Research Article

Treatment Efficiency by means of a Nonthermal Plasma Combined with Heterogeneous Catalysis of Odoriferous Volatile Organic Compounds Emissions from the Thermal Drying of Landfill Leachates

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The objective of the present work was to assess the odoriferous volatile organic compounds depuration efficiency of an experimental nonthermal plasma coupled to a catalytic system used for odor abatement of real emissions from a leachate thermal drying plant installed in an urban solid waste landfill. VOC screening was performed by means of HRGC-MS analysis of samples taken at the inlet and at the outlet of the nonthermal plasma system. Odor concentration by means of dynamic olfactometry, total organic carbon, mercaptans, NH$_3$, and H$_2$S were also determined in order to assess the performance of the system throughout several days. Three plasma frequencies (100, 150, and 200 Hz) and two catalyst temperatures (150°C and 50°C) were also tested. Under conditions of maximum capacity of the treatment system, the results show VOC depuration efficiencies around 69%, with average depuration efficiencies between 44 and 95% depending on the chemical family of the substance. Compounds belonging to the following families have been detected in the samples: organic acids, alcohols, ketones, aldehydes, pyrazines, and reduced sulphur compounds, among others. Average total organic carbon removal efficiency was 88%, while NH$_3$ and H$_2$S removal efficiencies were 88% and 87%, respectively, and odor concentration abatement was 78%.

1. Introduction

When rain water percolates through the solid waste of a landfill there is a process of dissolution and transport of water soluble elements and organic and inorganic substances, such as heavy metals, ammonium, inorganic anions, and volatile and semivolatile organic compounds, constituting a current that may end up reaching and significatively affecting the aquifers located in the peripheral areas of the facility and, therefore, potentially create risks to the environment and to human and animal health. A detailed summary of the composition of the leachates of urban solid waste (USW) landfills is included in [1], with data from the LEACH 2000 USA database.

Different approaches have been developed for the leachate treatment in order to mitigate the possible impact of USW landfills [2–4]. The methods that are most often implemented (whether alone or in combination) are infill recycling in order to increase the moisture of the waste that is being deposited and combined treatment with domestic sewage, aerobic and/or anaerobic biodegradation, chemical oxidation (including advanced oxidation), adsorption, precipitation, air stripping, microfiltration, ultrafiltration, nanofiltration, and reverse osmosis.

As an alternative to the mentioned techniques, in some cases thermal drying has also been used. It is based on an atomization process, during which ≅50 μm droplets are formed, which are subjected to the action of a hot air current
which causes the evaporation of the aqueous phase of the leachate from the surface of the droplets, thereby turning them into dry particles that, after a cooldown stage, are separated with a cyclone and a bag filter in series. As a residual effluent, a current of humid air with a “roasted” smell is produced. It has a high content of gaseous compounds withodoriferous significance, such as ammonia, amines, pyrazines, hydrogen sulfide, mercaptans, thioethers, carbonyls, alcohols, esters, and terpenes.

Preliminary studies concerning the characterization of the thermal drying process emissions of 3 urban solid waste (USW) leachate types allowed the determination of emission levels, for several compounds with odoriferous significance, H$_2$S (0.5–24 mg/Nm$^3$), NH$_3$ (5–277 mg/Nm$^3$), $\Sigma$ Alcohols C$_2$–C$_6$ (35–335 mg/Nm$^3$), and $\Sigma$ Carboxyls C$_3$–C$_6$ (60–345 µg/Nm$^3$), and emission levels of $\Sigma$ Pyrazines, which would explain the “roasted” odor descriptor assigned to them. Chemical reactions involving free radicals typically in turn react with the effluent pollutants and decompose the formation of free radicals from parent molecules, which and 250,000K, and, under these conditions, they induce an electric field at typical temperatures between 10,000 and $10^5$ s, which can translate to highly compact systems. Normal pollutant concentrations are around the 100 s of ppm, and therefore the direct interaction between the electrons and the pollutant molecules is negligible. The NTP induces chemical reactions at moderate conditions of approximately 1 atm and room temperature. The addition of oxidant substances is not required because the NTPs use the oxygen and water vapor to generate the reactive radicals.

In order to implement applications with significant yields, systems have been developed which combine the NTP with the use of adsorbents (such as activated carbon), different catalysts, and also supplementary UV postoxidation. The combination of a nonthermal plasma with catalytic processes (NTP-CAT) can be carried out by (a) in-plasma catalysis (IPC), where the selected catalyst is put directly inside the discharge chamber or by (b) postplasma catalysis (PPC), placing the catalyst after the discharge chamber. A heterogeneous catalyst is used, including Al$_2$O$_3$, SiO$_2$, TiO$_2$, MnO$_2$, platinum-based catalysts, and modified zeolites, and it can be introduced in the reactor in different forms. The details of the theoretical and practical aspects of NTP and NTP-CAT have been abundantly discussed in the available bibliography.

Table I includes a summary of the publications cited in the review article by Van Durme et al. [14] on the maximum abatement efficiencies for several VOC treated by means of coupled dielectric barrier discharge (DBD) nonthermal plasmas, both IPC and PPC, with different catalysts (DBD-NTP-CAT). It must be taken into account that most of the data in Table I and other publications cited in this work comes from laboratory tests treating currents with a single target pollutant [14–23].

In the present work a DBD-NTP-CAT pilot plant was installed in the Palautordera landfill (Barcelona, Spain), where the urban solid waste leachate is treated by means of thermal drying. The gas effluents emitted from the drying process are treated by means of an RTO system that uses biogas as fuel. A diagram of the installation is shown in Figure 1, including the two configurations that were tested (indicated as Options A and B, resp.). As will be explained in the following section, it must be noted that the quantitative efficiency tests were carried out with the Option B configuration.

### 2. Plasma Operation Conditions

Preliminary tests were carried out with the configuration indicated as Option A in Figure 1, that is, conducting the emission (diluted with clean air) directly into the plasma chamber, from where it was transferred to the chamber containing the catalyst. With this mode of operation it was found that, when the gas entered into contact with the electrodes, abundant small water droplets would condense that, because of the high operation electrical potentials, generated sparks which damaged the electrodes and made it impossible to carry out the corresponding efficacy tests. It was decided, therefore, that only ambient air would enter the plasma chamber. Then, the ionized ambient air was led into a mixing chamber together with the emission air as...
Table 1: VOC abatement efficiencies with DBD-NTP-CAT tests.

| Compounds  | Catalyst                  | Maximum abatement efficiency % | Publications cited by Van Durme et al. (2008) in [14]* |
|------------|---------------------------|---------------------------------|------------------------------------------------------|
| Benzen     | TiO₂                       | 60                              | Chae et al., 2004 (28)                                |
|            | TiO₂/SiO₂                  | 50                              |                                                      |
|            | TiO₂                       | 16                              | Lu et al., 2006 (4)                                  |
|            | Ag/TiO₂                    | 96                              | Kim et al., 2005 (12)                                |
| Toluene    | MnO₂/Al₂O₃                 | 55                              | Delagrange et al., 2006 (26)                         |
|            | CuO/MnO₂                   | 50                              | Chae et al., 2004 (28)                                |
|            | TiO₂                       | 16                              | Lu et al., 2006 (4)                                  |
|            | MnOₓ/Coₓ                    | >99                             | Subrahmanyan et al., 2006 (2)                        |
| SF₆        |                            | >99                             |                                                     |
| NF₃        | CuO/ZnO/MgO/Al₂O₃          | >99                             | Chang et al., 2004 (29)                              |
| CF₃        |                            | 66                              |                                                     |
| C₂F₆       |                            | 83                              |                                                     |
| HCOH       | Ag/CeO₂                    | 92                              | Ding et al., 2006 (30)                               |
| Trichloroethene (TCE) | Au-SBA-15           | >99                             | Magureanu et al., 2007 (34)                         |
| TCE        | MnO₂                       | 97                              | Subrahmanyan et al., 2007 (2)                        |
| Isoopropanol | MnOₓ/Coₓ                  | >99                             | Han et al., 2007 (35)                                |
| Dichloromethane | Various              | 34–51                           | Intriglio et al., 2007 (36)                         |

*The bracketed italic numbers correspond to the reference numbers in Van Durme et al. (2008) [14], and not the references of this paper.

Figure 1: Diagram of flow process of the NTP-CAT pilot plant configurations tested.

The conditions which were foreseen to provide the highest depuration efficiencies (200 Hz plasma frequency, 1:5 (v:v) dilution ratio, and 150°C catalyst temperature). The fourth campaign (Day 4 in Table 2) intended to provide data on the performance of the system under conditions that minimized the electrical cost associated to the heating of the catalyst (3:5 (v:v) dilution ratio and 50°C catalyst temperature). This fourth campaign was also meant to assess the effect of a decrease of the plasma frequency.

3. Materials and Methods for the Determination of the Treatment Efficiency of the NTP-CAT Pilot Plant

3.1. Sampling and “In Situ” Measurements. The samples were taken at the sampling points 1 and 2, as shown in Figure 1. Sampling point 1 was located at the inlet of the system and corresponds to the process gas before the dilution with fresh air, while sampling point 2 was located at the outlet of the system after the treatment. As the residence time of the gas in the system was very short, the inlet and outlet samples were taken simultaneously.

On-site determinations of hydrogen sulphide (H₂S), mercaptans (RSH), and ammonia (NH₃) were carried out by means of “ad hoc” colorimetric tubes (Dräger) by pumping the required sample volumes through the tubes. Limits of detection were 0.5 ppmV for RSH, 0.2 ppmV for H₂S, and 2 ppmV for NH₃. In order to determine the regularity of the gaseous effluent to be treated and the final emission, the total organic carbon (TOC) was also measured “in situ” with a portable flame ionization detector (FID) analyzer (Thermo FID, Mess-Analysentechnik GmbH).
Table 2: Odour concentration, VOC, NH₃, H₂S, and TOC concentrations, and % reduction in each day.

| Parameter                        | Day 1     | Day 2     | Day 3     | Average (Days 1–3) | Day 4.1 | Day 4.2 | Day 4.3 | Day 4.4 |
|----------------------------------|-----------|-----------|-----------|--------------------|---------|---------|---------|---------|
| Odour conc. (dynamic olfactom.)   |           |           |           |                    |         |         |         |         |
| Inlet conc. (ouE/m³)             | 13,777    | 38,968    | 20,632    | 24,459             |         |         |         |         |
| Outlet conc. (ouE/m³)            | 5,161     | 2,580     | 4,812     | 4,184              |         |         |         |         |
| Reduction (%)                    | 63        | 93        | 77        | 78                 |         |         |         |         |
| ΣVOCOD (μg/m³)                   | 148.4     | 470.6     | 195.9     | 271.6              | 98.3    | 92.6    | 41.2    | 42.5    |
| Inlet conc. (μg/m³)              | 51.8      | 145.8     | 55.2      | 84.3               | 45.8    | 19.3    |         |         |
| Outlet conc. (μg/m³)             | 18.1      | 19.2      | 13        | 16.8               | 81      | 65      |         |         |
| Reduction (%)                    | 65        | 69        | 72        | 69                 | 53      | 53      |         |         |
| NH₃                              |           |           |           |                    |         |         |         |         |
| Inlet conc. (ppmV)               | 139.3     | 143.0     | 115       | 132.4              | 265.2   | 265.2   | 170.5   | 170.5   |
| Outlet conc. (ppmV)              | 18.1      | 19.2      | 13        | 16.8               | 81      | 65      |         |         |
| Reduction (%)                    | 87        | 87        | 89        | 87                 | 53      | 53      |         |         |
| H₂S                              |           |           |           |                    |         |         |         |         |
| Inlet conc. (ppmV)               | 3.7       | 4.0       | 2.0       | 3.2                | <0.5    | <0.5    | <0.5    | <0.5    |
| Outlet conc. (ppmV)              | <0.5      | <0.5      | <0.5      | <0.5               | <0.5    | <0.5    | <0.5    | <0.5    |
| Reduction (%)                    | >86       | >88       | >75       | >83                | n.a.*   | n.a.*   | n.a.*   | n.a.*   |
| TOC                              |           |           |           |                    |         |         |         |         |
| Inlet conc. (mgC/m³)             | 62.3      | 46.5      | 51.5      | 53.4               | 167.3   | 150.1   | 140     | 140     |
| Outlet conc. (mgC/m³)            | 10.5      | 5.7       | 3.5       | 6.6                | 72.3    | 129.3   | 102.5   | 120.8   |
| Reduction (%)                    | 83        | 88        | 93        | 88                 | 57      | 14      | 27      | 14      |
| Plasma frequency (Hz)             | 200       | 200       | 200       | —                  | 200     | 150     | 200     | 100     |
| Effluent : dilution air ratio (v : v) | 1:5     |           |           |                    | 3:5     |         |         |         |
| Catalyst (temperature)           | MnO₂ (150°C) | MnO₂ (50°C) | | | |

* n.a.: not applicable; the compound was not detected at the inlet or at the outlet.

Note: the results indicated as < (value) correspond to the LOD of the corresponding technique.

Two fractions were considered regarding the VOC studied. The first fraction includes the VOC with a molecular size between 2 and 6 carbon atoms (C₂–C₆), which was sampled by pumping 15 L of gas through an activated charcoal cartridge (ORBO, 250 mg). The fraction containing the VOC with more than 6 carbon atoms (C₆ > C₆) was sampled by filling 5 L Nalophan bags by means of indirect aspiration with a lung-like device. All the samples were transported to the laboratory and analyzed as quickly as possible.

The 10 L samples contained in Nalophan bags for dynamic olfactometry analysis were taken using the same procedure described in the preceding paragraph for the VOC > C₆ and were sent immediately to the laboratory in a refrigerated and temperature-controlled container so they could be analyzed within 30 h, as indicated in the norm EN-13725:2005 [24].

3.2. Sample Treatment and Analysis

3.2.1. VOC Screening. HRGC-MS analyses were performed by means of a ThermoTrace HRGC-MS system with a 30 m × 0.25 mm ID × 1.4 μm BPX-624 column (SGE). Chromatographic conditions for both the analysis of the activated charcoal and the samples contained in Nalophan bags were carrier gas He at 1.5 mL/min, injection temperature 245°C, temperature program 35°C (5 min), and 5°C/min up to 250°C, staying at 250°C for 10 minutes. MS acquisition was performed in Full-Scan mode (m/z range 35 to 350 a.m.u.). Data treatment and identification of the analytes were carried out using the Xcalibur software (Thermo Fisher) with NIST and Wiley spectra libraries. The identification of an analyte was only considered positive if there was at least 85% matching between its spectrum and the library one.

Each of the VOC fractions was analyzed with the following conditions.

(1) Activated Charcoal Samples. The charcoal where the VOC were adsorbed was transferred to a vial and desorbed with 1,2,3-trimethylbenzene (Sigma Aldrich, p.a.) containing Toluene-D₈ as internal standard. The quantitation limit was 0.1 μg/m³.

(2) Samples Contained in Nalophan Bags. First, the samples are brought to the laboratory, where a Toluene-D₈ internal standard (Supelco) is added to a concentration of 25 μg/m³. Then a PDMS fiber (Supelco) was exposed to the sample for 60 minutes in order for the analytes to be adsorbed onto the fiber, after which the fiber was inserted into the HRGC system's injector at 245°C for the desorption of the adsorbed VOCs to take place. The quantitation limit was 0.1 μg/m³.

(3) Quantitation. The quantitation of the analytes present in both the activated charcoal and the Nalophan bag samples was carried out by using standards of some 100 compounds belonging to the VOC families that are commonly found
Table 3: Inlet and outlet concentration ranges and average concentration reduction for each compound family.

| Family                        | Inlet conc. range ($\mu g/m^3$) | Outlet conc. range ($\mu g/m^3$) | Average conc. reduction (%) |
|-------------------------------|----------------------------------|----------------------------------|----------------------------|
| $\Sigma$ Organic acids*       | 3.7–29.5                         | 0.2–3.5                          | 67.7                       |
| $\Sigma$ Alcohols and ethers  | 32.7–227.7                       | 6.9–33.3                         | 68.0                       |
| $\Sigma$ Aldehydes            | 2.1–232.8                        | 0.5–5.5                          | 65.3                       |
| $\Sigma$ Ketones              | 20.5–143.5                       | 2.7–34.6                         | 69.0                       |
| $\Sigma$ Esters               | 0.4–2.1                          | 0.1–1.0                          | 44.7                       |
| $\Sigma$ Pyrazines and pyridines | 2.2–54.3                    | 0.2–24.1                         | 54.2                       |
| $\Sigma$ Sulphur compounds    | 1.3–5.5                          | n.d. <0.1                        | 95.3                       |
| Other                         | 1.0–10.6                         | 0.2–2.2                          | 64.5                       |

* Excluding acetic acid, that had higher outlet than inlet levels, possibly due to new formation.

in environmental samples (including alcohols, esters, carboxyls, carboxylic acids, aromatic compounds, terpenes, and thioethers). These standards were used for the calculation of calibration curves for the corresponding compounds and relative response factors between each compound and Toluene-D8. Quantitation was then carried out by relative response to the Toluene-D8 internal standard, using relative response factors for most analytes. As it was impossible to have standards for all of the compounds that are typically present in this kind of effluents (and it is also impossible to know beforehand which ones will be present), the relative response factors of similar compounds were used when analytes were detected for which no standard was available.

3.2.2. Odor Concentration Analyses. Odor concentration was determined in an accredited lab by means of Dynamic Olfactometry according to the requirements indicated in the norm EN-13725:2005 [24]. Details of the applied techniques can be found in [25–35].

3.3. Results. A summary of the results of the VOC analyses, as well as the other studied parameters, is included in this section.

The VOC results shown in Tables 2 and 3 do not include all of the compounds of this group that were detected, and only the compounds with low enough odour thresholds to present significant odor implications, according to our experience [36], have been included in the study. These relevant odorous VOC (VOC$_{OD}$) belong for the most part to the following families: pyrazines and pyridines, organic acids, alcohols and ethers, aldehydes, ketones, esters, and reduced sulphur compounds (mercaptans and thioethers). Other nonodorous compounds were detected in the samples, such as aliphatic and aromatic hydrocarbons, but have not been taken into account, as they have higher odour thresholds [37] and do not significantly contribute to an emission’s odor impact [5, 27].

The experimental work was carried out during four days. During the first three days, the NTP system worked all the time with a plasma frequency of 200 Hz, and all of the indicated parameters were analyzed. During the fourth day, three frequencies were tested (100, 150, and 200 Hz, resp.). It must be pointed out that speciated VOC$_{OD}$ analyses were only performed at 200 Hz, while TOC, NH$_3$, H$_2$S, and mercaptans were determined at each of the three studied frequencies.

Table 2 shows the VOC$_{OD}$ concentration reduction, which corresponds to the average of all the individual VOC$_{OD}$ reductions. Also shown are the plasma frequencies corresponding to each assay, as well as the % reductions of TOC, NH$_3$, and H$_2$S in each day. The concentrations of mercaptans determined "in situ" were always under the detection limit (0.5 ppmV) in all of the samples analyzed during this study and therefore were not included in the table, but it must be pointed out that mercaptans were not detected in any of the HRGC-MS analyses of the charcoal cartridges, with LOD around 2 orders of magnitude smaller. Finally, a summary of the plasma operation conditions is also included in the table (catalyst type, temperature, and effluent/clean air dilution rates).

Table 3 shows the VOC$_{OD}$ families present in the samples, along with the mean reduction efficacy for each kind of compound. Generally, the most relevant families in terms of concentrations were alcohols, aldehydes, ketones, and organic acids, and there was also a highly significant presence of pyrazines, which are usually formed in thermal drying processes such as the one carried out in the plant where the NTP system was tested and which have a strong roastedlike odor [5]. Their inlet concentrations were between 0.1 and 40.1 $\mu g/m^3$. It must be pointed out that, for certain compounds, their concentration in the outlet samples may sometimes be higher than in the inlet or even newly appear and therefore individually have negative % reductions.

Figure 2 shows two speciated VOC$_{OD}$ ($C_n > C_8$) TIC chromatograms corresponding to the SPME analyses carried out at the inlet and the outlet of the NTP system samples taken during Day 3.

As an example, Table 4 shows the speciated VOC$_{OD}$ ($C_n > C_8$) screening results corresponding to Day 4.1, along with the % reduction for each compound.

4. Discussion and Conclusions

As shown in Table 2, the NH$_3$ depuration efficiency, working in the conditions of maximum capacity of the treatment...
system, was quite good, ≥87%, which is similar to those reported in [10] of 93.3%, in [38] of 80%, and in [36] of ≥100%. The results shown in [39], however, are significantly different, with a reported NH$_3$ depuration efficiency of 44%. The average H$_2$S reduction that was determined during this work is >83%, which is comparable to the results found in [8, 39], which are 96.3% and 91%, respectively.

As can be seen in Table 3, the % reductions for the VOC$_{OD}$ concentrations were in the 45–95% range, while Table 2 shows that the average total % reductions are between 53% and 75%. These depuration efficiencies may prove to be somewhat low when dealing with critical emissions or those with high odor loads. These values are close to those reported in [40], where depuration efficiencies for thiocethers were 78–100% and 32–51% for other VOC and with those in [39] which indicate abatement efficiencies in the 45–93% range for oxygenated VOC such as aldehydes, ketones, and alcohols. The average total organic carbon content reduction of 88% shows good agreement with the determined VOC reduction efficiencies achieved during this study, although it must be pointed out that these results are significantly different from the 35% TOC reduction reported in [39].

Regarding the odor concentration abatement, the results published in [40] (70–90%) and [39] (72–88%) are similar to our results (78%). Finally, the dynamic olfactometry and $\Sigma$VOC$_{OD}$ data shows that the reductions are quantitatively comparable (with reduction values of 78% and 89%, resp.).

The results obtained in different plasma conditions during Day 4 show that a lower catalyst temperature, together with a higher effluent: dilution air ratio, translates to a small decrease in the NH$_3$ abatement efficacy, as well as a more significant decrease in the VOC$_{OD}$ and TOC reductions. Furthermore, the experiment carried out during Day 4 shows that lower plasma frequencies correspond to a clear decrease in the TOC reduction efficacy.

As has been previously mentioned, the most important VOC$_{OD}$ families detected in the samples, in terms of concentration, were alcohols, aldehydes, organic acids, and pyrazines. The highest depuration efficiencies were those corresponding to sulphur compounds and ketones (95% and 69% on average, resp.), while esters and pyrazines (the latter compounds have an olfactive descriptor coincident with that of the studied emission) showed the poorest removals with the NTP-CAT system studied during the present work, with values just near 50% in the experimental conditions that were used. In this regard, it must be pointed out that these reduction percentages show good agreement with those published in different references (e.g., [38, 40]).

On the other hand, even though it falls outside the scope of the present work, an aspect that must be highlighted is the fact that the levels of depuration efficiencies of odoriferous compounds do not correspond with the experimentally observed odor concentration reductions determined in the laboratory by means of dynamic olfactometry (carried out according to the norm EN-13725:2003) as, while a clear reduction of odor intensity between the samples taken before and after the NTP-CAT system could be organoleptically perceived, this was not clearly reflected by the results of the olfactometric assays. The causes that may explain these not fully satisfactory odor depuration efficiencies may be attributed, among others, to a possible incomplete degradation of some VOC$_{OD}$ and that some of the resulting by-products may have lower olfactive thresholds (OTV) than their precursors (e.g., [39, 41]), which may be due to the selected operation configuration and/or the catalyst that was chosen for this pilot test. It must be pointed out that, in some cases, a new formation of oxidized species (such as acetic acid or some carbonyls) was observed, but, due to the limited number of samples that were analyzed, it was not possible to arrive at definitive conclusions, and these compounds were excluded from the VOC$_{OD}$ lists.

It can be concluded, therefore, that the NTP-CAT technology that was tested, in its current state and under the applied conditions, will likely not be capable of providing sufficient performance in critical deodorization operations (e.g., high odor or VOC$_{OD}$ loads, very close populated receptors, etc.) which require very stringent demands regarding odor.

**Figure 2:** TIC chromatograms of an inlet (a) and outlet (b) sample.
Table 4: Odoriferous VOC$_{OD}$ ($C_n > C_6$) screening results and % reduction, corresponding to Day 4.1.

| Compound              | Inlet conc. ($\mu g/m^3$) | Outlet conc. ($\mu g/m^3$) | % Reduction |
|-----------------------|-----------------------------|----------------------------|-------------|
| 2-Hexanone            | 0.2                         | <0.1                       | >50.0       |
| 2-Hexen-1-ol acetate  | 0.4                         | 0.4                        | 0.0         |
| 2-Hexen-1-ol          | 1.7                         | 1.2                        | 29.4        |
| Dimethylpyridine      | 0.6                         | 0.8                        | -33.3       |
| Cyclohexanone         | 1.3                         | <0.1                       | >92.3       |
| 2-Ethylpyridine       | 0.1                         | <0.1                       | 0.0         |
| 2,6-Dimethylpyrazine  | 0.9                         | 0.6                        | 33.3        |
| 2,3-Dimethylpyrazine  | 0.9                         | 0.6                        | 33.3        |
| 2-Ethyl-2-hexenal     | 0.2                         | 0.2                        | 0.0         |
| Benzaldehyde          | 0.2                         | <0.1                       | >50.0       |
| 2-Ethylhexanal        | 0.5                         | <0.1                       | >80.0       |
| Hexanoic acid         | 0.3                         | 0.2                        | 33.3        |
| Terpene               | 0.3                         | 0.2                        | 33.3        |
| 2,4,5-Trimethylpyridine| 0.3                        | <0.1                       | >66.7       |
| 2-Methyl-6-ethylpyrazine| 0.4                  | 0.2                        | 50.0        |
| Trimethylpyrazine      | 13.7                       | 8.9                        | 35.0        |
| Limonene              | 0.8                         | <0.1                       | >87.5       |
| Eucalyptol            | 1.3                         | 0.8                        | 38.5        |
| Dihydroisophorone     | 0.6                         | <0.1                       | >83.3       |
| 2-Ethyl-3,5-dimethylpyrazine| 4.9                  | 3                           | 38.8        |
| 3-Ethyl-2,5-dimethylpyrazine| 19.5             | 9.8                        | 49.7        |
| Alcohol               | 2.4                         | 1.9                        | 20.8        |
| 2-Ethylhexanoic acid  | 3.5                         | <0.1                       | >97.1       |
| Camphor               | 2.7                         | 1.7                        | 37.0        |
| Octanoic acid         | 3.5                         | 3.4                        | 2.9         |
| 3-Undecanone          | 6.9                         | 4.8                        | 30.4        |
| Possible sulphur derivative | 5.5                    | <0.1                       | >98.2       |
| 2-Methyl-1-dodecanol  | 4.8                         | 3.1                        | 35.4        |
| Alcohol               | 14.3                       | 8.7                        | 39.2        |

$\Sigma$VOC$_{OD}$ ($C_n > C_6$) 92.7 51.5 44.4

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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