Supplementary Materials: Development of a Refrigerant-Free Cryotrap Unit for Pre-Concentration of Biogenic Volatile Organic Compounds in Air

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Table S1. List the chemical properties of target species for ambient measurements.

Compounds	CAS	Formula	Molar mass	Purity (%)	Producer
Isoprene	78-79-5	C_5H_8	68.1	100	Linder, USA
α-pinene	80-56-8	$C_{10}H_{16}$	136.2	96.7	Dr. Ehrenstorfer, Germany
camphene	79-92-5	$C_{10}H_{16}$	136.2	95.0	Supelco, USA
sabinene	3387-41-5	$C_{10}H_{16}$	136.2	97.9	ChromaDex, USA
β-Myrcene	123-35-3	$C_{10}H_{16}$	136.2	95.0	ChromaDex, USA
β-pinene	127-91-3	$C_{10}H_{16}$	136.2	98.8	Dr. Ehrenstorfer, Germany
2-carene	4497-92-1	$C_{10}H_{16}$	136.2	96.6	ChromaDex, USA
3-carene	13466-78-9	$C_{10}H_{16}$	136.2	92.6	Dr. Ehrenstorfer, Germany
D-limonene	5989-27-5	$C_{10}H_{16}$	136.2	92.3	ChromaDex, USA
p-Cymene	99-87-6	$C_{10}H_{14}$	134.2	99.1	Dr. Ehrenstorfer, Germany
Ocimene	13877-91-3	$C_{10}H_{16}$	136.2	98.0	Supelco, USA
Cineole	470-82-6	$C_{10}H_{18}O$	154.3	98.7	ANPLE, China
γ-terpinene	99-85-4	$C_{10}H_{16}$	136.2	94.5	Dr. Ehrenstorfer, Germany
Isolongifolene	1135-66-6	$C_{15}H_{24}$	204.4	98.0	Supelco, USA
Longifolene	475-20-7	$C_{15}H_{24}$	204.4	81.1	ChromaDex, USA
α-cedrene	469-61-4	$C_{15}H_{24}$	204.4	94.7	ChromaDex, USA
β-caryophyllene	87-44-5	$C_{15}H_{24}$	204.4	98.0	YuanyeBio, China
α-caryophyllene	6753-98-6	$C_{15}H_{24}$	204.4	96.7	ChromaDex, USA

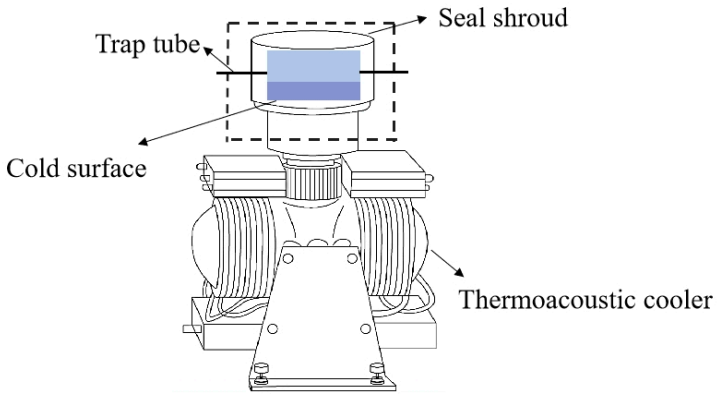

Figure S1. A drawing of thermoacoustic cooler which is used in this device.

Table S2. Detection limit (DL), repeatability and their mixing ratios (mean concentration ± standard deviation) (ppbv) of measured VOC compounds in the air of Guangzhou sie at June 2022.

Species	R^2	DL (ppbv)	Repeatability	AVG
Alkanes (*n*=29)				
Ethane	0.999	0.036	1.8%	1.67 ± 0.862
Propane	0.999	0.016	1.0%	3.58 ± 2.43
iso-Butane	0.999	0.012	0.8%	2.08 ± 2.36
n-Butane	0.999	0.020	0.4%	3.33 ± 2.28
Cyclopentane	0.999	0.008	0.2%	0.236 ± 0.181
iso-Pentane	0.999	0.012	0.1%	5.48 ± 5.44
n-Pentane	0.999	0.010	1.3%	5.40 ± 6.31
2,2-Dimethylbutane	0.996	0.010	2.0%	0.228 ± 0.247
2,3-Dimethylbutane	0.999	0.015	1.3%	0.192 ± 1.069
2-Methylpentane	0.998	0.012	1.6%	0.807 ± 0.771
3-Methylpentane	0.998	0.012	2.4%	0.790 ± 0.696
n-Hexane	0.997	0.008	1.4%	1.05 ± 1.04
2,4-Dimethylpentane	0.999	0.009	1.4%	0.123 ± 0.105
Methylcyclopentane	0.997	0.008	1.9%	0.439 ± 0.376
2-Methylhexane	0.998	0.012	1.3%	0.553 ± 0.465
2,3-Dimethylpentane	0.999	0.013	1.8%	0.153 ± 0.129
Cyclohexane	0.999	0.011	1.8%	0.133 ± 0.102
3-Methylhexane	0.998	0.009	1.1%	0.362 ± 0.310
2,2,4-Trimethylpentane	0.998	0.003	1.6%	0.135 ± 0.119
n-Heptane	0.998	0.009	1.9%	0.250 ± 0.196
Methylcyclohexane	0.997	0.013	2.4%	0.142 ± 0.113
2,3,4-Trimethylpentane	0.998	0.010	1.3%	0.044 ± 0.032
2-Methylheptane	0.999	0.010	1.6%	0.038 ± 0.030
3-Methylheptane	0.999	0.009	1.5%	0.043 ± 0.033
n-Octane	0.999	0.009	1.6%	0.084 ± 0.052
Nonane	0.997	0.011	1.8%	0.087 ± 0.043
n-Decane	0.998	0.009	1.5%	0.040 ± 0.020
n-Undecane	0.995	0.011	1.7%	0.057 ± 0.028
n-Dodecane	0.988	0.021	5.5%	0.000±0.000
Alkenes (*n*=11)				
Ethylene	0.999	0.039	1.2%	1.52 ± 0.780
Propylene	0.997	0.018	2.8%	0.222 ± 0.171
trans-2-Butene	0.998	0.009	1.7%	0.313 ± 0.593
1-Butene	0.998	0.020	1.6%	0.161 ± 0.095
cis-2-Butene	0.998	0.018	1.7%	0.071 ± 0.229
1,3-Butadiene	0.998	0.014	1.4%	0.026 ± 0.029
Isbutene	0.995	0.012	2.1%	0.650 ± 0.455
1-Pentene	0.998	0.019	1.4%	0.059 ± 0.102
trans-2-Pentene	0.999	0.009	1.6%	0.077 ± 0.104

cis-2-Pentene	0.997	0.006	3.9%	0.021 ± 0.028
1-Hexene	0.998	0.013	1.8%	0.090 ± 0.053
		Alkynes (*n*=1)		
Acetylene	0.997	0.039	3.5%	1.01 ± 0.737
		Carbonyls (*n*=12)		
Acrolein	0.988	0.022	6.3%	0.076 ± 0.072
Propanal	0.998	0.030	5.4%	0.257 ± 0.183
Acetone	0.998	0.013	1.5%	0.154 ± 0.140
MTBE	0.999	0.010	4.9%	0.710 ± 0.975
Methacrolein	0.997	0.009	3.4%	0.055 ± 0.050
n-Butanal	0.999	0.012	8.6%	0.159 ± 0.106
MethylVinylKetone	0.995	0.010	6.5%	0.203 ± 0.163
MethylEthylKetone	0.997	0.011	5.5%	0.179 ± 0.119
2-Pentanone	0.988	0.025	9.2%	0.000 ± 0.000
Pentanal	0.956	0.022	10.3%	0.123 ± 0.114
3-Pentanone	0.957	0.032	13.9%	0.000 ± 0.000
Hexanal	0.950	0.016	14.7%	1.30 ± 0.832
		Aromatics (*n*=16)		
Benzene	0.998	0.007	1.3%	0.303 ± 0.223
Toluene	0.999	0.006	1.4%	2.73 ± 2.36
Ethylbenzene	0.999	0.009	1.7%	0.652 ± 0.531
m/p-Xylene	0.998	0.008	1.5%	1.99 ± 2.83
o-Xylene	0.998	0.007	1.3%	0.852 ± 0.734
Styrene	0.998	0.008	1.6%	0.343 ± 0.328
Isopropylbenzene	0.998	0.007	1.6%	0.030 ± 0.018
n-Propylbenzene	0.999	0.009	1.8%	0.027 ± 0.016
m-Ethyltoluene	0.999	0.015	1.5%	0.053 ± 0.039
p-Ethyltoluene	0.993	0.014	0.5%	0.033 ± 0.022
1,3,5-Trimethylbenzene	0.999	0.020	0.8%	0.027 ± 0.020
o-Ethyltoluene	0.999	0.010	0.9%	0.033 ± 0.022
1,2,4-Trimethylbenzene	0.993	0.029	0.8%	0.107 ± 0.077
1,2,3-Trimethylbenzene	0.995	0.012	0.7%	0.039 ± 0.027
m-Diethylbenzene	0.998	0.060	1.0%	0.013 ± 0.008
p-Diethylbenzene	0.982	0.050	7.2%	0.030 ± 0.019
		Haloalkane (*n*=31)		
Freon-114	0.998	0.025	0.8%	0.021 ± 0.005
Chloromethane	0.999	0.020	2.0%	0.160 ± 0.187
Vinylchloride	0.999	0.023	1.0%	0.170 ± 0.048
Bromomethane	0.999	0.019	1.0%	0.004 ± 0.002
Chloroethane	0.998	0.020	1.1%	0.002 ± 0.004
Freon-11	0.976	0.015	4.3%	0.362 ± 0.054
1,1-Dichloroethene	0.999	0.017	1.0%	0.001 ± 0.005
Freon-113	0.998	0.009	1.2%	0.069 ± 0.013
Dichloromethane	0.996	0.026	1.2%	1.97±1.61
1,1-Dichloroethane	0.999	0.024	1.0%	0.004 ± 0.004
cis-1,2-Dichloroethene	0.999	0.009	0.8%	0.005 ± 0.003
Chloroform	0.958	0.026	5.1%	0.060 ± 0.033
1,1,1-Trichloroethane	0.988	0.015	3.5%	0.002 ± 0.002
CarbonTetrachloride	0.999	0.010	4.9%	0.074 ± 0.018
1,2-Dichloroethane	0.997	0.031	1.0%	0.115 ± 0.095
Trichloroethylene	0.999	0.024	1.3%	0.119 ± 0.167
1,2-Dichloropropane	0.998	0.016	1.6%	0.081 ± 0.073
Bromodichloromethane	0.995	0.023	2.0%	0.002 ± 0.003
trans-1,3-Dichloropropene	0.997	0.033	5.8%	0.025 ± 0.011
cis-1,3-Dichloropropene	0.998	0.034	2.0%	0.001 ± 0.003
1,1,2-Trichloroethane	0.998	0.015	1.5%	0.014 ± 0.011
Tetrachloroethylene	0.998	0.031	1.4%	0.054 ± 0.045
1,2-Dibromoethane	0.998	0.028	2.0%	0.001 ± 0.004
Chlorobenzene	0.995	0.033	1.9%	0.003 ± 0.003
Bromoform	0.991	0.030	2.1%	0.004 ± 0.005

1,1,2,2-Tertrachloroethane	0.991	0.021	2.0%	0.002 ± 0.003
1,3-Dichlorobenzene	0.995	0.021	2.0%	0.003 ± 0.003
1,4-Dichlorobenzene	0.997	0.011	1.9%	0.024 ± 0.019
BenzylChloride	0.993	0.038	5.9%	0.001 ± 0.001
1,2-Dichlorobenzene	0.995	0.017	2.0%	0.003 ± 0.004
Acetonitrile	0.993	0.043	4.5%	0.326 ± 0.320
		BVOCs (*n*=4)		
Isoprene	0.998	0.007	1.6%	1.15 ± 0.511
α-pinene	0.994	0.004	3.8%	0.105 ± 0.048
β-pinene	0.994	0.004	3.7%	0.016 ± 0.006
D-limonene	0.993	0.005	5.3%	0.013 ± 0.007

Table S3. The optimization results water trap temperature, desorption temperature and desorption time of target analytes. The number of repetitions of each analysis was 3 times.

The optimization results of water trap temperature of target analytes			
Compound	0 ℃ (Average[a] ±SD[b])	-20 ℃ (Average ±SD)	-40 ℃ (Average ±SD)
α-Pinene	4.22 ± 0.012	4.47 ± 0.018	4.63 ± 0.012
Camphene	3.37 ± 0.009	3.61 ± 0.019	3.73 ± 0.009
Sabinene	4.41 ± 0.016	4.58 ± 0.010	4.77 ± 0.016
β-Myrcene	4.40 ± 0.016	4.58 ± 0.010	4.76 ± 0.016
β-Pinene	4.40 ± 0.016	4.58 ± 0.010	4.77 ± 0.016
2-Carene	4.48 ± 0.007	4.71 ± 0.013	4.89 ± 0.007
3-Carene	4.63 ± 0.010	4.92 ± 0.006	5.12 ± 0.010
D-limonene	4.24 ± 0.025	4.34 ± 0.031	4.57 ± 0.025
p-Cymene	5.33 ± 0.013	5.49 ± 0.004	5.51 ± 0.013
Ocimene	4.40 ± 0.016	4.52 ± 0.003	4.59 ± 0.016
Cineole	4.84 ± 0.074	4.57 ± 0.021	4.48 ± 0.074
γ-Terpinene	4.69 ± 0.016	4.85 ± 0.006	5.00 ± 0.016
Isolongifolene	4.67 ± 0.042	4.73 ± 0.059	4.12 ± 0.042
Longifolene	4.55 ± 0.010	4.30 ± 0.079	3.60 ± 0.010
α-Cedrene	4.86 ± 0.039	4.62 ± 0.082	3.92 ± 0.039
β-Caryophyllene	4.32 ± 0.035	3.81 ± 0.093	2.97 ± 0.035
α-Caryophyllene	4.59 ± 0.086	3.65 ± 0.106	2.90 ± 0.086

The optimization results of desorption temperature of target analytes			
Compound	150 ℃ (Average ±SD)	200 ℃ (Average ±SD)	250 ℃ (Average ±SD)
α-Pinene	4.69 ± 0.030	4.84 ± 0.070	4.81 ± 0.042
Camphene	3.76 ± 0.030	3.87 ± 0.051	3.85 ± 0.037
Sabinene	4.83 ± 0.022	5.01 ± 0.061	4.98 ± 0.040
β-Myrcene	4.83 ± 0.021	5.01 ± 0.061	4.98 ± 0.040

β-Pinene	4.83 ± 0.022	5.01 ± 0.061	4.98 ± 0.040
2-Carene	4.92 ± 0.018	5.08 ± 0.058	5.05 ± 0.033
3-Carene	5.16 ± 0.017	5.32 ± 0.056	5.30 ± 0.032
D-limonene	4.58 ± 0.011	4.69 ± 0.073	4.72 ± 0.052
p-Cymene	5.74 ± 0.012	5.90 ± 0.048	5.89 ± 0.028
Ocimene	4.74 ± 0.016	4.90 ± 0.055	4.87 ± 0.032
Cineole	4.76 ± 0.016	4.89 ± 0.008	5.04 ± 0.043
γ-Terpinene	5.09 ± 0.013	5.26 ± 0.057	5.26 ± 0.031
Isolongifolene	4.98 ± 0.013	5.12 ± 0.022	5.16 ± 0.024
Longifolene	4.57 ± 0.019	4.71 ± 0.047	4.78 ± 0.045
α-Cedrene	4.92 ± 0.019	5.09 ± 0.043	5.18 ± 0.048
β-Caryophyllene	4.08 ± 0.036	4.27 ± 0.060	4.42 ± 0.076
α-Caryophyllene	3.97 ± 0.047	4.23 ± 0.032	4.38 ± 0.139

The optimization results of desorption time of target analytes						
Compound	**1 min (Average ±SD)**	**2 min (Average ±SD)**	**3 min (Average ±SD)**	**4 min (Average ±SD)**	**5 min (Average ±SD)**	7 min (Average ±SD)
α-Pinene	4.88 ± 0.031	4.99 ± 0.031	5.01 ± 0.031	4.99 ± 0.031	4.94 ± 0.031	4.86 ± 0.031
Camphene	3.94 ± 0.026	4.08 ± 0.026	4.09 ± 0.026	4.06 ± 0.026	3.99 ± 0.026	3.88 ± 0.026
Sabinene	4.80 ± 0.067	5.10 ± 0.067	5.12 ± 0.067	5.11 ± 0.067	5.06 ± 0.067	5.01 ± 0.067
β-Myrcene	4.79 ± 0.067	5.10 ± 0.067	5.12 ± 0.067	5.11 ± 0.067	5.06 ± 0.067	5.01 ± 0.067
β-Pinene	4.79 ± 0.067	5.10 ± 0.067	5.12 ± 0.067	5.11 ± 0.067	5.06 ± 0.067	5.01 ± 0.067
2-Carene	4.92 ± 0.046	5.13 ± 0.046	5.15 ± 0.046	5.14 ± 0.046	5.11 ± 0.046	5.07 ± 0.046
3-Carene	5.13 ± 0.049	5.36 ± 0.049	5.37 ± 0.049	5.37 ± 0.049	5.34 ± 0.049	5.31 ± 0.049
D-limonene	4.36 ± 0.089	4.76 ± 0.089	4.80 ± 0.089	4.79 ± 0.089	4.77 ± 0.089	4.74 ± 0.089
p-Cymene	5.46 ± 0.106	5.88 ± 0.106	5.93 ± 0.106	5.94 ± 0.106	5.92 ± 0.106	5.91 ± 0.106
Ocimene	4.43 ± 0.112	4.92 ± 0.112	4.96 ± 0.112	4.96 ± 0.112	4.93 ± 0.112	4.90 ± 0.112
Cineole	4.12 ± 0.150	4.55 ± 0.150	4.84 ± 0.150	4.95 ± 0.150	5.15 ± 0.150	5.25 ± 0.150
γ-Terpinene	4.88 ± 0.084	5.29 ± 0.084	5.32 ± 0.084	5.31 ± 0.084	5.29 ± 0.084	5.27 ± 0.084
Isolongifolene	2.47 ± 0.086	4.04 ± 0.086	4.75 ± 0.086	5.02 ± 0.086	5.18 ± 0.086	5.20 ± 0.086
Longifolene	2.30 ± 0.233	3.34 ± 0.233	4.21 ± 0.233	4.60 ± 0.233	4.83 ± 0.233	4.89 ± 0.233
α-Cedrene	2.73 ± 0.070	3.57 ± 0.070	4.53 ± 0.070	4.97 ± 0.070	5.21 ± 0.070	5.29 ± 0.070
β-Caryophyllene	1.97 ± 0.069	2.58 ± 0.069	3.60 ± 0.069	4.19 ± 0.069	4.50 ± 0.069	4.61 ± 0.069
α-Caryophyllene	2.60 ± 0.038	3.02 ± 0.038	3.36 ± 0.038	4.14 ± 0.038	4.56 ± 0.038	4.81 ± 0.038

[a] Average: The average of the Log (Area). [b] SD: Standard deviation.

Table S4. Comparisons the average mixing ratios of BVOCs measured in Guangzhou and other sites in China.

References	Guangzhou Jun. 2022	Guangzhou Jun. 2011-May. 2012	Guangzhou Aug. 29, 2020	Guangzhou* Aug. 30-31, 2020	Hongkong Jun.-Jul. 2007	Beijing Jun.-Aug. 2014	Xi'an May. 2017
	This study	**[1]**	**[2]**	**[2]**	**[3]**	**[4]**	**[5]**
Sampling site	Suburb	Forest park	Suburb	Suburb	Urban-park	Urban	Mountain
Sampling method	Online	Canister samples	Canister samples	Canister samples	Canister samples	Online	Cartridges
Enrichment method	Cryotrap-thermoacoustic cooler	Cryotrap-liquid nitrogen	Cryotrap-liquid nitrogen	Cryotrap-liquid nitrogen	Cryotrap-liquid argon	solid adsorbents	Tenax\Carbograph-solid adsorbents
Analytical method	GC-MS/FID	GC-FID	GC-MS/FID	GC-MS/FID	GC-FID	GC-FID	Offline-TD-GC-MS/FID
BVOCs	Concentrations						
Isoprene	1.15	4.90	1.21	1.75	1.33	0.927	1.30
α-pinene	0.105	0.184	0.009	0.023	-	0.018	-
β-pinene	0.016	-	0.016	0.017	0.24	-	-
D-limonene	0.013	-	-	-	0.19	-	-
Monoterpenes	0.13	0.184	0.025	0.04	0.43	0.018	0.21

* pollution.

- data were not available in the relative reference.

Table S5. Comparisons of VOCs with high levels in Guangzhou with other cities in China.

	Guangzhou	**Guangzhou**	**Guangzhou**	**Guangzhou**	**Wanqingsha**	**Beijing**	**Shanghai**	**Wuhan**
References	This study	[6]	[7]	[2]	[8]	[9]	[10]	[11]
Sampling site	Suburb	Suburb	Urban	Suburb	Suburb	Suburb	Urban	Urban
Top ten VOCs	Concentrations							
iso-Pentane	5.48	1.72	2.52	1.18	2.31	0.79	1.03	0.94
n-Pentane	5.40	1.37	1.78	0.52	1.69	0.33	0.55	0.44
Propane	3.58	4.34	3.90	3.67	4.63	1.97	2.99	5.49
n-Butane	3.33	3.07	3.62	3.82	3.86	0.81	1.57	1.74
Toluene	2.73	4.59	2.97	1.42	4.90	0.39	1.31	0.71
iso-Butane	2.08	2.67	1.82	2.10	2.22	0.47	0.91	1.09
m/p-Xylene	1.99	1.41	1.44	1.15	2.75	0.27	1.07	0.27
Dichloromethane	1.97	-	-	-	-	0.68	1.57	1.16
Ethane	1.67	3.66	1.43	1.52	2.40	1.84	3.69	3.91
Ethylene	1.52	2.99	1.21	0.61	1.56	0.69	1.53	1.97

* pollution.

- data were not available in the relative reference.

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