Phytochemistry, Vol. 28, No. 7, pp. 1817–1824, 1989.
Printed in Great Britain.

0031-9422/89 $3.00+0.00

VOLATILE COMPONENTS OF CELERY AND CELERIAC

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(*Received in revised form* 11 *November* 1988)

Key Word Index—*Apium graveolens* var. *dulce*; *Apium graveolens* var. *rapaceum*; Umbelliferae; celery; celeriac; aroma volatiles.

Abstract—Volatile components of celery and celeriac were analysed using routine procedures. A high proportion of each isolate consisted of monoterpene hydrocarbons (46.0% celery; 24.8% celeriac) and phthalides (42.3% celery; 37.4% celeriac). Major components of both isolates were limonene and 3-butyl-4,5-dihydrophthalide (or sedanenolide). Celery volatiles contained higher concentrations of γ-terpinene and α-pinene, whereas those of celeriac had greater quantities of α-terpinene and β-pinene. A celery-like odour was associated during GC elution with each of the 16 phthalides reported, of which (*E*)-3-butylidene-4,5-dihydrophthalide (or (*E*)-ligustilide) and *cis,syn*-3-butyl-3a,4,5,7a-tetrahydrophthalide (or cnidilide) could not be detected in celeriac.

INTRODUCTION

Cultivated celery (*Apium graveolens* L. var. *dulce*) and celeriac (*Apium graveolens* L. var. *rapaceum*) are closely related members of the Umbelliferae, but, whereas the leaf stem is the edible part of celery, the part of the celeriac that is eaten is the swollen base of the stem, often referred to incorrectly as a 'root'. In both cases, they can be eaten raw in salads or as cooked vegetables or in soups, stews, etc. The flavour of celeriac is similar to that of celery, but it has never equalled celery in wide popularity, and this is reflected in the number of publications on the volatile components of the two vegetables.

The volatiles of celery have been studied by several workers [1–11], although some of the early work was, in fact, performed on celery seeds. In all, about 165 components have been characterized, and notable features are the reported presence of several terpenes and some characteristic phthalides. Only five publications describe the volatile constituents of celeriac [10, 12-15], which amount to about 40 in number. In this paper, we report the results of direct comparative analyses of the volatile components of celery (*Apium graveolens* L. var. *dulce*, 'Celebrity') and celeriac (*Apium graveolens* L. var. *rapaceum*, 'Monarch'), which have not been published hitherto.

RESULTS AND DISCUSSION

Fresh celery and celeriac were purchased from a local supermarket, and aroma extracts were prepared and validated using well-established procedures [16, 17]. The odour of each extract was representative of the vegetable extracted; both were described as strongly celery-like, whilst the celeriac aroma extract possessed additional slight buttery and caramel overtones. Four extractions were performed for each vegetable, and the replicate extracts were combined before concentration by high vacuum-low temperature distillation [18]. The resultant isolates, on appropriate re-dilution, possessed strong characteristic celery aromas as described above for the respective extracts.

Isolates were analysed by GC, GC-MS and by the technique of GC odour port assessment (GC-OPA) [19], and results are given in Tables 1 and 2. Fused silica capillary GC columns were used, containing either bonded-phase DB5 or BP20. Best resolution was achieved using the DB5 column, and the majority of the aroma components were identified using this phase (Table 1); however, a small number of components was identified only from the BP20 column (Table 2). Literature Kováts retention indices [20–25] of most components are also included in the Tables, and confirm the general elution sequence. Where positive identities are given, the mass spectra obtained on GC-MS agreed with those in the literature [10, 14, 20, 21, 25–34].

Quantitatively, as assessed from total peak areas, the celery isolate was stronger than the celeriac isolate (obtained identically) by a factor of about six. In all, from the celery volatiles, 68 components (comprising *ca* 92.0% of the isolate) were positively identified, with a further 12 (*ca* 1.0%) partially characterized. Of the fully identified components, 28 are reported as celery volatiles for the first time, and these are indicated in Tables 1 and 2 by ¶. With regard to the celeriac volatiles, 46 components (comprising *ca* 79.0% of the isolate) were positively identified, with a further 3 (*ca* 2.0%) partially characterized. Of the fully identified components, 21 are reported as celeriac volatiles for the first time, and these are indicated in Tables 1 and 2 by ‖.

Some interesting features of the chemical composition of the two isolates relate to their content of terpenoid compounds. As a group, the 18 monoterpene hydrocarbons represented as much as 46.0% of the celery isolate, but only 24.8% of the celeriac sample. Nine of the monoterpene hydrocarbons identified in celery—namely camphene, α-thujene, terpinolene, β-phellandrene, 4-isopropenyl-1-methylbenzene and the *p*-menthatrienes—could not be detected in the celeriac isolate. The major component in both isolates was limonene, constituting

Table 1. Volatile components of celery and celeriac (DB5 column GC-MS)

Component and ref. MS (lit)*	R_t (min)	Kováts Index (lit)†	Celery % RA‡	Celery Odour §	Celeriac % RA‡	Celeriac Odour §
‖ Dimethyl sulphide	5.24		tr.		0.2	
‖¶ 2,3-Dimethylbutane	6.31		—		0.2	
‖¶ 2-Methylpentane	6.41		0.1		1.9	
‖¶ 3-Methylpentane	6.91		0.1		0.7	
‖¶ Hexane	7.59	600	0.1		6.0	
‖¶ Methylcyclopentane	9.12		tr.		1.6	
‖¶ 2-Methylbutan-2-ol [20]	9.62	631	tr.		tr.	
‖ 3-Methylbutanal	10.66	649	—		0.3	
‖¶ Cyclohexane	11.17	677	tr.		0.3	
‖¶ 2-Methylbut-2-enal	17.18		tr.		tr.	
‖¶ Pyridine	17.41	695	tr.		0.1	
¶ 1,2- or 1,3-Dimethylcyclohexane	20.73		tr.		—	
‖ Hexanal [21]	21.99	780	tr.		0.1	
¶ Octane [20]	22.63	800	0.1		—	
¶ 1,3- or 1,2-Dimethylcyclohexane	23.30		tr.		—	
‖ 2-Furaldehyde [20]	26.03	815	tr.		0.1	
a branched C_9 hydrocarbon	29.11		tr.		—	
‖¶ Nonane [20]	32.74	900	0.3		1.8	boiled milk
Heptanal [20]	32.98	883	—		0.1	warmed-over meat, stale boiled potatoes
¶ α-Thujene [21]	35.69	938	0.1		—	
α-Pinene [21]	36.53	942	0.5	fragrant	0.2	fragrant, sweet
Camphene [21]	38.13	948**	0.1		—	
¶ Benzaldehyde [20]	39.09	947	tr.	nutty	—	
Sabinene [21]	40.29	976	0.2		tr.	oily
β-Pinene [21]	40.68	981	1.8		4.4	
Myrcene [21]	41.55	983**	0.9	sl. fragrant, stored apples, unpleasant	0.7	stale, wet hay, green, fragrant, unpleasant
¶ Cumene (i.e. isopropylbenzene)	42.02		tr.		—	
Octanal [20]	42.54	985	tr.	linseed oil, putty, nutty, stale green, unpleasant	0.5	linseed oil, putty, nutty, stale green, unpleasant
α-Terpinene [21]	43.99	1015**	0.1		3.0	
p-Cymene [21]	44.73	1020	0.1		0.1	oily, nutty
Limonene [20, 21]	45.72	1022**	35.5	sweet, fruity, sl. citrus, green, fragrant	14.9	fruity, fragrant
¶ β-Phellandrene [27]	45.83	1025	tr.		—	
cis-β-Ocimene [21]	46.46	1025	0.1	oily, petrol, unpleasant	tr.	petrol, unpleasant
‖¶ Phenylacetaldehyde	46.54	1024	tr.	floral, green, fragrant	0.2	roses, geraniums, fragrant
γ-Terpinene [21]	47.92	1057	6.6	throaty, pungent	1.6	

¶3-Methylthiophen-2-carboxaldehyde [28]	49.79		tr.	sl. roasted cereal	—	
Terpinolene [21]	50.16	1083**	tr.		—	
¶Linalool [21]	50.78	1086**	tr.	roses, fragrant	—	
Nonanal [20]	51.05	1087	tr.	fragrant	0.1	fragrant, herbal
¶a *p*-Menthatriene [21, 29]	52.83		tr.		—	
¶a *p*-Menthatriene [21, 29]	53.97		tr.		—	
an Undecatriene [22]	54.12		tr.		—	
a Pentylcyclohexadiene [30]	55.85		0.3	fragrant, oily, green	1.8	fragrant, sweet, sickly
Pentylbenzene	56.00		tr.		0.1	
(*E*,*Z*)-Undeca-1,3,5-triene [22]	56.69	1166††	tr.	fruity, fragrant, very pleasant	—	
Terpinen-4-ol	57.58	1175	0.1		tr.	
α-Terpineol	58.57	1185	0.1		0.3	
¶a *p*-Menthatriene [21, 29]	59.34		tr.		—	
Carveol	60.70	1209 1222	tr.		—	
4-Isopropenyl-1-methylbenzene [21]	60.91	1277**	tr.		—	
‖¶2-Methoxy-4-vinylphenol (i.e. *p*-vinylguaiacol)	67.86		tr.	cooked beans, musty, stale	1.4	cloves, cinnamon, cooked beans
p-Mentha-1,3,8-triene [21, 29]	69.31		0.1		—	
¶α-Cedrene [31]	75.94	1436	tr.	sweet, fruity	—	
Caryophyllene [20, 31, 32]	76.35	1428	1.2		0.1	roasted cereal, fragrant
α-Humulene [31]	78.59	1465	0.2		—	
¶*ar*-Curcumene [32]	79.52	1500‡‡	0.4	fragrant, stale, petrol	—	
β-Selinene [31]	80.88	1530§§	1.0	green, fragrant	—	
a Selinene [31]	81.29		0.2	fragrant	—	
¶δ-Cadinene [31]	82.71	1524	tr.		—	
an Alkyl phthalide	84.74		tr.	*celery*	—	
an Alkyl phthalide	88.30		tr.	*celery*	—	
‖a 3-Butylhexahydrophthalide [33]	90.34		0.1	sweet, sickly, *cooked celery*	2.1	*braised celery*, peppery, smoky
3-Butylphthalide [10, 14, 25, 34]	91.49	1633‖‖	2.5	sweet, fruity	1.5	*fresh celery*, green, fragrant, sweet
‖¶γ-Dodecalactone [20]	92.09	1647	tr.	fruity, fragrant	tr.	
‖a 3-Butylhexahydrophthalide [33]	92.14		tr.		3.3	*celery*, fruity, fragrant
‖¶Methyl 6-pentanoylcyclohex-1-enoate (i.e. methyl sedanonate) [33]	92.28		tr.	*celery*	0.2	*fresh celery*, pungent
(*Z*)-3-Butylidenephthalide [25, 34]	92.58	1660‖‖	0.1	*celery*, fruity, fragrant	0.5	*fresh celery*, pungent
a Phthalide M192	93.10		0.5	*fresh celery*, sweet, fragrant, pungent	0.1	*celery*, chicory
a Phthalide M192	93.36		tr.		tr.	*raw celery*, pungent
(*E*)-3-Butylidenephthalide [25, 34]	95.73	1709‖‖	1.0	*cooked celery*	0.2	*cooked celeriac*, sickly
3-Butyl-4,5-dihydrophthalide (i.e. sedanenolide) [10, 14, 25, 33, 34]	96.69	1702‖‖	28.1	*celery*, pungent	13.4	*cooked celeriac*
cis-3-Butyl-3a,4,5,6-tetrahydrophthalide (i.e. *cis*-sedanolide) [10, 25, 34]	96.92	1713‖‖	5.0	*celery*, pungent	5.3	*fresh celery*, medicinal, oil of wintergreen
(*Z*)-3-Butylidene-4,5-dihydrophthalide (i.e. (*Z*)-ligustilide) [25, 34]	97.20	1724‖‖	3.5	*celery*, pungent	3.5	*fresh celery*, smoky, astringent, sl. medicinal

Table 1. *Continued*

Component and ref. MS (lit)*	R_t (min)	Kováts Index (lit)†	Celery % RA‡	Celery Odour §	Celeriac % RA‡	Celeriac Odour §
trans-3-Butyl-3*a*,4,5,6-tetrahydrophthalide (i.e. *trans*-sedanolide) [10, 25, 34]	97.94		1.4	*celery*, less pleasant, sap-like, camphor, petrol	7.3	*fresh celery*, peppery
(*E*)-3-Butylidene-4,5-dihydrophthalide (i.e. (*E*)-ligustilide) [25, 34]	100.57	1789‖ ‖	0.1	*celery*, camphor	—	

*MS lit. [26] when not cited.
†Kováts index lit. [20] for nearest stationary phase (OV 101), confirming general elution sequence.
‡RA = relative abundance; if <0.1% then quoted as trace (tr.); — not detected.
§By GC odour port assessment; odours most relevant to celery and celeraic are italicised.
**Lit. [21].
††Lit. [22].
‡‡Lit [23].
§§Lit. [24].
‖‖Lit. [25].
¶Reported as celery volatiles for the first time.
‖Reported as celeriac volatiles for the first time.

Table 2. Additional volatile components of celery and celeriac (BP20 column GC–MS)

Component and ref. MS (lit)	R_t (min)	Kováts Index (lit)*	% RA† Celery	% RA† Celeriac
Butanedione [20]	2.36	963	tr.	—
trans-β-Ocimene [27]	11.82	1250	tr.	tr.
cis-*p*-Mentha-2,8-dien-1-ol [21]	35.39		tr.	tr.
¶Santalene [31]	37.35	1671‡	tr.	—
‖Neral [21]	37.84	1680	tr.	tr.
a Dihydrosesquiterpene (M206)	40.53		tr.	—
¶*cis, syn*-3-butyl-3*a*,4,5,7*a*-tetrahydrophthalide (i.e. cnidilide) [34]	73.42		tr.	—

*Kováts index lit. [20] for PEG 20 M.

†RA = relative abundance; tr. = trace, i.e. <0.1%; — not detected.

‡lit. [24].

¶reported as celery volatiles for the first time.

‖reported as celeriac volatiles for the first time.

35.5% of the celery volatiles and 14.9% of the celeriac isolate. However, an interesting distinguishing feature is that, whereas the celery volatiles also contained higher concentrations of γ-terpinene (6.6 vs 1.6%) and α-pinene (0.5 vs 0.2%), the celeriac volatiles were characterized by greater quantities of the alternative isomers, i.e. α-terpinene (3.0 vs 0.1%) and β-pinene (4.4 vs 1.8%). Considering now the nine identified sesquiterpene hydrocarbons as a group, they represented 3% of the celery aroma—this being largely due to caryophyllene (1.2%) and β-selinene (1.0%). Interestingly, however, only one of the nine, namely caryophyllene, could be detected in the celeriac isolate (and at only 0.1%). The notable absence (or low level) of β-selinene, in particular, is another distinguishing facet, since it is a well-known and reputedly important aroma component of celery [5, 35].

Of greater relevance is the identification in this work of 16 phthalides (one is a derivative), 12 of which were positively characterized, and are presented in Fig. 1. Phthalide structures and nomenclature in the literature have been ambiguous and highly confused. In this paper, the semi-systematic nomenclature used is based on that of Cocker *et al.* [36] and is explained in the figure footnote. As a class, the phthalides represented as high a proportion as 42.3% of the celery isolate and 37.4% of the celeriac sample. Twelve members were present in both celery and celeriac, but two of the celery phthalides, namely (*E*)-ligustilide and cnidilide, could not be detected in the celeriac volatiles. To the best of our knowledge, this is the first reported presence of cnidilide and methyl sedanonate in celery, and of the latter compound and the two isomeric 3-butylhexahydrophthalides in celeriac. The major phthalide in both isolates was sedanenolide, constituting 28.1% of the celery volatiles and 13.4% of the celeriac sample. The other main differences between the two isolates were the presence of higher concentrations in celery of 3-butylphthalide (2.5 vs 1.5%), (*E*)-3-butylidinephthalide (1.0 vs 0.2%), (*E*)-ligustilide and cnidilide, whereas the celeriac isolate contained higher concentrations of the two isomeric 3-butylhexahydrophthalides (5.4 vs 0.1% combined), *cis*-sedanolide (5.3 vs 5.0%), *trans*-sedanolide (7.3 vs 1.4%), (*Z*)-3-butylidinephthalide (0.5 vs 0.1%) and methyl sedanonate (0.2% vs trace).

As shown in Table 1, the highly characteristic odour of celery was associated only with the phthalides during chromatographic elution. All the representatives identified were described as celery-like and all are substituted at C_3. However, a braised or cooked celery note was associated with one isomer of 3-butylhexahydrophthalide, (*E*)-3-butylidinephthalide and sedanenolide; a medicinal celery nuance correlated with *cis*-sedanolide and (*Z*)-ligustilide, whilst a distinct camphoraceous/celery description applied to the alternative isomers, i.e. *trans*-sedanolide and (*E*)-ligustilide. The correlation of celery odour with phthalide structures dates back to the late 1920's when Berlingozzi and co-workers studied the chemistry and odour properties of alkyl and alkylidene phthalides, e.g. ref. [37]. Working with synthesized 5,6-dihydro-, 3a,4,5,6-tetrahydro- and hexahydrophthalides, they showed that replacement of one of the C_3 hydrogens by an alkyl group produced a celery aroma. When both were replaced by an alkyl group, the odour was less intense. However, celery odour was most intense when the C_3 hydrogens were replaced by an alkylidene group; odour intensity increased as the alkylidene group increased from C_1 to C_4. This was confirmed later by Kariyone and Shimizu [38].

Indeed, four alkylidenephthalides, i.e. 3-(2-methylpropylidene)phthalide, 3-(3-methylbutylidene)phthalide, 3-(2-methylpropylidine)-3a,4-dihydrophthalide and 3-(3-methylbutylidine)-3a,4-dihydrophthalide, were detected in celery stem volatiles by Gold and Wilson in 1963 [39, 40] and found to possess significant celery-like aroma. Since then, the celery odour of such compounds has been confirmed [41] and some representatives of this group have been reported four times as celery volatiles [6, 12, 14, 30], but characterization has not been convincing. Specific mass spectral searches were made in the present study for these four compounds, but none was detected.

Some previous workers have been unable to detect 3-butylphthalide together with sedanolide in celery, and have suggested that the latter has been confused with the former [4, 33]. However, both (and also the isomeric sedanolides) were very clearly separated and readily characterized in this investigation. A confusion has also

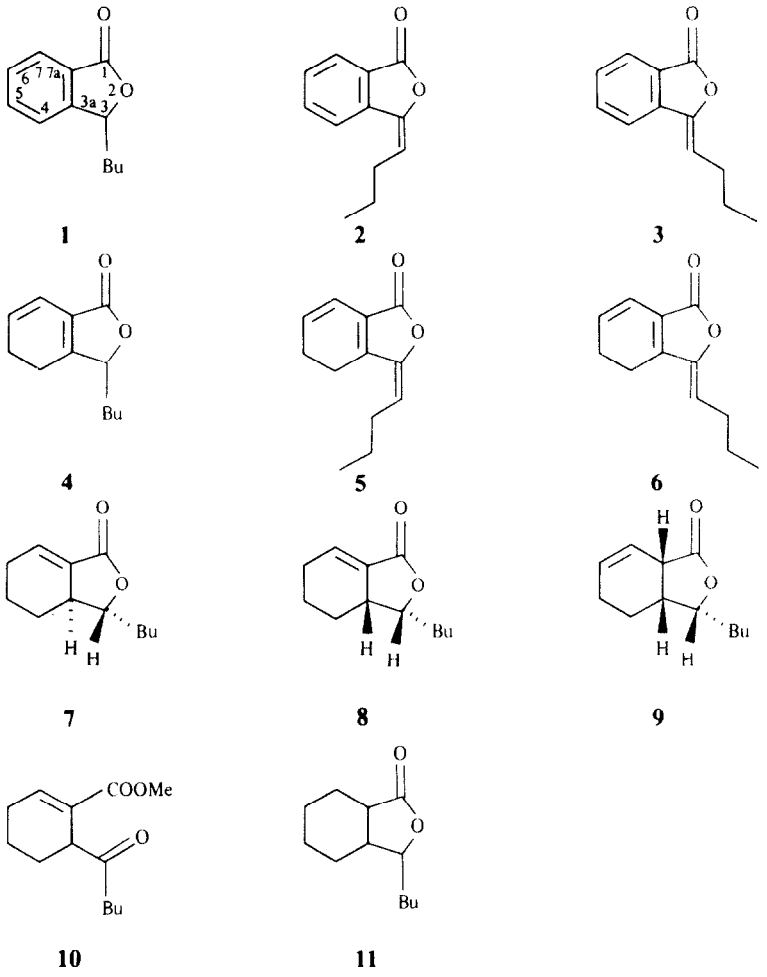

Fig. 1. Bu is the straight chain butyl group. The stereochemical convention suggested by Cocker *et al.* [36] is used here, i.e. *cis* and *trans* used to describe orientation of 3a,7a H atoms; *syn* and *anti* used to describe orientation of 3,3a H atoms; where a 7,7a double bond eliminates the necessity to refer to the ring fusion, *cis* and *trans* are used to describe orientation of the 3,3a H atoms. **1** = 3-butylphthalide; **2** = (*E*)-3-butylidenephthalide; **3** = (*Z*)-3-butylidenephthalide; **4** = 3-butyl-4,5-dihydrophthalide or sedanenolide (also named senkyunolide) [34]; **5** = (*E*)-3-butylidene-4,5-dihydrophthalide or (*E*)-ligustilide; **6** = (*Z*)-3-butylidene-4,5-dihydrophthalide or (*Z*)-ligustilide; **7** = *trans*-3-butyl-3a,4,5,6-tetrahydrophthalide or *trans*-sedanolide (also named neocnidilide) [36]; **8** = *cis*-3-butyl-3a,4,5,6-tetrahydrophthalide or *cis*-sedanolide (also named isocnidilide) [36]; **9** = *cis,syn*-3-butyl-3a,4,5,7a-tetrahydrophthalide or cnidilide [36]; **10** = methyl 6-pentanoylcyclohex-1-enoate or methyl sedanonate [33]; **11** = 3-butylhexahydrophthalide (four possible isomers *cis,anti*; *trans, anti*; *cis, syn*; *trans, syn* [36]–of which two detected).

occurred in the literature regarding the identification of sedanolide and sedanenolide, in that frequently either one or the other has been reported but not both. The reason for this is that they can have similar chromatographic properties and can prove difficult to resolve. Once again, good resolution of the sedanenolide and the isomeric sedanolides was achieved in the present study, especially when using the DB5 fused silica capillary GC column. Very recently, adequate resolution for characterisation has also been achieved by Bindler and Laugel, analysing the volatiles of celeriac brandy [42], and to a lesser extent also by Toulemonde *et al.* in their analysis of lovage phthalides, although they report only one sedanolide isomer [25].

Three compounds, which are probably related biosynthetically and/or degradatively to the phthalides, eluted on the DB5 GC column in between the monoterpene and sesquiterpene hydrocarbon groups. Two were present in both celery and celeriac and are the previously reported [30, 35] pentylcyclohexadiene (M_r 150) and pentylbenzene (M_r 148), the latter being formed from the former by disproportionation or dehydrogenation [35]. The former was present at a higher concentration in celeriac than celery (1.8% vs 0.3%), agreeing with the recent findings for lovage 'root' [43]. The third compound was detected only in the celery isolate and had a mass spectrum showing similarities with that of the pentylcyclohexadiene; it was identified as (*E,Z*)-undeca-1,3,5-triene, reported recently as a celery volatile by Berger *et al.* [22]. It can be regarded as the ring-opened form of the pentylcyclohexadiene. However, linear undecaenes are more potent odorants than their alicyclic

isomers [22], the detection odour threshold of (*E,Z*)-undeca-1,3,5-triene being 0.001–0.002 ng, the lowest in air for any hydrocarbon [44].

The identified series of four *p*-menthatrienes (including *p*-mentha-1,3,8-triene) has not been established previously in the volatile components of celery, and they could not be detected in the celeriac isolate. Together with the closely related 4-isopropenyl-1-methylbenzene, they have been reported as volatiles of parsley [29, 45], another member of the Umbelliferae. However, two other volatile components, which are to some extent characteristic of the Umbelliferae and identified previously in both parsley [29, 45] and celery [11], namely apiole and myristicin, could not be detected in the present study.

EXPERIMENTAL

Fresh celery (*Apium graveolens* L. var. *dulce*, 'Celebrity') and celeriac (*Apium graveolens* L. var. *rapaceum*, 'Monarch') were purchased from a local supermarket supplied by G. S. Shropshire and Sons, Cambs.

Isolation of volatile components. Celery stems (750 g), cut in *ca* 1 cm slices, in dist. H_2O (500 ml) were extracted for 3 hr in a modified [16] Likens and Nickerson apparatus [17] using triply distilled 2-methylbutane (20 ml). Four extractions were performed and the extracts combined before low temp.–high vacuum concentration [18] to 200 μl at 2.6×10^{-3} kPa. The procedure was repeated using peeled celeriac cut in *ca* 1 cm cubes. A blank isolate was obtained as above, using dist. H_2O only in the extraction flask.

GC. FID-GC: (a) 60 m × 0.32 mm i.d. fused silica capillary column coated with DB5 bonded phase (1 μm film); He, 2 ml/min; temp. prog., 40° for 5 min then 2°/min to 225°; detector and injector heaters, 250°; injection vol., typically 1 μl at 25:1 split and attenuation 1×64, i.e. 32×10^{-11} A fsd. (b) 25 m × 0.32 mm i.d. fused silica capillary column coated with BP20 bonded phase (0.5 μm film); He, 2 ml/min; temp. prog., 60° for 10 min then 2°/min to 200°; detector and injector heaters, 225°; other parameters as for the DB5 column.

GC odour port assessment (GC-OPA). Isolates were assessed sensorially at the DB5 column outlet after chromatographic separation using the technique of GC-OPA as previously described [19].

GC-MS. A Kratos MS25 instrument was used, linked on-line to a Kratos DS55S data processing system. Capillary GC conditions as above were used, for both polar and non-polar columns. Injection vol. was 2 μl at 25:1 split. Significant operating parameters of the MS were: ionization voltage, 70 eV; ionization current, 100 μA; source temp., 200°; accelerating voltage, 2 kV; resolution, 600; scan speed, 1 sec/decade (repetitive throughout run).

Quantitative assessment. Duplicate analyses were performed. Quantitative data were derived from the TIC monitor obtained during GC-MS and, for trace components, by extrapolation from integrator (Hewlett Packard 3370B) data obtained from the GC-FID chromatogram recorded during routine GC.

Acknowledgements—We thank Mr W. G. Gunn and Mr A. E. Cakebread for running the GC-MS, and Mrs J. Liddell for technical assistance.

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