines the cost and the sustainability
763 of the industrial processes. The cost of solvent replacement in an absorption column is relatively
764 high. In addition, the used absorbent, which does not ensure high gas purity, must be utilized. Thus,
765 frequent replacement of absorbents significantly increases the cost of the process and adversely
766 affects the environment [131]. Various absorbent regeneration processes are available, i.e., thermal
767 desorption, adsorption using common adsorbents, distillation, or barbotage using inert gas [236].
768 The most commonly used regeneration methods are presented in Figure 3. All DES regeneration
769 processes usually require high temperatures to ensure high VOCs removal efficiency. Therefore, not
770 all well-studied DES can be used as absorbents. Some of them are characterized by low thermal
771 stability, low degradation temperature, and high vapor pressure, which can result in the degradation
772 and loss of DES during the regeneration process [64,237].

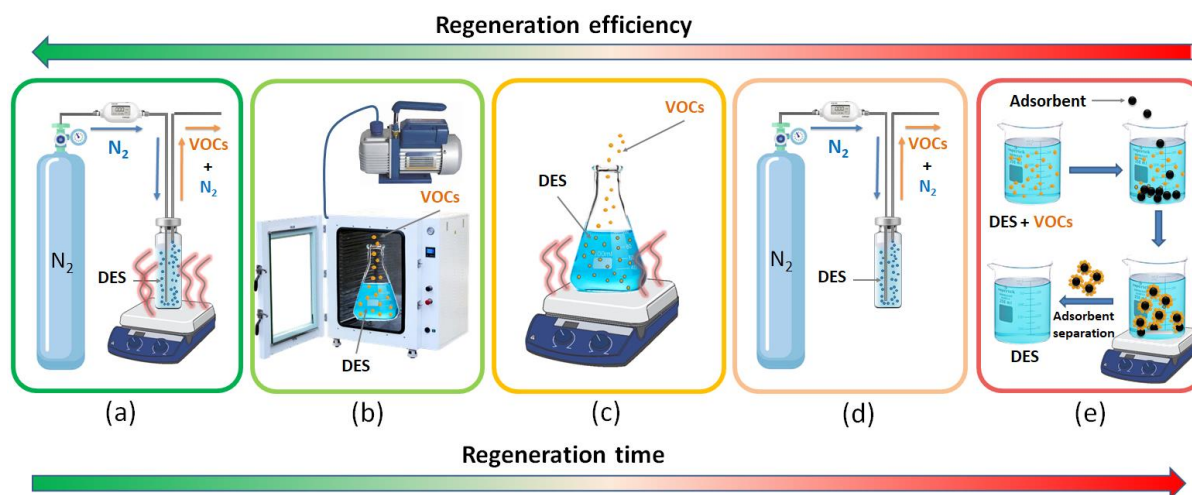
773 Among available regeneration processes, conventional thermal desorption is one of the most
774 common used. It involves continuous stirring of the DES at an elevated 60 °C for 48 hours. As a result
775 of heating the absorbent, the non-covalent bonds formed between DES and VOCs elongate and
776 become weaker. The VOCs then evaporate from the solvent and they are transferred to the gas
777 phase. The regeneration process allows efficient regeneration of absorbents based on quaternary
778 ammonium salts and cyclodextrins, as well as conventional DES. VOCs including toluene, DCM,
779 thiophene, acetaldehyde and butanone can be almost completely removed from DES without
780 noticeable loss of absorption capacity. After five cycles, the absorption capacity remains between 97
781 and 99%. In addition, no change in absorbent mass was observed before and after five absorption-
782 desorption cycles [72,147,148,177]. From an industrial point of view, five cycles is not enough to
783 make the process economical. However, the authors did not specify after how many cycles the
784 decreases in DES uptake efficiency would be observed. Moreover, the absorbent regeneration time is
785 too long to ensure process continuity in a typical industrial system, which consists of one absorption
786 column and one desorption column. Increasing the process temperature is one approach to improve
787 efficiency and reduce DES regeneration time. However, this can lead to the evaporation of DES
788 components, changing the properties of the absorbents and leading to their loss. Therefore, the
789 regeneration temperature should be chosen according to the properties of the absorbent used [238].

790 Another method to ensure a shorter regeneration time for absorbents is the application of
791 nitrogen barbotage. This is the reverse process of conventional absorption. Passing nitrogen through
792 DES containing absorbed VOCs in contact with the gas captures the contaminants and transfers them
793 from the liquid to the gas phase. This process can effectively remove DMDS, toluene, and limonene
794 from ionic and non-ionic DES [155,191]. After five cycles of absorption-desorption, no significant
795 changes are observed in the structure of DES and absorption capacity. In addition, the time of
796 regeneration can be shortened to 5 hours [191]. However, only a combination of the above methods
797 ensures almost 100% regeneration of DES within 1 hour [61,74,149,152,153].

798 The DES regeneration process can also be conducted under reduced pressure (0.05-0.5 kPa)
799 without additional energy input. The application of reduced-pressure regeneration is favorable for
800 unstable DES, which degrade at elevated temperatures. However, high regeneration efficiency can
801 be achieved only for the most volatile VOCs. The evaporation under vacuum efficiency decreases as
802 the boiling point of VOCs increases [239]. Increasing the process temperature reduces the time and
803 increases the efficiency of the VOCs desorption [25,146,188]. Complete DES regeneration can be
804 achieved within one hour at 40 °C [145].

805 Another method for DES regeneration after VOCs absorption is adsorption. The process uses
 806 the equilibrium relationship between the adsorbent, DES, and adsorbate to break the adsorption
 807 equilibrium by changing the temperature or pH value of the solvent [240,241]. To regenerate DES, an
 808 adsorbent with a high affinity for VOCs must be appropriately selected. To date, activated carbon,
 809 silica gel, and aluminum oxide (III) have been tested for VOCs adsorption from used DES [132,191].
 810 The highest DMDS desorption efficiency was demonstrated for silica gel. In addition, it was observed
 811 that as the mass of adsorbent increases, the desorption efficiency of VOCs from DES increases. This is
 812 due to the increased number of active sites in the adsorbent to which DMDS molecules can attach.
 813 However, from an economic point of view, increasing the mass of adsorbent is disadvantageous due
 814 to the cost of purchasing new sorbent and the necessity to regenerate or utilize a higher amount of
 815 used adsorbent. In addition, it has been demonstrated that the desorption efficiency closely depends
 816 on the type of DES. Some DES components can bind strongly to the adsorbent, blocking the active
 817 sites. The adsorption efficiency ranged from 84.5 to 96.6% [191]. This indicates that it is not the most
 818 favorable method for DES regeneration.

819 A back extraction method using common solvents, i.e., water [242], diethyl ether [142], or n-
 820 heptane [243], can also be used to regenerate DES. However, a large amount of waste solvent
 821 remains after the process, which must be disposed of properly. Consequently, the cost of the process
 822 increases. In addition, most of the solvents used are volatile substances that can contaminate the
 823 atmosphere, and the gas purification process is no longer a “green” method. Therefore, this
 824 approach should not be used for the regeneration of DES.



825
 826 **Figure 3** DES regeneration methods a) nitrogen barbotage with heating; b) evaporation under vacuum; c) thermal
 827 regeneration; d) nitrogen barbotage; e) adsorption.

828 4. Critical remarks

829 Currently, published studies demonstrate the great potential of DES application in the industrial
 830 gas separation processes. Nevertheless, many problems and limitations can be observed in the
 831 presented works, which must be solved before using them on an industrial scale. The first major
 832 limitation is the lack of a large amount of data on the physicochemical properties of the developed
 833 DES. Most researchers limit their studies to basic physical parameters' determination, such as
 834 density, viscosity, and MP of DES. Crucial parameters are missed, including polarity, surface tension,
 835 volatility, and degradation temperature. In addition, only toxicity and biodegradability data are

836 available for the most popular DES. Therefore, it is hard to estimate the environmental impact of
837 DES. Many publications assume the green nature of DES, but any studies do not confirm this theory.
838 A systematic analysis of the mentioned parameters is necessary to conduct further studies on an
839 enlarged scale.

840 The cost of some DES is relatively high compared to commercially available absorbents, i.e.,
841 glycols. Therefore, some of them are unsuitable for industrial applications. One possibility to reduce
842 the cost of the absorption process is regeneration. However, some well known DES are characterized
843 by relatively high vapor pressure. Despite their high efficiency of VOCs absorption, it is impractical to
844 use some DES in industrial conditions due to the introduction of additional pollutants into the
845 treated biogas. As a result, the purified gas may contain more contaminants than before the
846 purification process. The vapor pressure of most DES is still unknown. However, based on the DES
847 studied, it can be assumed that a large group of DES can be characterized by high vapor pressure. The
848 regeneration of such absorbents at high temperatures is almost impossible. At elevated
849 temperatures, volatile absorbents are vaporized, resulting in the loss of absorbents and the
850 transmission of DES vapors into the ambient air.

851 The main drawback of the published works is the lack of information on multiple DES
852 regeneration. Most DES were subjected to the regeneration stage only five times. After that, it was
853 proven that DES do not change their structural, physical, and functional properties. However, it is still
854 unclear after how many absorption-desorption cycles, DES significantly reduces its absorption
855 capacity. Without this information, it is impossible to fully estimate the economics of the process. In
856 addition, regeneration processes should reflect real conditions. However, most of the published
857 processes consisted only of heating the used absorbents. Such a process takes a very long time,
858 about 24 hours, and is uneconomical. Therefore, it is not suitable for industrial conditions. Typically,
859 the setup consists of absorption and desorption columns that work simultaneously. Therefore,
860 regeneration of the absorbent should not take longer than the absorption process. A much better
861 solution is to heat the DES and supply a stream of nitrogen, which accelerates the regeneration
862 process.

863 A comparison of DES with conventional and commercially available absorbents indicates that DES
864 can absorb hydrophobic VOCs better or comparably. In contrast, the absorption capacity of
865 hydrophilic contaminants from the O-VOCs group is similar to or worse than water. Therefore, for
866 absorption of hydrophilic VOCs, it is better to consider using water, which is a more "green
867 absorbent." On the other hand, the evaluation of DES efficiency was usually done by examining the
868 partition coefficient of VOCs between the absorbent and the gas phase. However, this is not the best
869 assessment of the new absorbents, as the applicability of the new solvents is affected by many more
870 parameters, i.e., gas flow rate, gas temperature, or composition of the purified stream. These are
871 parameters whose effect on the efficiency of VOCs capture can only be studied using a dynamic
872 absorption system. However, only a few works consider such studies. To date, only a few works have
873 examined the effect of the gas matrix on the absorption efficiency of VOCs. Most of the presented
874 studies were limited solely to the examination of VOC absorption from nitrogen. Due to the inert
875 nature of nitrogen, these studies contribute valuable information about the interaction of VOCs with
876 DES components, but they do not reflect real conditions. The presence of various pollutants, i.e.,
877 carbon dioxide in high concentrations, can rapidly block DES active sites and significantly reduce
878 absorption efficiency. Most of the presented papers describe absorption experiments at room
879 temperature and atmospheric pressure. However, the waste or flue gas temperature is higher (50-80



880 °C). Therefore, more research is needed to fill the huge gap between laboratory studies and
881 industrial processes.

882 In addition, some studies presented the use of mathematical modeling to evaluate the utility of
883 DES in absorption processes. Predictive models, i.e., COSMO-RS, or COSMO-SAC, are perfect for DES
884 preselection because there is no need to introduce real data or to conduct long-term and expensive
885 calculations. However, the main drawback of the models is the large error. Therefore, mathematical
886 modeling should only be complementary to experimental studies.

887 Large-scale (preparative and semipreparative) studies are lacking in published papers. To
888 consider the applicability of DES in an industrial setting, enlarged-scale studies should be conducted
889 using real waste streams. To date, only one paper has considered increasing the scale of the process
890 to 1 L of absorbent. Moreover, barbotage columns were used in all works dedicated to dynamic
891 absorption with DES. Due to their low efficiency, they are not widely used in industry. Therefore,
892 additional research should also be done on the application of other types of absorbers, i.e., spray
893 towers, irrigated packed beds, and plate towers.

894

895 **4. Conclusions and outlook**

896 Currently, a limited number of studies can be found in the literature on the application of DES to
897 VOCs captured from gas streams. The available research is fundamental studies that mainly describe
898 the mechanism of VOCs absorption from the model gas by DES. However, there is still a lack of
899 information on the removal mechanisms of some VOC groups. Therefore, the depth of fundamental
900 and experimental studies (i.e., physicochemical data and molecular interactions between deep
901 eutectic solvent components) are still highly desirable. Published studies demonstrate the great
902 potential of DES as an alternative to commercially available absorbents. This is due to their high VOC
903 absorption efficiency, favorable physicochemical properties, regenerability, and simple and
904 inexpensive synthesis. However, the main advantage in the absorption of VOCs using DES is the
905 ability to tune their properties by selecting HBA and HBD. Therefore, an appropriately designed DES
906 can selectively capture specific contaminants by forming specific non-covalent interactions.

907 Shortly, we can expect to extend the research using more tailored DES to absorb the selected VOC
908 groups. Probably, future research will also be directed to the removal of multiple VOC groups
909 simultaneously from the model as well as real gas streams. There is still a lack of enlarged-scale
910 studies on industrial gas streams or gaseous fuels. However, because of increased scientific interest
911 in this topic, we can soon expect semi-preparative or industrial-scale studies. In addition, the
912 continued development of DES and the appearance of reports of new DES groups in the literature
913 will probably increase similar work using new DES with unique properties for absorbing VOCs. On the
914 other hand, greater awareness among scientists of the negative environmental impact of organic
915 compounds will prompt scientists to better test DES for biodegradability and toxicity.

916

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920 Centre, Poland within the grant project (No. UMO-2021/43/D/ST8/01791).

921 **Abbreviations:**



922 BP - boiling point; BTEX - benzene, ethylbenzene, toluene, xylenes; BTPB -
923 butyltriphenylphosphonium bromide; C - camphor; Car - carvone; Captisol - Sulfobutylether- β -CD;
924 CDs - cyclodextrins; CF - chloroform; ChCl - choline chloride; Chol-C6-Lac - Hexylcholinium lactate;
925 Chol-C6-Lev - Hexylcholinium levulinate; Chol-C8-Lac - Octylcholinium lactate; Chol-C8-Lev -
926 Octylcholinium levulinate; COSMO-RS - conductor like screening model for real solvents; COSMO-
927 SAC - conductor-like screening model segment activity coefficient; CRYSMEB - methylated beta
928 cyclodextrin; D3 - hexamethylcyclopentasiloxane; D4 - octamethylcyclotetrasiloxane; D5 -
929 decamethylcyclopentasiloxane; D6 - dodecamethylcyclohexasiloxane; DA - decanoic acid; DCM -
930 dichloromethane; DDA - dodecanoic acid; DEG - diethylene glycol; DES - deep eutectic solvents; DFT
931 - density functional theory; DMDS - dimethyldisulfide; E - eucalyptol; EC50 - half maximal effective
932 concentration; EEA - European Environment Agency; EG - ethyl glycol; FP - freezing point; G -
933 guaiacol; Glu - glucose; Gly - glycerol; Gu - guaiacol; GWP - global warming potential; HBA -
934 hydrogen bond acceptor; HBD - hydrogen bond donor; HP- β -CD - hydroxypropyl- β -CD; ILs - ionic
935 liquids; K - vapor-liquid partition coefficient; L2 - hexamethyldisiloxane; L3 - octamethyltrisiloxane;
936 L4 - decamethyltetrasiloxane; L5 - dodecamethylpentasiloxane; LA - lactic acid; LD50 - Lethal Dose,
937 50%; Lev - levulinic acid; Lid - lidocaine; MEK - Methyl ethyl ketone; MP - melting point; NA -
938 nonanoic acid; OA - octanoic acid; OECD - Organisation for Economic Cooperation and Development;
939 O-VOCs - Oxygenated volatile organic compounds; Ox - oxalic acid; PC - 1,2-propanediol; Ph -
940 phenol; RAMEB - randomly methylated β -cyclodextrin; QAS - quaternary ammonium salts; QPS -
941 quaternary phosphonium salts; R-32 - difluoromethane; R-125 - pentafluoroethane; R-134a -
942 1,1,1,2-tetrafluoroethane; Res - resorcinol; RT - room temperature; ST - surface tension; SUPRADES
943 - supramolecular deep eutectic solvents; Syr - Syringol; TBABr - tetrabutylammonium bromide;
944 TBACl - tetrabutylammonium chloride; TBPBr - tetrabutylphosphonium bromide; TBPCl -
945 tetrabutylphosphonium chloride; TCE - tetrachloroethane; TCEtOH - 2,2,2-trichloroethanol; TCM -
946 carbon tetrachloride; Tdeg - degradation temperature; TEACl - Tetraethylammonium chloride;
947 TEBACl - Triethyl benzyl ammonium; TEG - tetraethylglycol; Tg - glass transition temperature; Th -
948 thymol; TOABr - tetraoctylammonium bromide; TPABr - tetrabutylphosphonium bromide; TriEG -
949 triethylene glycol; TTABr - tetradecyltrimethylammonium bromide; U - urea; UA - undecanoic acid;
950 US EPA - United States Environmental Protection Agency; WHO - World Health Organization; VFG -
951 Volatile fluorinated gases; VMS - Volatile methyl siloxanes; VOCs - volatile organic compounds;
952 VOSCs - Volatile organosulfur compounds; VOX - Volatile organochlorine compounds; Vp - vapor
953 pressure; Xyl - xylose
954

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