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Exploring Odor Minimization in Post-Consumer Plastic Packaging Waste through the Use of Probiotic Bacteria

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Abstract: Plastic packaging represents a large proportion of the plastic consumption throughout the world. The negative environmental impact associated with plastic packaging waste can be in part abated by recycling plastics, and increasing numbers of regulatory frameworks are being adopted towards this goal. Despite recent advances in modern recycling technologies, the production of high-quality polyolefin recyclates remains a challenge. Among other functional requirements, odor plays a crucial role in the acceptance of recycled packaging. This presents a challenge, as odor contamination in plastic packaging waste can stem from diverse sources, such as spoilage processes, and strongly depends on the quality of the post-consumer input material. The present study addressed this issue by exploring potential odor abatement of malodors in packaging waste through the use of probiotic bacteria. Specifically, probiotics were added to a mixed post-consumer plastic packaging waste fraction, which was subsequently evaluated using human sensory and gas chromatography–olfactometric analyses. A comparison of treated with untreated plastic waste fractions revealed significant sensory differences. Further structural elucidation of the causative odorants confirmed a reduction in malodorous microbial metabolites, although complete odor removal was not achieved. However, this environmentally friendly approach may represent an essential step towards overcoming the odor burden in post-consumer plastic packaging recyclates.

Keywords: decontamination; deodorization; gas chromatography–olfactometry; mixed post-consumer plastic packaging waste; polyolefin; probiotics; recycling; smell; washing process

1. Introduction

Plastic is the number one material for packaging, as is evident from the vast amount of plastic packaging material produced globally every year, estimated at 78 million tons [1]. Despite these quantities, only a fraction is recycled, with 10% of the global plastic packaging flow currently part of a closed-loop (2%) or cascaded recycling (8%), while the majority enters landfill or is leaked into the environment [1]. To cope with growing quantities of plastic waste and to conform with increasingly stringent legal regulations for recycling quotas [2], a functional plastics recycling chain that yields high-quality recycled plastic material is more important than ever [3]. In relation to polyolefins, insufficient quality and, in particular, odor-related problems are still major impediments for the widespread re-use of recycled plastic materials [4]. The unwanted smell of recyclates can be traced back to the high contamination of input material, primarily from post-consumer plastic waste [5]. Although methods that aim at odor reduction exist, for instance by physical deodorization or advanced

washing processes, including the application of hot water or chemical additives, a complete odor removal has hitherto not been achieved [6–13]. Moreover, a high-energy input or the addition of potentially environmentally critical chemicals, such as detergents, absorbers or entrainers, are often still required.

Ecologically friendly alternatives are in rising demand in many industrial fields. Probiotics, as valuable non-pathogenic bacterial cultures, are increasingly applied in many sectors, with the fields of human health and food quality representing the most traditional and best established areas [14,15]. The benefits of probiotics are manifold, including their potential ability to displace other microorganisms that may be harmful. This is achieved not only by the competition for nutrients and space, but also by their potential excretion of enzymes and antibiotic compounds that keep pathogens at bay [16]. More recently, the implementation of probiotics in livestock farming has been increasingly explored, especially in aquaculture [17–20]. A successful reduction of pathogens was even reported in hospital hygiene control using surface cleaning agents containing spores of diverse *Bacillus* spores [21]. Many probiotic strains additionally exhibit a potential to reduce organic matter via exo-enzymatic metabolism. Such reactions have been reported to support the degradation of biofilms, as well as the conversion of odorous contaminants into non-odorous volatiles, yielding innovative and promising areas of application in wastewater or industrial cooling waters [22–26]. For instance, a substantial reduction of a sulfurous, rotten egg-like smell was achieved by applying different bacteria of the genus *Bacillus* to industrial wastewater [27].

With regard to the smell associated with recycled plastics, the elimination or reduction of odor-active contaminants, or even avoidance of their formation at an early stage, is of prime importance. Recently, it was demonstrated that despite an observed decrease in odorous volatiles in mixed plastic film waste through washing, most of these components were still present after extrusion [28]. In terms of the extended storage periods of post-consumer plastic packaging waste, commonly up to three to four weeks in households, with several additional weeks until final processing, an early avoidance of spoilage processes presents a promising approach to reduce generation of odorous compounds from rotting processes of organic matter in plastic packaging waste [5].

In view of these challenges, the present study aimed not only at a molecular characterization of the causative odorous contaminants, but also at a reduction of odorous microbial metabolites in post-consumer plastic packaging waste by the application of a probiotic bacterial solution during the recycling process. Our focus was on the film fraction, primarily lightweight packaging, which has been previously reported to be especially susceptible to smell contamination [29]. To this aim, the efficiency of odor removal or even avoidance of odor generation in post-consumer plastic packaging waste using a commercial probiotics formulation was investigated on a pilot scale. Molecular characterization of the causative odorants in the probiotics-treated sample in comparison to an untreated reference sample was carried out using a combination of human sensory evaluations together with targeted high-resolution gas chromatography–mass spectrometry analyses.

2. Materials and Methods

2.1. Sample Material

2.1.1. Post-Consumer Plastic Packaging Waste Fraction

Post-consumer plastic packaging waste was sourced from household waste collected through a local council collection system known as 'yellow bag' collection in the state of Bavaria, Germany. The examined waste comprised the fraction no. 352 'lightweight packaging (LWP): mixed plastics' [30], that was sorted by means of state-of-the-art sorting technology (sorting date: 24 July 2019, Bavaria, Germany) [29] and chosen because of the typically intense off-odor associated with this post-consumer packaging waste. This fraction consisted mainly of plastic films that had been shredded post-sorting to 50 mm particles for storage. The waste contained 55% polyolefin and had a dry matter content of 93% (w/w).

2.1.2. Probiotic Bacterial Solution

A commercial bacterial suspension, 'PIP Aquatec Basic' (*Chrisal*), was applied, which is sold as a strong probiotic concentrate for the treatment and maintenance of large industrial water systems, such as cooling water and process water [31,32]. According to the manufacturer, the probiotic bacteria metabolize organic dirt and thus prevent water cloudiness, odor and biofilm formation. Further, it is claimed that the bacteria potentially displace microorganisms in water systems that produce odorous volatile gases while releasing odorless volatile compounds only. These positive effects are purportedly achieved by several different probiotic species (including different *Bacillus* spores) with 50 million germs per mL. For the present study, and according to the manufacturer's dosage, 2 L/m^3 was used for the treatment of the waste, which was diluted with water at a ratio of 1:10 for use as a spray solution. Appropriate adaption and growth of the probiotic bacteria on the input waste material has been ensured in microbiological pretests on a laboratory scale. Since the time of application was found to play an important role, probiotics were applied directly after sorting and shredding of the input waste material (Section 2.2.).

2.2. Pretreatment With Probiotic Bacteria and Washing Process of the Separated Post-Consumer Packaging Waste

Two 250 kg batches of post-consumer plastic packaging waste (fraction no. 352, cf. Section 2.1.) were sourced from a local sorting plant (date: 24 July 2019, Bavaria, Germany). Each batch was collected in a separate big bag (1 m^3). The first batch (250 kg) was sprayed with 20 L of the diluted probiotic bacterial solution; the second batch remained untreated and served as a reference. In order to ensure sufficient wetting of the input material and that the bacterial solution adhered evenly to the waste, 50 kg aliquots were placed on a conveyor belt and sprayed with 5 L of the bacterial solution. Afterwards, the bag was turned several times. This spraying and mixing procedure was repeated four times in total. A standard pressure spray bottle (Primex 5 Comfort, Gloria, Witten, Germany) fitted with a spray lance and nozzle with a barrel volume of 5 L was used to apply the bacterial solution. Both batches were subsequently stored outside for 40 days (24 July 2019–2 September 2019 Germany) until further processing.

A subsequent washing process after 40 days storage was carried out at a technical center (Herbold Meckesheim GmbH, Meckesheim, Germany). To eliminate any carry-over effects from water circulation (120 m^3), the circulated washing water was replaced with fresh water between each washing step of the individual samples. Both batches (sprayed waste and untreated reference waste) were cleaned with cold water in a two-stage washing process (Figure 1). After the initial removal of coarse impurities in the pre-washing stage, a friction washer performed the main washing. The residence time of each waste input material within the wet processing was approximately 10 min. Subsequently, the sample material was first mechanically dried in a centrifuged dryer followed by thermal drying with a hot air flow of approximately 80 °C (T1015 Mechanical Dryer and TNT 500 Thermal Dryer, both Herbold Meckesheim GmbH, Meckesheim, Germany). Afterwards, aliquots of both waste samples were stored at −20 °C until sensory analyses to prevent any further bacterial activity.

2.3. Sensory Evaluation

2.3.1. Panel

Sensory analyses were made by a trained panel comprising 14 panelists (age 24–57 years) from the Department of Sensory Analytics of Fraunhofer IVV (Freising, Germany). Panelists did not exhibit any known illness or olfactory dysfunction at the time of the assessments. Their olfactory capabilities were tested in weekly sensory training sessions comprising the recognition and the description of about 150 odorants presented in the form of odorant pens.

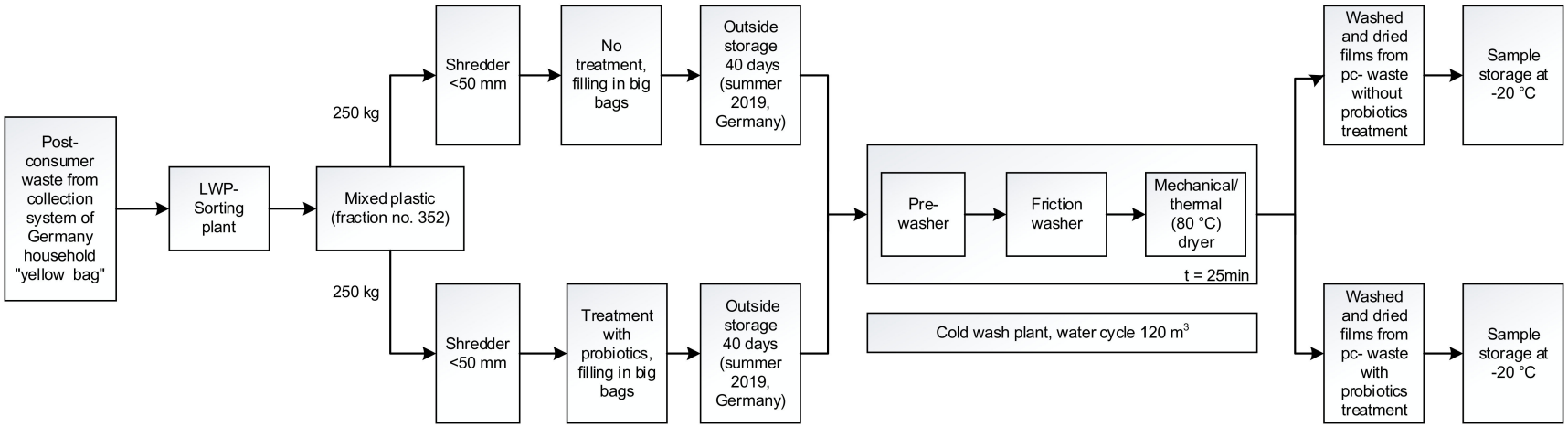

Figure 1. Schematic of sample collection of the post-consumer (pc-) plastic packaging waste and treatment with probiotics, including storage conditions and the washing process in comparison to the respective reference batch.

2.3.2. Triangle Test

A triangle test according to DIN EN ISO 4120:2007 was performed to determine possible sensory differences between the samples; this forced-choice approach is applicable especially in such cases when the nature of the potential difference is unknown. Overall, 19 participants were asked to determine the deviating sample out of a series of three samples or, if no difference was perceivable, to randomly choose one of them. In each case, 2g (±0.1 g) of the reference (untreated) plastic packaging waste or the probiotics-treated waste was respectively placed into a 140 mL covered glass vessel, randomly distributed, encoded and presented to the panelists. This procedure was performed in triplicate so that each panelist had to evaluate three series (in total nine single samples), leading to a total of 57 responses. Based on a significance level of $\alpha = 0.001$, at least 30 correct answers were required to prove a perceivable sensory difference.

2.3.3. Odor Profile Analysis

The in-house odor profile analysis was based on the industry standard DIN EN ISO 13299:2016-09. Panelists (cf. Section 2.3.1.) were required to describe their perceived orthonasal impressions, which were subsequently collated across the panel and shortlisted based on a consensus approval of the panel (more than 50% of panelists) to establish a selection of odor attributes that best characterize the perceived odor of the samples. Subsequently, each panelist was required to rate the intensity of each attribute in both samples on a scale from 0 (no perception) to 10 (strong perception), which was performed in comparison to odorant references that were presented in form of odorant pens. The reference compounds were 2-methylisoborneol (moldy/musty), geosmin (earthy), 3-phenylpropanoic acid (flowery/beeswax-like), (*E*)-2-nonenal (fatty/cardboard-like/cucumber-like) and octanal (soapy/citrus-like). Statistical differences were calculated by a paired, two-tailed distributed Student's *t*-test at $\alpha = 0.05$.

2.4. Characterization of Odorants

2.4.1. Solvent Extraction of Volatiles

Solvent extraction proceeded immersing 10 g (±0.1 g) of the respective sample material in 200 mL of dichloromethane and stirring for 30min at room temperature. After filtration through cotton wool, the volatile fraction, including odor-active compounds, was gently separated by solvent-assisted flavor evaporation (SAFE) according to Engel et al. [33]. The SAFE procedure was carried out under high vacuum (10^{-4} mbar) with a water bath temperature of 50 °C while the apparatus temperature was kept at 55 °C. After drying over anhydrous sodium sulfate, the distillate was concentrated to a total volume of ~100 µL by means of Vigreux distillation and subsequent microdistillation [34]. The successful extraction of odor-active compounds was verified by comparing the odor impression of a single drop of the obtained distillate applied on a filter paper with the odor of the respective input waste material. Details of chemicals and reference compounds used are listed in Appendix A.

2.4.2. Comparative Odor Extract Dilution Analysis

Relative odor intensities of single odor-active regions in gas chromatography–olfactometry (GC-O) (cf. Section 2.4.3) between treated and untreated samples were compared via comparative odor extract dilution analyses (cOEDA) [35,36]. The undiluted distillates (cf. Section 2.4.1), corresponding to odor dilution (OD) 1, were diluted stepwise (1 + 2 v/v) with dichloromethane to produce solutions for GC-O analysis that corresponded to OD factors 3, 9, 27, 81, 243 and 729. The higher the OD factor of a detected compound, the more relevant it is for the overall odor of the sample.

2.4.3. Gas Chromatography–Olfactometry

The undiluted distillates and 2 µL aliquots of each dilution (cf. Section 2.4.2) were analyzed by GC-O with a Trace GC Ultra (Thermo Fisher Scientific GmbH, Dreieich, Germany) using two capillary columns: DB-FFAP or DB-5 (both 30 m × 0.3 mm, film thickness 0.25 µm; J&W Scientific, Agilent Technologies, Waldbronn, Germany). Sample injection was applied using the cold on-column-technique with an initial oven temperature of 40 °C, which was then raised at 8 °C/min to either 235 °C (DB-FFAP) or 250 °C (DB-5). The final temperature was held for 5 min in both cases. Helium was used as carrier gas at a flow rate of 2.2 mL/min. At the end of the capillary column, the effluent was equally split in two parts and transferred via a Y-type glass splitter and deactivated fused silica capillaries (0.5 m × 0.2 mm, A-Z Analytik-Zubehör GmbH, Langen, Germany) to a flame ionization detector (FID, 250 °C) and an odor detection port (ODP, 270 °C). GC-O analyses of the original distillates were performed by three trained panelists.

The tentative identification of odorants was based on linear retention indices (RIs) on the two capillary columns of different polarities (DB-FFAP and DB-5) and calculated by analyzing a homologous series of *n*-alkanes [37], together with the perceived odor quality at the ODP in comparison to the respective reference compound. For further unequivocal identification, the molecular structure was elucidated by mass spectral data (if obtained) and also compared to reference compounds (cf. Section 2.4.4).

2.4.4. Two-Dimensional Gas Chromatography-Mass Spectrometry/Olfactometry

Two-dimensional gas chromatography–mass spectrometry/olfactometry (2D-GC-MS/O) was performed for the unequivocal identification of odorants based on mass spectral data using a system of two gas chromatographs (Varian CP-3800, Agilent Technologies, Waldbronn, Germany) equipped with a DB-FFAP column in the first dimension and a DB-5 column in the second dimension (cf. Section 2.4.3. for capillary column parameters). Both GCs were connected via a CTS 1 cryotrap system cooled with liquid nitrogen to −100 °C (Gerstel GmbH & Co. KG, Mülheim, Germany). Automated sample injection of 4 µL of the original distillates (OD1) by the cold on-column technique was performed with a MPS 2XL multipurpose sampler (Gerstel). After 2 min at an initial oven temperature of 40 °C in the first GC system, the temperature was raised at 8 °C/min to 230 °C and held for 5 min. The helium carrier gas flow was 9.0 mL/min. At the end of the first GC system, the effluent was split (1:1) via a Y-type glass splitter to an FID (250 °C) and an OPD (270 °C).

The area containing the analyte of interest was separated after passing the first GC system and transferred to the cryotrap by means of a MCS2 multi-column switching system (Gerstel). The subsequent transfer of volatiles to the second GC system was achieved by thermodesorption at 250 °C. In the second GC system, the initial temperature of 40 °C (2 min) was raised (8 °C/min) to 250 °C (1 min), with the effluent at the end of the capillary column again split in two equal parts, allowing the simultaneous detection of odor qualities at the ODP (270 °C) and mass spectra by means of a Saturn 2200 ion trap mass spectrometer (Agilent Technologies, Waldbronn, Germany). Mass spectral data were generated through electron ionization in positive mode at 70 eV ionization energy, over a *m/z* range of 35–399.

3. Results

3.1. Sensory Evaluation

With the initial focus on revealing possible odor differences, the post-consumer plastic packaging waste sample that had been treated with the probiotic bacterial solution was compared to the untreated reference sample via a triangle test (cf. Section 2.3.2). This forced-choice test returned 34 correct responses from a total of 57, indicating that the panel was able to discriminate the samples based on the perceived odor (α-risk: 0.001).

To further characterize the odor difference, 14 trained panelists agreed on five odor attributes as descriptors for both samples (cf. Section 2.3.3). These included moldy/musty, earthy, flowery/beeswax-like, fatty/cardboard-like/cucumber-like and soapy/citrus-like. The highest intensity ratings in both samples (treated and untreated) were for the attributes moldy/musty (mean intensity ratings of 6.7 and 7.3) and earthy (mean intensity ratings of 5.4 and 5.6), with ratings similar for both samples (Figure 2). By comparison, the attributes fatty/cardboard-like/cucumber-like and soapy/citrus-like were rated with lower but comparable intensities of 3.1/2.6 and 3.1/2.9 (reference sample/probiotics-treated sample). The lowest intensities were obtained for the attribute flowery/beeswax-like, which was rated at an intensity of 1.4 in both samples.

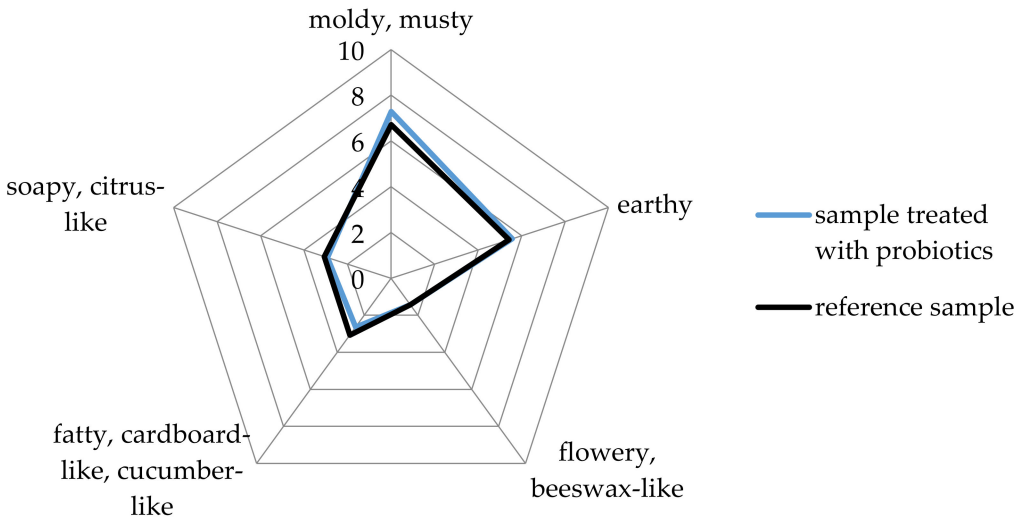

Figure 2. Mean intensity ratings during comparative odor profile analysis of the untreated reference and the probiotics-treated sample for the chosen attributes. Scale: 0 (no perception) to 10 (strong perception); $n = 14$.

Despite the differences perceived between the samples in the triangle tests, the mean intensity rating differences of individual attributes between the samples during odor profile analysis were not significant (paired, two-tailed distributed Student's t-test at $\alpha = 0.05$). This might be attributable to the relatively high variance in individual intensity ratings for single descriptors, as is regularly observed in sensory evaluations, possibly due to differences in individual odor sensitivities and/or odor thresholds [38].

3.2. Identification and Characterization of Causative Odorants

Gas chromatography–olfactometry (GC-O) analyses on the original distillate of the untreated reference sample performed by three trained panelists led to the detection of 57 odor-active regions (Table 1). Further analyses of each dilution (cOEDA, cf. Section 2.4.2) allowed an assignment of the most potent odorants to the respective odor dilution (OD) factors.

Commonly, most of the odorous substances in post-consumer plastic packaging waste have been found to be trace compounds that co-elute to a relevant extent with other (odorless) volatile substances, as previously observed in other studies [5]. Accordingly, in the majority of the cases two-dimensional GC-MS/O was required for unequivocal identification of odorants in both the reference and the probiotics-treated sample.

Table 1. Odorants identified in distillates of the reference (REF) and the probiotics-treated (PRO) plastic packaging waste sample, and comparison of respective OD factors obtained by cOEDA.

No.[a]	Odorant	Odor Quality [b]	OD [c]		RI [d] on		Identification Criteria [e]
			REF	PRO	DB-FFAP	DB-5	
1	2,3-butanedione	butter-like	9	1	984	<700	RI, O
2	ethyl 3-methylbutanoate	fruity, blueberry-like	3	1	1072	885	RI, O, MS
3	1-hexen-3-one	super glue-like, lighter gas-like	3	9	1096	774	RI, O
4	propyl 2-methylbutanoate	fruity, pineapple-like	9	9	1132	950	RI, O
5	(*Z*)-4-heptenal	fishy, fatty	9	<1	1230	895	RI, O
6	styrene	almond-like, pungent	27	9	1251	894	RI, O, MS
7	octanal	citrus-like, soapy	9	9	1280	1002	RI, O, MS
8	1-octen-3-one	mushroom-like	27	9	1291	979	RI, O, MS
9	2-acetyl-1-pyrroline	popcorn-like, roasty	81	9	1321	932	RI, O
10	(*Z*)-rose oxide	flowery, soapy	9	9	1344	1135	RI, O, MS
11	dimethyl trisulfide	garlic-like, cabbage-like	81	9	1365	970	RI, O, MS
12	2-mercapto-3-pentanone	cat urine-like, black currant-like	243	9	1367	946	RI, O
13	trimethylpyrazine	earthy, musty	27	9	1401	1022	RI, O, MS
14	(*E*)-2-octenal	fatty	243	81	1417	1058	RI, O, MS
15	2-furfurylthiol	roasted coffee bean-like	1	<1	1427	916	RI, O
16	acetic acid	vinegar-like	9	3	1435	738	RI, O, MS
17	methional	cooked potato-like	27	3	1444	905	RI, O
18	(*Z*)-2-nonenal	fatty, cardboard-like, green	1	<1	1494	1145	RI, O
19	3-isobutyl-2-methoxypyrazine	bell pepper-like, pea-like	27	9	1506	1177	RI, O
20	(*E*)-2-nonenal	fatty, cardboard-like	243	81	1523	1160	RI, O, MS
21	linalool	flowery, fresh	9	9	1534	1102	RI, O, MS
22	2-methylpropanoic acid	cheesy, sweaty	27	9	1552	765	RI, O
23	2-methylisoborneol	earthy, musty	729	27	1583	1191	RI, O, MS
24	2-acetyl-1-pyridine	popcorn-like	9	<1	1588	1026	RI, O, MS
25	butanoic acid	cheesy, sweaty	27	3	1618	804	RI, O, MS
26	acetophenone	marzipan-like, flowery	1	<1	1645	1069	RI, O, MS
27	3-methylbutanoic acid	cheesy, sweaty	81	27	1653	861	RI, O, MS
28	(*E*,*E*)-2,4-nonadienal	fatty	9	<1	1690	1213	RI, O, MS
29	pentanoic acid	sweaty, pungent	9	<1	1723	888	RI, O, MS
30	naphthalene	fuel-oil-like, fecal	9	9	1733	1191	RI, O, MS

Table 1. *Cont.*

No.[a]	Odorant	Odor Quality [b]	OD [c]		RI [d] on		Identification Criteria [e]
			REF	PRO	DB-FFAP	DB-5	
31	2-methylpentanoic acid	fruity, pungent	1	1	1751	932	RI, O
32	α-damascone	apple juice-like	27	27	1754	1394	RI, O, MS
33	3-methylpentanoic acid	fruity, sweaty, cinnamon-like	1	<1	1777	938	RI, O
34	2-phenylethyl acetate	rose-like, flowery	27	27	1815	1257	RI, O, MS
35	*trans*-anethole	anise-like	3	1	1821	1289	RI, O, MS
36	α-isomethylionone	flowery, rose-like	729	729	1835	1477	RI, O, MS
37	guaiacol	smoky, smoked ham-like	81	9	1852	1087	RI, O, MS
38	verdyl acetate	banana-like	729	729	1888	1420	RI, O, MS
39	β-ionone	violet-like, flowery	27	27	1918	1483	RI, O, MS
40	benzothiazole	rubber-like, car tire-like	81	27	1937	1227	RI, O, MS
41	heptanoic acid	plastic-like, sweaty, dusty	9	<1	1940	1087	RI, O
42	verdyl propionate	banana-like	27	9	1950	1527	RI, O, MS
43	1-methyl-β-ionone	flowery	3	3	1992	1583	RI, O, MS
44	γ-nonalactone	fruity, coconut-like	81	27	2018	1360	RI, O, MS
45	octanoic acid	musty, coriander-like, fatty	243	27	2039	1179	RI, O, MS
46	2,4-/2,5-dimethylphenol	fecal, smoky	3	<1	2069	1164	RI, O, MS
47	*p*-cresol	horse stable-like, fecal	729	27	2078	1085	RI, O, MS
48	γ-decalactone	peach-like, fruity	243	243	2129	1472	RI, O, MS
49	patchouli alcohol	moldy, earthy	27	27	2161	1664	RI, O, MS
50	sotolone	lovage-like, celery-like	1	3	2175	1102	RI, O
51	γ-undecalactone	peach-like, soapy	81	27	2255	1572	RI, O, MS
52	2,6-dimethoxyphenol	smoky, smoked ham-like	3	<1	2283	1650	RI, O, MS
53	γ-dodecalactone	fruity, peach-like	3	<1	2361	1679	RI, O, MS
54	unknown	mushroom-like	729	729	2387	1754	-
55	indole	fecal	3	<1	2448	1310	RI, O, MS
56	skatole	fecal, mothball-like	27	9	2474	1391	RI, O, MS
57	unknown	fecal	9	<1	2595	n.d. [f]	-

[a] Consecutive numbering of odorants according to their elution on capillary column DB-FFAP. [b] Odor quality perceived at the odor detection port by three trained panelists. [c] Odor dilution (OD) factor on capillary column DB-FFAP according to Grosch [35]. [d] Retention index (RI) on capillary columns DB-FFAP and DB-5 according to Van den Dool and Kratz [37]. [e] Identification of odorants based on retention index (RI), odor quality (O) and mass spectrum (MS); respective data were compared to those of a reference compound. [f] n.d. not detected.

For the reference sample, five odor-active substances were still perceivable at the highest dilution, which corresponded to OD factor 729, indicating a major contribution of these substances to the overall odor of the respective input plastic packaging waste material. These included 2-methylisoborneol (Table 1, no. 23, earthy, musty), α-isomethylionone (36, flowery, rose-like), verdyl acetate (38, banana-like), *p*-cresol (47, horse stable-like, fecal), and an unknown mushroom-like smelling compound (54). Another five odorants that corresponded to the second highest OD factor of 243 were successfully identified as 2-mercapto-3-pentanone (12, cat urine-like, black currant-like), (*E*)-2-octenal (14, fatty), (*E*)-2-nonenal (20, fatty, cardboard-like), octanoic acid (45, musty, coriander-like, fatty) and γ-decalactone (48, peach-like, fruity). Seven odorous substances were perceived at OD 81: 2-acetyl-1-pyrroline (9, popcorn-like, roasty), dimethyl trisulfide (11, garlic-like, cabbage-like), 3-methylbutanoic acid (27, cheesy, sweaty), guaiacol (37, smoky, smoked ham-like), benzothiazole (40, rubber-like, car tyre-like), γ-nonalactone (44, fruity, coconut-like) and γ-undecalactone (51, peach-like, soapy); whereas additional 13 odorants were detected at OD 27: styrene (6, almond-like, pungent), 1-octen-3-one (8, mushroom-like), trimethylpyrazine (13, earthy, musty), methional (17, cooked potato-like), 3-isobutyl-2-methoxypyrazine (19, bell pepper-like, pea-like), 2-methylpropanoic acid (22, cheesy, sweaty), butanoic acid (25, cheesy, sweaty), α-damascone (32, apple juice-like), 2-phenylethyl acetate (34, rose-like, flowery), β-ionone (39, violet-like, flowery), verdyl propionate (42, banana-like), patchouli alcohol (49, moldy, earthy) and skatole (56, fecal, mothball-like). Additionally, 27 odorous substances most likely contributed to the overall odor of the sample to a lesser extent, being perceived in dilutions below OD 27, among them fruity or flowery smelling substances (no. 2, 4, 7, 10, 21, 26, 31, 35, 43, 53) and several fecal, fatty or sweaty smelling odorants (no. 18, 28, 29, 30, 33, 41, 46, 52, 55, 57).

Of the total 57 odorous contaminants detected in the untreated reference sample, 43 substances were also perceived in the undiluted distillate (OD 1) of the plastic packaging waste sample that had been treated with the probiotic bacterial cultures. On the other hand, no additional odorants were detectable, and only four odorants, namely α-isomethylionone (36), verdyl acetate (38), the unknown mushroom-like smelling compound (54) and γ-decalactone (48), were identified in the treated sample at the highest OD factors of 729 or 243, respectively. A direct comparison of both samples further revealed that 15 of the 43 odorants were detected at the same OD factor in each case. In contrast, and with the sole exception of 1-hexen-3-one (3), the remaining 27 odorants showed lower OD factors in the treated sample, indicating that more than 70% of the odorous contaminants were perceived at lower intensity during cOEDA. Odorous compounds that differed by more than three dilution steps between the two samples were 2-mercapto-3-pentanone (12), 2-methylisoborneol (23) and *p*-cresol (47).

4. Discussion

4.1. Comparison of Human Sensory Evaluation and Analytical Results

The triangle test yielded significant differences in the overall odors of the untreated reference sample and the sample treated with probiotic bacterial cultures. To further objectify this difference an odor profile analysis was performed, which confirmed a major odor load for both samples. Overall, the moldy/musty and earthy smell impressions dominated the overall profiles of both samples, with both attributes rated with high intensities (6.7/7.3 and 5.4/5.6, respectively). Comparison with the GC-O data indicated that 1-octen-3-one (8), trimethylpyrazine (13), octanoic acid (45), patchouli alcohol (49) and especially the earthy/musty smelling 2-methylisoborneol (23) are the main compounds likely to be responsible for this odor, especially the latter, which yielded the highest OD factor in the reference sample. Further, the fatty smelling odorants (*E*)-2-octenal (14), (*E*)-2-nonenal (20) and (*E*,*E*)-2,4-nonadienal (28) are likely to contribute to the fatty odor impression rated with moderate intensities in both samples (3.1/2.6). Interestingly, the attributes soapy and flowery were rated with the lowest intensities despite the fact that the instrumental olfactometric analyses revealed a variety of flowery and soapy smelling substances (no. 7, 10, 21, 34, 36, 39, 43). While most of these flowery or

soapy smelling compounds were perceivable at the highest OD for both samples, they are likely to be covered by other potent malodorous compounds, for example the moldy smelling 2-methylisoborneol. One reason might be that complex mixtures of odor-active compounds may cover or enhance the impact of single odorants, even to an extent that individual odor impressions are no longer attributable to single odorous compounds [35,39–41]. It should also be noted that many of these flowery and soapy smelling substances exhibit a higher volatility. Due to the complete evaporation of the distillate on the GC column compared to vapor pressures at room temperature, higher OD factors are therefore less important for these odorants [42].

The complexity of the odorant composition might also explain why the two samples did not exhibit significant differences in any of the rated attributes despite the clear discernibility of the samples in the triangle test. Additionally, as determined by cOEDA, potent odorants (OD factor ≥ 27 in the reference sample) with the highest differences in OD factors between the reference and the probiotics-treated sample (no. 9, 11, 12, 17, 23, 25, 37, 45, 47) showed deviations of a maximum of three OD steps. However, these compounds were still perceivable in both samples in any case, meaning that none of these compounds was fully eliminated through the probiotic treatment. This might explain why the odor profile analysis did not provide a satisfactory differentiation between the two samples on the basis of the defined odor attributes. Otherwise, in the case of the attributes soapy and flowery, which were rated with similar intensities in both samples, the sensory evaluation correlated well with the cOEDA results, as most flowery or soapy smelling substances were detected at the same OD factor in both samples. Accordingly, cOEDA served as a necessary approach to distinguish molecular differences in single odorous constituents between the samples.

4.2. Potential Origin of Identified Odorants

A large number of odorous contaminants was identified in the post-consumer plastic packaging waste of the present study, which is in agreement with previous studies on household plastic waste or recycled plastic materials [5,9,29,40,43]. However, 17 of the detected odorants (no. 2, 3, 6, 10, 13, 19, 30, 31, 33, 34, 37, 40, 41, 45, 46, 52, 57), which represent around 30% of all detected substances, are reported here for the first time as odor-active contaminants in plastic packaging waste and recycled plastics. A reason for this large number of previously unreported odorants might be the inhomogeneity of post-consumer material, and variations in degree of contamination and, accordingly, odor load.

Generally, odorants stem from various sources or via diverse formation pathways. The majority of the odor-active compounds identified here represent typical metabolites of microorganisms, such as 2-methylisoborneol (23), *p*-cresol (47) and diverse short-chain carboxylic acids (cf. Section 4.3). This indicates that the off-odor primarily originates from substances emitted by bioconversion of organic residues typically found in post-consumer waste. Only about 20% of the identified odorants most likely stem from residual fragranced filling goods, such as washing and cleaning agents. Compounds such as α-isomethylionone (36) and verdyl acetate (38) are especially commonly used as fragrance compounds [44]. The pronounced appearance of microbial metabolites is in line with previous findings on post-consumer plastic packaging waste and plastic film fractions [5,29,45]. Deviant from the smell composition of the mixed polyolefin fractions, recycled post-consumer high-density polyethylene (HDPE) and polypropylene (PP) has been found to be additionally dominated by the soapy/flowery smells, corresponding to the detection of typical fragrance compounds, as discussed above [9,13,40].

Additionally, several odorants are also likely to stem from the plastic polymer, and possibly additives, representing oxidation and degradation products thereof. In previous studies, unsaturated aldehydes, short-chain carboxylic acids and γ-lactones have been reported as odor-active compounds in plastics [46–49].

4.3. Odor Reduction by the Application of Probiotic Bacterial Cultures

Significant differences between the odors of the untreated reference sample and the sample treated with probiotic bacterial cultures were found via triangle tests and cOEDA. Thereby, ten odorants exhibited lower OD factors by two or more dilution steps, and 14 odorous contaminants present in the

reference sample were not perceived at all in the undiluted distillate of the probiotics-treated sample. Although cOEDA is only a screening method to obtain an approximate odor contribution weighting of the individual substances, it nevertheless serves the goal of obtaining an indication of relative quantitative differences [36]. Accordingly, major differences in OD steps or even non-detection versus detection between samples is a strong indicator that the related odorant quantities were influenced to a major extent by the process under investigation.

The vast majority of odorants showing noteworthy OD differences in this study most likely relate to the bioconversion of organic residue materials, since they have been reported elsewhere to be common metabolites of microorganisms. In particular, especially the group of short-chain carboxylic acids (no. 16, 25, 29, 41, 45) and methylated carboxylic acids (no. 22, 27, 31, 33) exhibited lower OD factors in the probiotics-treated sample or were not detected at all. Generation of volatile acids has been reported for different bacterial species, such as *Clostridium*, *Lactobacillus*, *Pediococcus* and *Leuconostoc* [50,51], and might explain their previous detection in organic waste and wastewater [52–54]. Various other compounds can also be categorized as bacterial metabolites of fatty acids, for instance 2,3-diketones (no. 1), diverse esters (no. 2, 4) and lactones (no. 44, 48, 51, 53), which have been reported to be generated by microorganisms such as *Staphylococcus, Stigmatella, Loktanella* and *Dinoroseobacter* [55,56]. On the other hand, such low molecular weight compounds themselves may serve as nutrient and energy source for the microbiota. In the present study, the majority of the detected fatty acid derivatives was perceived at lower OD factors in the probiotics-treated sample than the untreated reference sample, indicating a notable trend of reduction by the application of the probiotic bacterial solution.

Apart from that, substantial OD differences were observed for the sulfur compounds dimethyl trisulfide (11), 2-mercapto-3-pentanone (12) and methional (17). All of these compounds have been previously identified as odorous contaminants in post-consumer plastic waste and are linked to the bacterial spoilage of organic waste [29,43]. Another group of potential odorous metabolites of microorganisms, which were perceived with lower OD factors in the distillate of the probiotics-treated sample, were nitrogen compounds, such as the earthy/green smelling pyrazines trimethylpyrazine (13) and 3-isobutyl-2-methoxypyrazine (19), and popcorn-like smelling pyrroles and pyridines (2-acetyl-1-pyrroline (9), 2-acetyl-1-pyridine (24)). Pyrazines represent prominent volatiles produced by bacteria, with methoxypyrazines exhibiting an especially intense odor [55]. The same trend applied for aromatic N-heterocyclic compounds like benzothiazole (40) and, in particular, indole (55) and skatole (56) with a characteristic fecal odor [55,57–61]. Likewise, the terpenoid 2-methylisoborneol, which exhibited the highest OD factor of 729 in the untreated sample but only OD 27 in the probiotics-treated sample, is known to be formed by actinomycetes, cyanobacteria and myxobacteria [55,62,63]. On the other hand, also several phenolic compounds were primarily detected in the reference sample (no. 37, 46, 47, 52), most likely stemming from rotting processes of organic matter. A prominent example is the fecal smelling *p*-cresol (47) that is also a common constituent of feces [61,64].

Unlike the detected microbial metabolites, the majority of typical fragrance constituents (no. 7, 10, 21, 32, 34, 36, 38, 39, 42, 49) was perceived at the same OD factors in both samples (Figure 3). Especially α-isomethylionone (36) and verdyl acetate (38) were perceived even into the highest dilution of the two samples by means of olfactory detection, substantiating the comparable contamination of both samples with regard to typical fragrances. Consequently, the present study shows that, in this case and under these process conditions, probiotics are not capable of degrading such fragrance-related odorants. As such, the contamination degree of the input plastic packaging waste material with these fragrances remained unaffected by probiotic bacterial treatment. This finding calls for new strategies in the fragrance industry in view of designing recycling and biodegradation strategies for filling goods and scent constituents, appropriate measures to avoid fragrance compound contamination of the plastic material, and additional decontamination strategies, ideally combined as a systemic strategy.

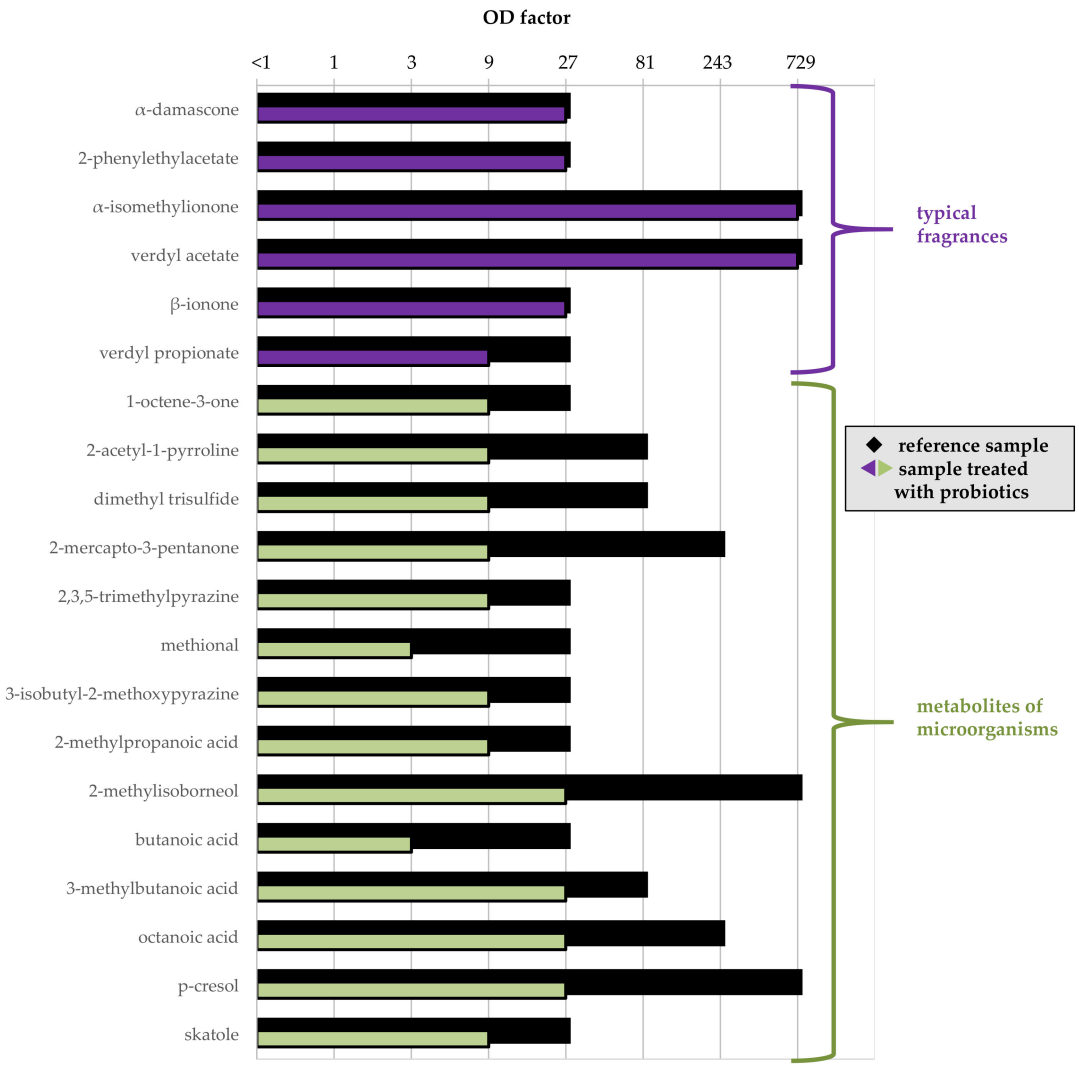

Figure 3. Odor dilution (OD) factors of the most important odor-active compounds (OD ≥ 27 in the reference sample), representing either typical fragrances or characteristic metabolites of microorganisms detected in distillates of the untreated reference post-consumer plastic packaging waste sample in comparison to the same waste fraction treated with probiotic bacterial cultures.

The reduction in OD factors between the untreated and treated sample indicates a quantitative reduction of these odor-active compounds in the plastic packaging waste fraction after probiotic treatment. Moreover, no additional odorants were detected in the latter, indicating that only non-odorous derivatives were formed, while odor-forming microorganisms may have been additionally limited in growth. Nevertheless, a complete inhibition of odor-forming microbial activity was not achieved, potentially due to the initial odor load being too abundant to be fully removed. Hence, the treated sample still exhibited a relatively strong odor.

On the contrary, the probiotic bacteria affecting other properties of the plastic material is unlikely given that the probiotic aqueous solution did not contain any further chemicals. Additional moisture content may not be of importance since the material was mechanically and thermally dried at the end of the evaluated recycling process. Regarding the applied probiotic bacteria themselves, a modification of plastic polymers is unlikely, given that in literature it was only shown for specifically isolated bacteria to be able to decompose polyethylene terephthalate (PET) [65]. On top, common post-consumer plastic waste also contains various kinds of probiotic bacteria, originating, amongst others, from diverse food residues such as dairy products, whilst a modification of the plastic packaging has not been reported so far.

In short, we deem the implementation of probiotics in the plastics recycling chain as a promising strategy towards the reduction of unwanted organic matter and odor. It is important to mention that post-consumer plastic packaging waste is commonly stored for extended periods of up to several weeks or months starting from the collection in households until usage for recyclate production. This favors abundant spoilage processes and, consequently, odor contamination. New strategies in the collection, logistics and storage of plastic packaging waste, together with the right timing of implementing treatment with probiotics, are in our opinion crucial for successful implementation of such biological strategies into a systemic waste management and recycling concept. This is especially true when it comes to potent odorants that annoy consumers even at lowest concentration levels. Their removal will be essential for consumer acceptance and future recycling strategies for deodorizing post-consumer plastic packaging waste.

5. Conclusions

The focus of the present study was on the odor contamination of post-consumer plastic packaging waste. A combinatory approach of using human sensory and gas chromatography–olfactometric analyses allowed the determination of potential odor minimization through the use of probiotic bacteria. More precisely, the high odor load of the investigated post-consumer plastic packaging waste was evident from the 57 odor-active compounds detected, the majority of which represent typical odorous bacterial metabolites with various chemical structures. The application of probiotic bacterial cultures to the input material, followed by 40 day storage (summer 2019 in Germany), led to significant differences in the overall odor profile compared with an untreated reference. Characterization of the causative odor-active compounds by means of cOEDA revealed a reduction in the majority of odorants, specifically those substances that most likely originated from microbial degradation of organic matter. On the other hand, no effect was observed for contaminants that presumably originated from residual fragranced filling goods. The reduced perceived intensity of microbial odorants, as determined by cOEDA, demonstrated a measurable effect of the applied probiotics. Although the probiotics-treated plastic packaging waste fraction still showed a high odor contamination, the detected reduction of odorous microbial metabolites in this study makes the use of probiotics a promising alternative solution to odor optimization in plastic packaging recycling. In view of the current extended storage periods of plastic waste, especially in the case of post-consumer packaging waste, further means such as an earlier implementation of probiotic treatment will be required.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

Chemicals

The solvents pentane and dichloromethane (freshly distilled prior to use), as well as anhydrous sodium sulfate, were obtained from Th. Geyer GmbH (Renningen, Germany); liquid nitrogen was supplied by Linde Gas (Pullach im Isartal, Germany). For the determination of retention indices, a homologues series of linear *n*-alkanes in pentane was used (Fluka, Steinheim, Germany and Sigma-Aldrich, Steinheim, Germany). The following reference compounds were obtained from

Sigma-Aldrich (Steinheim, Germany): acetic acid ≥99%, acetophenone ≥98%, 2-acetyl-1-pyridine ≥99%, benzothiazole ≥96%, γ-decalactone ≥98%, 2,6-dimethoxyphenol ≥99%, (*RS*)-(±)-3,7-dimethyl-1,6-octadien-3-ol ((*RS*)-(±)-linalool) ≥97%, 2,5-dimethylphenol ≥99%, dimethyl trisulfide ≥98%, ethyl 3-methyl butanoate ≥98%, 2-furfurylthiol ≥98%, heptanoic acid ≥99%, (*Z*)-4-heptenal ≥98%, 3-hydroxy-4,5-dimethylfuran-2(5*H*)-one (sotolone) ≥97%, 2-methoxyphenol (guaiacol) ≥99% (*trans*)-1-methoxy-4-(1-propenyl) benzene ((*trans*)-anethole) ≥99%, 2-methylbutanoic acid ≥98%, 3-methylbutanoic acid ≥99%, 3-methylindole (skatole) ≥98%, 4-methylphenol (*p*-cresol) ≥ 99%, 3-(methylthio)propanal (methional) ≥97%, 3-methyl-4-(2,6,6-trimethyl-2-cyclohexenyl)-3-buten-2-one (α-isomethylionone) ≥99%, (*E*,*E*)-nona-2,4-dienal ≥85%, γ-nonalactone ≥98%, (*E*)-2-nonenal ≥ 97%, (*Z*)-2-nonenal ≥90%, octanal ≥99%, octanoic acid ≥98%, (*E*)-2-octenal ≥94%, 1-octen-3-one ≥50%, (*Z*)-rose-oxide of unknown purity, styrene ≥99%, 1,2,7,7-tetramethylbicyclo[2.2.1]heptan-2-ol (2-methylisoborneol) ≥98%, 1-(2,6,6-trimethyl-2-cyclohex-1-en-1-yl)-but-2-en-1-one (α-damascone) ≥95%, (3*E*)-4-(2,6,6-trimethylcyclohex-1-en-1-yl)-but-3-en-2-one (β-ionone) ≥96%, (*E*)-1-(2,6,6-trimethylcyclohex-1-en-1-yl)-pent-1-en-3-one (1-methyl-β-ionone) of unknown purity, trimethylpyrazine ≥99% and γ-undecalactone ≥98%.

Verdyl acetate (3*a*,4,5,6,7,7*a*-hexahydro-4,7-methano-1*H*-inden-6-yl acetate; ≥97%) and verdyl propionate (3*a*,4,5,6,7,7*a*-hexahydro-4,7-methano-1*H*-inden-6-yl propionate ≥96%) were obtained from Essencia AG (Winterthur, Switzerland), patchouli alcohol (3,4,4$\alpha\beta$,5,6β,7,8,8α-octahydro-4α,8$\alpha\beta$,9,9-tetramethyl-1,6-methanonaphthalen-1β(2*H*)-ol, ≥98%) was obtained from Biozol (Eching, Germany) and 2,3-butanedione ≥99%, butanoic acid ≥99,5%, 2,4-dimethylphenol ≥97%, indole ≥98.5%, 2-methylpentanoic acid ≥98%, 3-methylpentanoic acid ≥97%, naphthalene ≥99%, pentanoic acid ≥99% and 2-phenylethyl acetate ≥99% were obtained from Fluka (Steinheim, Germany). Other reference compounds were 2-acetyl-1-pyrroline (1-(3,4-dihydro-2*H*-pyrrol-5-yl)ethanone, ≥95%, aromaLAB AG, Freising, Germany), 1-hexen-3-one ≥90% (ABCR, Karlsruhe, Germany), 3-isobutyl-2-methoxypyrazine ≥99% (Acros Organic, Geel, Belgium), 2-mercapto-3-pentanone ≥95% (TCI Europe, Zwijndrecht, Belgium), propyl-2-methyl butanoate of unknown purity (Symrise AG, Holzminden, Germany) and 2-methylpropanoic acid ≥99%, as well as γ-dodecalactone ≥97 %, from SAFC (Steinheim, Germany).

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