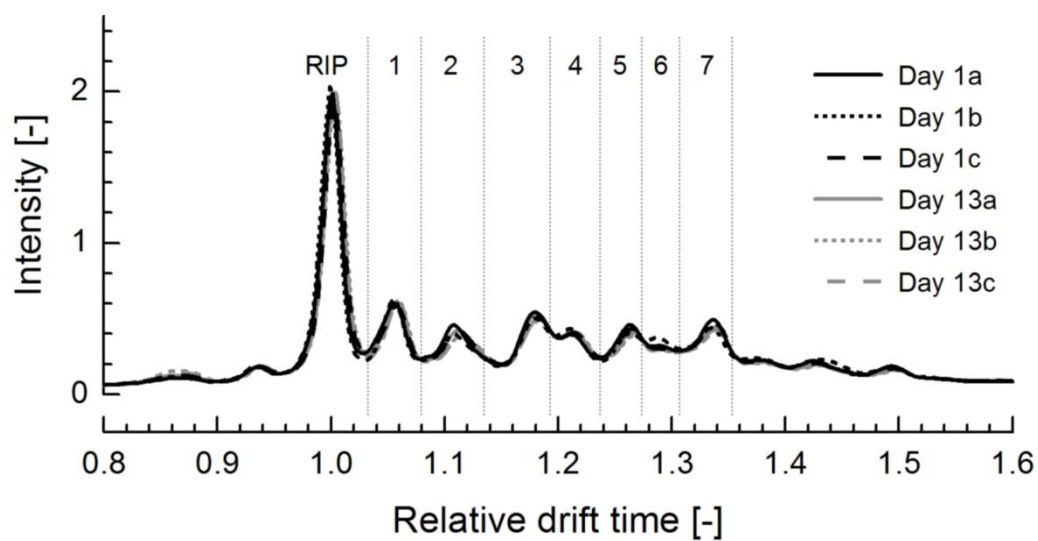


Figure S1. IMS sample inlet system. From M. Tzschoppe et al., Using ion mobility spectrometry for screening the autoxidation of peanuts, *Food Control* 2016, 64, 17–21, with permission from Elsevier.



	Peak 1	Peak 2	Peak 3	Peak 4	Peak 5	Peak 6	Peak 7
Peak area [$10^{-2} \cdot \text{A.U.}$]	0.76 ± 0.04	0.57 ± 0.04	0.87 ± 0.07	0.45 ± 0.06	0.48 ± 0.06	0.17 ± 0.04	0.68 ± 0.06
RSD [%]	5.7	6.3	8.5	13.3	11.7	23.5	8.5

Figure S2. IM spectra of cocoa liquor (used for testing purposes) that was measured repeatedly on several days. Peak areas in table are mean value \pm standard deviation ($n = 6$) of the seven peaks in IM spectra.

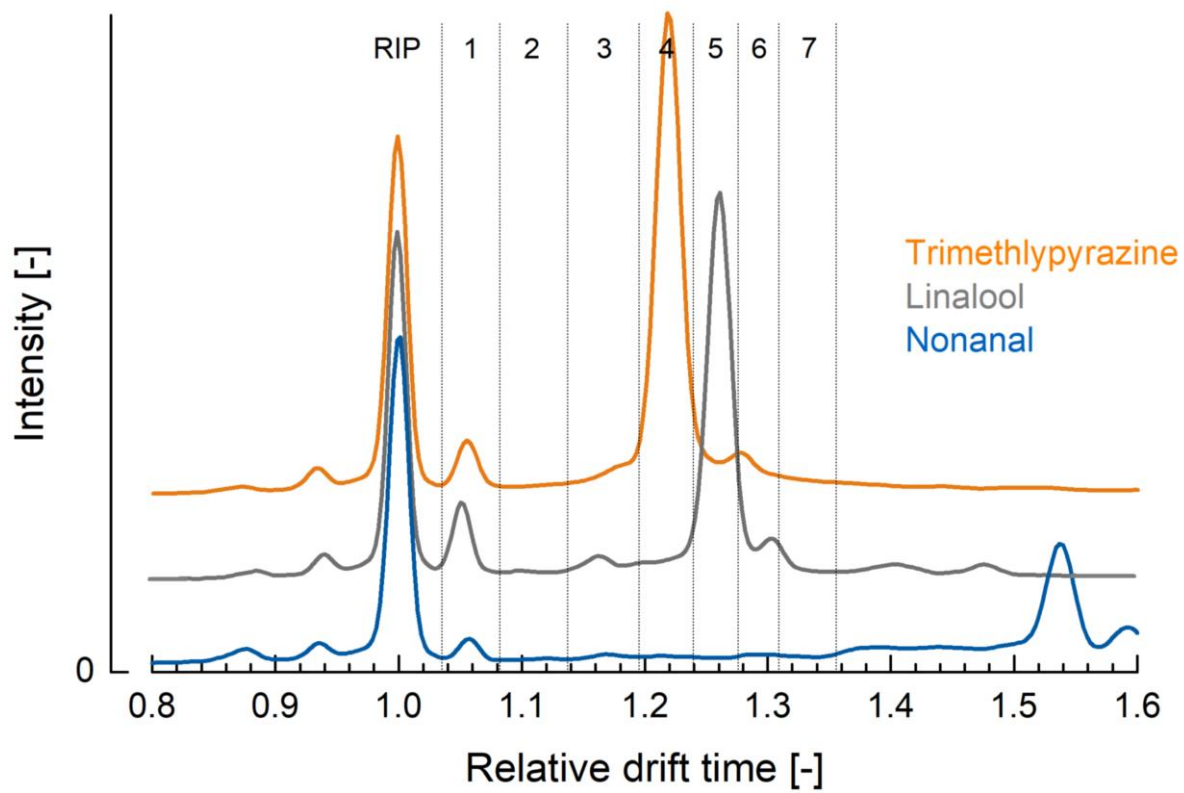


Figure S3. IM spectra of pure flavor compounds usually found on cocoa and chocolate. Intensity axis: RIP intensity for each compound is 2.0.

Table S1. Absolute peak areas of flavor compounds determined by HS-SPME-GC-MS (n = 2).

Compound	Absolute peak area [$10^4 \cdot \text{A.U.}$]				
	Fermented cocoa beans	Un-roasted cocoa nibs	Roasted cocoa nibs	Preground cocoa liquor	Finely ground, pre-treated cocoa liquor
Acetoin	14.6	21.1	-	-	-
3-Methylbutan-1-ol	44.5	12.6	-	-	-
2-Methylpropanoic acid	42.2	-	-	-	-
2,3-Butanediol*	28.5	22.5	305.0	137.4	76.1
Pentyl acetate	43.2	40.0	-	-	-
1-Butanol-3-methyl-acetate*	64.1	61.3	37.7	89.1	-
1-Butanol-2-methyl-acetate	17.9	27.8	-	-	-
2-Heptanol	46.5	17.8	-	-	-
2,5-Dimethylpyrazine	-	-	36.6	26.0	12.4
2,3-Dimethylpyrazine	-	-	26.5	19.3	14.7
Benzaldehyde*	34.2	100.6	40.1	41.2	36.2
Beta-myrcene	39.6	17.6	8.7	9.0	6.9
2-Ethyl-6-methylpyrazine	-	-	3.1	3.1	2.9
Trimethylpyrazine*	8.8	19.5	79.0	59.9	55.1
Limonene	3.7	6.1	3.4	4.4	4.6
Phenylacetaldehyde	-	-	9.4	18.8	20.6
2-Isobutyl-4,5-dimethyl-3-oxazoline	-	-	78.3	-	-
1-(1H-Pyrrol-2-yl)ethanone	-	-	11.8	20.5	21.1
Acetophenone*	23.0	21.0	20.1	21.4	16.0
3-Ethyl-2,5-dimethylpyrazine	-	-	13.9	16.0	16.7
Tetramethylpyrazine*	77.4	181.2	281.2	276.8	313.9
2-Nonanone	15.0	29.6	17.7	21.1	19.5
Linalool*	21.6	35.1	20.7	25.9	26.7
Nonanal*	16.8	6.8	-	2.9	9.2
2-Isopropyl-5-methyl-2-hexenal	-	-	78.6	13.8	-
2-Phenylethanol*	71.1	87.8	181.5	257.3	169.1
Massoia lactone	-	-	56.0	127.7	65.1
2-Ethyl-3,5,6-trimethylpyrazine	-	4.5	11.7	11.1	12.6
Ethyl octanoate	7.4	4.2	5.0	7.9	6.9
2-Phenethyl acetate*	29.1	56.2	42.5	64.3	65.8
5,6-Dihydro-2H-pyran-2-one	-	-	-	11.0	19.2
2-Phenyl-2-butenal	-	8.1	11.0	21.4	20.9
Amyl benzoate	10.0	12.0	8.7	14.4	15.7
Caffeine	-	-	-	14.7	10.8

* Marked compounds are the selected volatiles for correlation analysis.