

## **Electronic Supplementary Information**

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### ***Radial multi-stationary phase thin-layer chromatography for the field-ready fingerprinting of herbal material***

Sarah May Sibug-Torres<sup>†</sup>, Isagani D. Padolina<sup>‡</sup>, Erwin P. Enriquez\*<sup>†</sup>

<sup>†</sup> Department of Chemistry, Ateneo de Manila University, Katipunan Ave., Loyola Heights, Quezon City, Philippines 1108

<sup>‡</sup> Pascual Pharma Corp., 23rd Floor, The Taipan Place, F. Ortigas Jr. Road, Ortigas Center, Brgy. San Antonio, Pasig City, Metro Manila, Philippines 1605

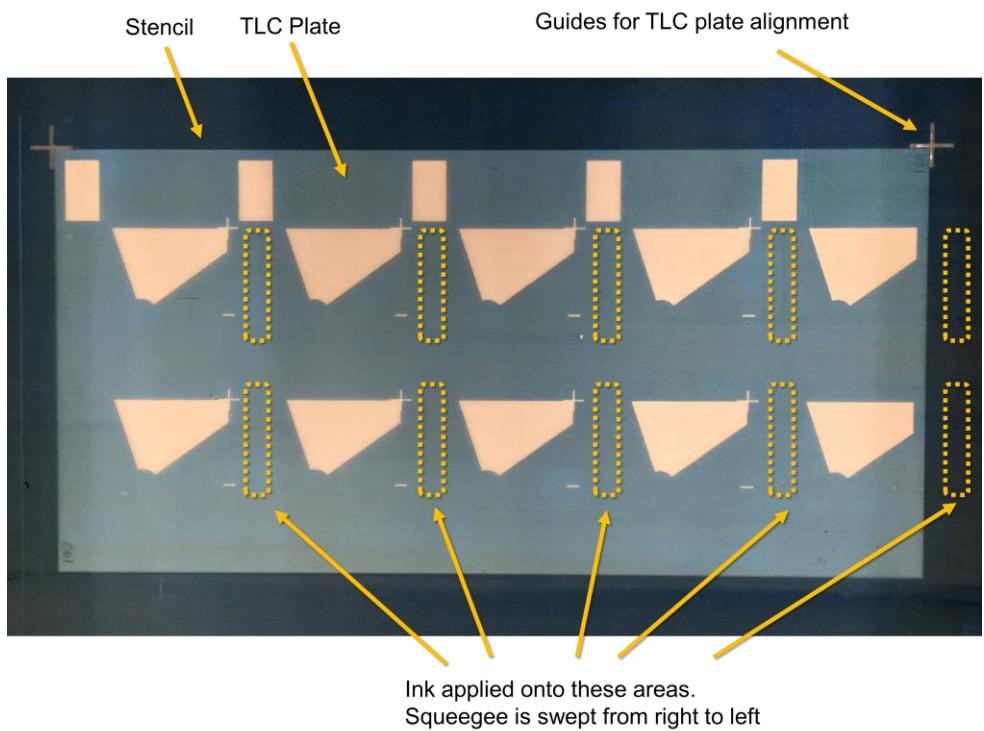
**\*Corresponding Author**

Tel: +63 2 426 6001 ext. 5620, Fax: +63 2 426 1323, Email: epenriquez@ateneo.edu

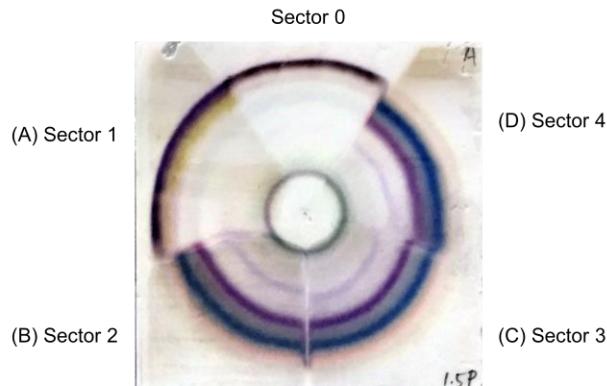
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## **Contents**

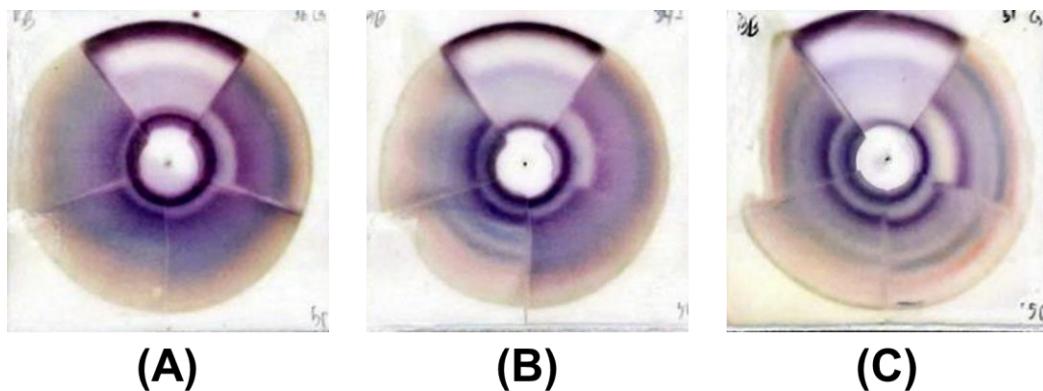
Fig. ESI-1. Illustration of the screen printing method.....	2
Fig. ESI-2. Effect of PEO-1M on screen printed MSP-TLC plates.....	3
Fig. ESI-3. Effect of screen printing ink volume.....	3
Table ESI-1. Full sample details and MSP-TLC profiles of sample set.....	1
Fig. ESI-4. Preprocessing of triplicate samples.....	9
Table ESI-2. Inner cross-validation performances of <i>B. balsamifera</i> models constructed using different combinations of MSP-TLC profile sectors.....	10
Table ESI-3. Inner cross-validation performances of <i>V. negundo</i> models constructed using different combinations of MSP-TLC profile sectors.....	11
Fig. ESI-5. Predicted classifications of <i>B. balsamifera</i> samples.....	12
Fig. ESI-6. Predicted classifications of <i>V. negundo</i> samples.....	13
Table ESI-4. Comparison of classification performances of MSP-TLC system, adapted TLC method from ref. [1], and the standard Pharmacopoeia TLC method .....	14



**Fig. ESI-1.** Illustration of the screen printing procedure used to fabricate MSP-TLC plates. A TLC plate is aligned under the screen printing stencil, and organosilane ink solution is applied onto the indicated outlines. A squeegee is swept right to left, pushing the ink through the stencil and onto the TLC plate substrate

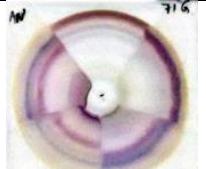
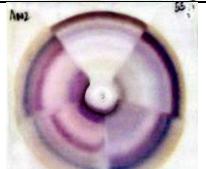
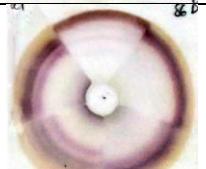
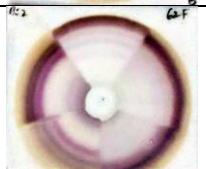
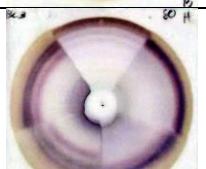
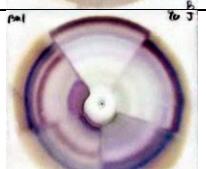
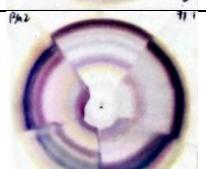
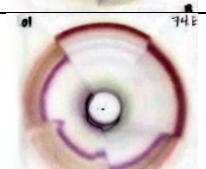


**Fig. ESI-2.** Effect of PEO-1M on screen printed MSP-TLC plates. An MSP-TLC plate was screen printed using 4:1 (v/v) ethanol-water, 5% wt. acetic acid solutions with (A) 1.00% wt. PEO-1M in Sector 1, (B) 1.50  $m$  PTMS with 1.25% wt. PEO-1M in Sector 2, (C) 1.50  $m$  PTMS with 1.00% wt. PEO-1M in Sector 3, and (D) 1.50  $m$  PTMS with 0.75% wt. PEO-1M in Sector 4. Sector 0 is bare, unmodified silica gel. A *B. balsamifera* reference sample was developed on the plate using 60:40 (v/v) ethanol-water mobile phase. The resulting MSP-TLC profile shows slight shifts in the positions of the profile bands to lower  $R_f$  values with increasing PEO-1M, suggesting a hydrophobic contribution of PEO-1M to the stationary phase.

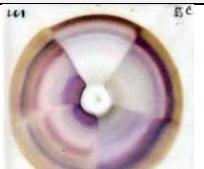
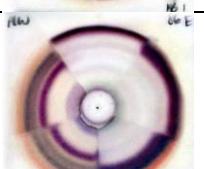
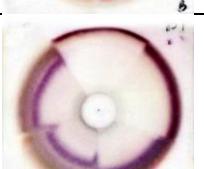
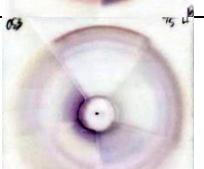
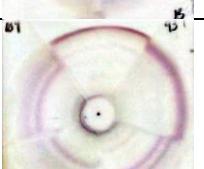


**Fig. ESI-3.** Effect of organosilane ink volume used for screen printing. Shown are MSP-TLC profiles of a *B. balsamifera* reference sample developed using 50:50 (v/v) ethanol-water on MSP-TLC plates screen printed with OTES inks at (A) 50  $\mu$ L, (B) 60  $\mu$ L, (C) 70  $\mu$ L of ink applied per sector. The smudged appearance of the TLC bands in (A) and (B) suggests that insufficient volumes of ink were applied to uniformly modify the TLC sorbent layer.

**Table ESI-1.** Full sample details and MSP-TLC profiles of sample set. Sample classes: within-specifications *B. balsamifera* (BB WS), off-specifications *B. balsamifera*, within-specifications *V. negundo* (VN WS), off-specifications *V. negundo* (OS VN), and *B. balsamifera-V. negundo* mixtures (MIX)

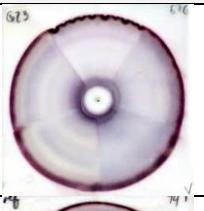
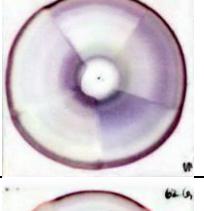
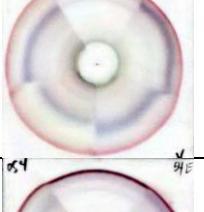
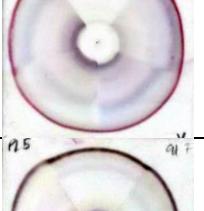
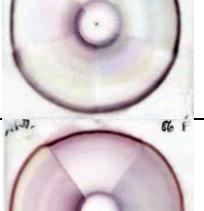
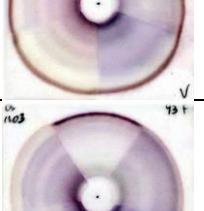
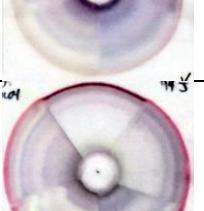
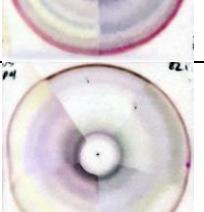
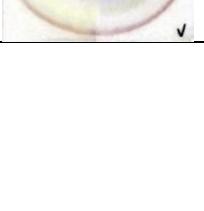
Class	ID	Source	Date Collected	Processing History	MSP-TLC Profile
BB WS	BB01	Farm 1, Luzon, Philippines	June 2017	Air Dried	
BB WS	BB02	Farm 1, Luzon, Philippines	June 2017	Oven Dried	
BB WS	BB03	Farm 2, Visayas, Philippines	April 2016	Air Dried	
BB WS	BB04	Farm 2, Visayas, Philippines	April 2016	Oven Dried	
BB WS	BB05	Farm 2, Visayas, Philippines	July 2016	Oven Dried	
BB WS	BB06	Farm 3, Luzon, Philippines	Feb 2017	Air Dried	
BB WS	BB07	Farm 3, Luzon, Philippines	Feb 2017	Oven Dried	
BB WS	BB08	Farm 4, Visayas, Philippines	June 2017	Air Dried	



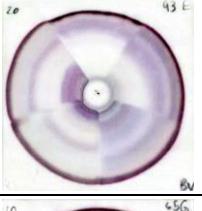
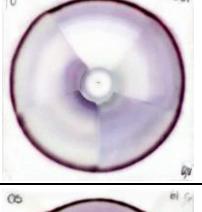
Class	ID	Source	Date Collected	Processing History	MSP-TLC Profile
BB WS	BB18	Farm 12, Luzon, Philippines	April 2017	Oven Dried	
BB WS	BB19	Farm 13, Luzon, Philippines	June 2016	Oven Dried	
BB WS	BB20	Farm 13, Luzon, Philippines	July 2016	Dehydrator	
BB WS	BB21	Farm 13, Luzon, Philippines	Nov 2017	Oven Dried	
BB WS	BB22	Farm 14, Luzon, Philippines	Nov 2017	Oven Dried	
BB OS	BB OS1	Farm 4, Visayas, Philippines	Oct 2017	Stored in humid conditions, 24 hrs	
BB OS	BB OS2	Farm 4, Visayas, Philippines	Oct 2017	Stored in humid conditions, 48 hrs	
BB OS	BB OS3	Farm 4, Visayas, Philippines	Feb 2016	Steam Sterilized	
BB OS	BB OS4	Farm 4, Visayas, Philippines	July 2016	Fermented	

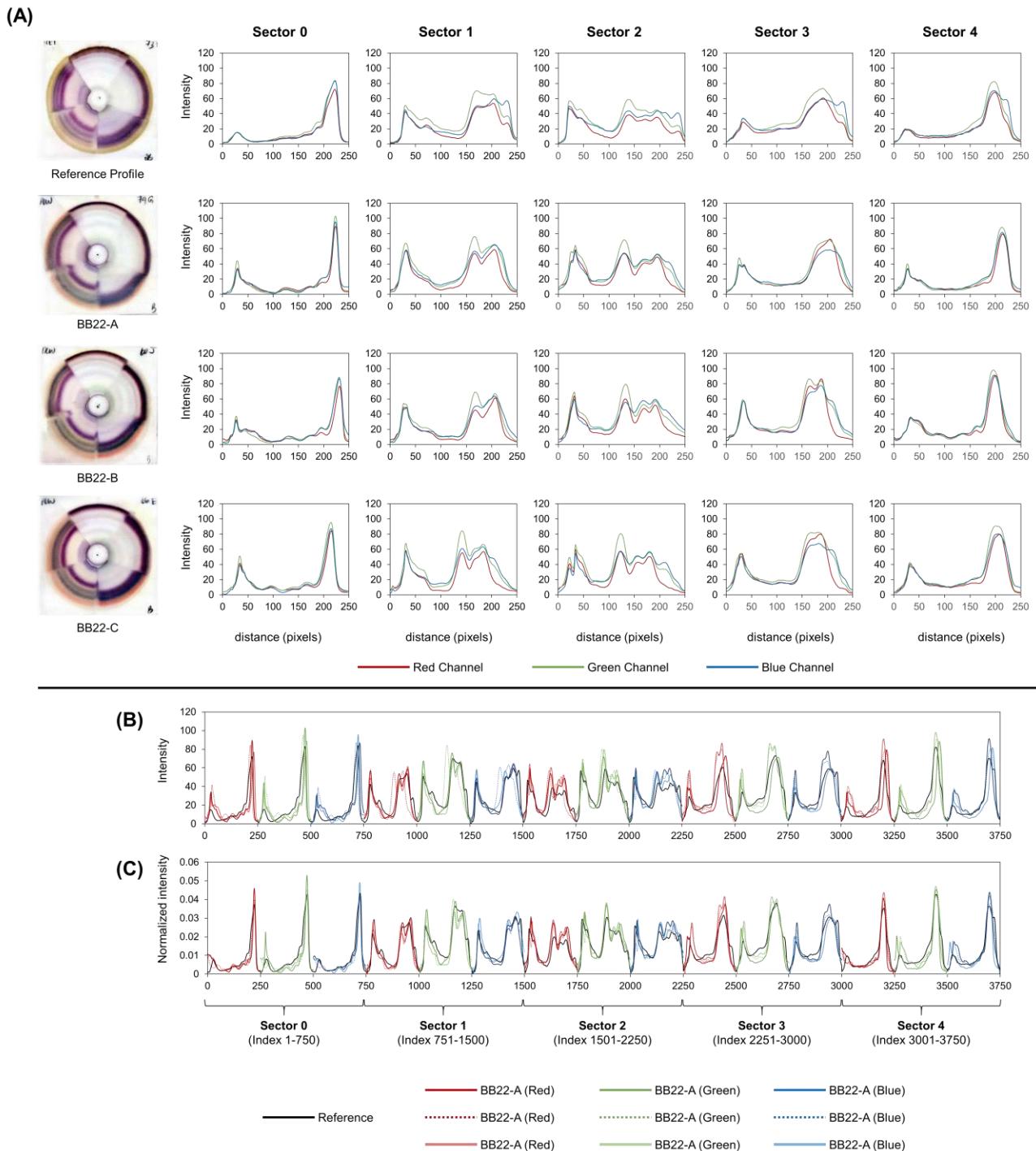
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BB OS	BB OS5	Farm 4, Visayas, Philippines	July 2016	Incomplete drying followed by storage	
BB OS	BB OS6	Farm 4, Visayas, Philippines	July 2016	High Heat Drying, 100°C	
BB OS	BB OS7	Farm 13, Visayas, Philippines	Nov 2017	Stored in humid conditions, 48 hrs	
BB OS	BB OS8	Farm 13, Visayas, Philippines	Nov 2017	High Heat Drying, 100°C	
BB OS	BB OS9	Farm 4, Visayas, Philippines	Feb 2016	Oven Dried, <i>B. lacera</i>	
VN WS	VN01	Farm 1, Luzon, Philippines	Feb 2017	Oven Dried	
VN WS	VN02	Farm 2, Luzon, Philippines	Feb 2017	Air Dried	
VN WS	VN03	Farm 2, Luzon, Philippines	Feb 2017	Oven Dried	
VN WS	VN04	Farm 3, Luzon, Philippines	May 2017	Air Dried	

Class	ID	Source	Date Collected	Processing History	MSP-TLC Profile
VN WS	VN05	Farm 4, Visayas, Philippines	June 2017	Air Dried	
VN WS	VN06	Farm 5, Visayas, Philippines	June 2017	Air Dried	
VN WS	VN07	Farm 6, Luzon, Philippines	Feb 2017	Oven Dried	
VN WS	VN08	Farm 7, Luzon, Philippines	Feb 2017	Oven Dried	
VN WS	VN09	Farm 8, Luzon, Philippines	Nov 2017	Air Dried	
VN WS	VN10	Variety A - Farm 9, Luzon, Philippines	Nov 2017	Air Dried	
VN WS	VN11	Variety B - Farm 9, Luzon, Philippines	Nov 2017	Air Dried	
VN WS	VN12	Farm 10, Luzon, Philippines	Feb 2017	Oven Dried	
VN WS	VN13	Farm 11, Luzon, Philippines	June 2017	Air Dried	

Class	ID	Source	Date Collected	Processing History	MSP-TLC Profile
VN WS	VN14	Farm 12, Luzon, Philippines	June 2017	Air Dried	
VN WS	VN15	Farm 13, Luzon, Philippines	Nov 2017	Oven Dried	
VN OS	VN OS1	Farm 2, Luzon, Philippines	Feb 2017	High Heat Drying, 100°C	
VN OS	VN OS2	Farm 2, Luzon, Philippines	Feb 2017	Stored in humid conditions	
VN OS	VN OS3	Farm 2, Luzon, Philippines	Feb 2017	Fermented	
VN OS	VN OS4	Farm 2, Luzon, Philippines	Feb 2017	Incomplete drying followed by storage	
VN OS	VN OS5	Farm 4, Visayas, Philippines	Nov 2017	Stored in humid conditions, 24 hrs	
VN OS	VN OS6	Farm 5, Visayas, Philippines	Nov 2017	Stored in humid conditions, 48 hrs	
VN OS	VN OS7	Farm 9, Luzon, Philippines	Nov 2017	Stored in humid conditions, 24 hrs	

Class	ID	Source	Date Collected	Processing History	MSP-TLC Profile
VN OS	VN OS8	Farm 9, Luzon, Philippines	Nov 2017	Stored in humid conditions, 48 hrs	
MIX	BB95: VN05	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 95% <i>B. balsamifera</i> 5% <i>V. negundo</i>	
MIX	BB90: VN10	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 90% <i>B. balsamifera</i> 10% <i>V. negundo</i>	
MIX	BB80: VN20	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 80% <i>B. balsamifera</i> 20% <i>V. negundo</i>	
MIX	BB70: VN30	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 70% <i>B. balsamifera</i> 30% <i>V. negundo</i>	
MIX	BB60: VN40	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 60% <i>B. balsamifera</i> 40% <i>V. negundo</i>	
MIX	BB50: VN50	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 50% <i>B. balsamifera</i> 50% <i>V. negundo</i>	
MIX	BB40: VN60	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 40% <i>B. balsamifera</i> 60% <i>V. negundo</i>	

Class	ID	Source	Date Collected	Processing History	MSP-TLC Profile
MIX	BB30: VN70	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 30% <i>B. balsamifera</i> 70% <i>V. negundo</i>	
MIX	BB20: VN80	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 20% <i>B. balsamifera</i> 80% <i>V. negundo</i>	
MIX	BB10: VN90	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 10% <i>B. balsamifera</i> 90% <i>V. negundo</i>	
MIX	BB05: VN95	Farm 11, Luzon, Philippines	Nov 2017	Oven Dried, 5% <i>B. balsamifera</i> 95% <i>V. negundo</i>	



**Fig. ESI-4.** Preprocessing of triplicate samples. (A) MSP-TLC profiles of *B. balsamifera* reference sample (ID: BB19) and technical replicates of sample BB22 are shown alongside their densitograms per sector. (B) Unaligned, unfolded MSP-TLC densitograms of replicate samples show mismatched peak positions. After alignment with VPdtw, the position of the peaks in each technical replicate now match, as shown in (C).

**Table ESI-2.** Inner cross-validation performances of *B. balsamifera* models constructed using different combinations of MSP-TLC profile sectors. Selected sector model is highlighted in yellow.

Sector Combinations	% Sensitivity	% Specificity
3	87.2 % (s.d. 5.7%)	89.6 % (s.d. 5.3%)
2, 3	84.3 % (s.d. 5.9%)	91.9 % (s.d. 6%)
1, 2, 3	84.5 % (s.d. 4.8%)	91.2 % (s.d. 5.9%)
2, 3, 4	83.5 % (s.d. 7%)	90.1 % (s.d. 7.3%)
1, 2, 3, 4	84.1 % (s.d. 6.4%)	89.0 % (s.d. 6.5%)
1, 3	83.7 % (s.d. 5.9%)	88.3 % (s.d. 7%)
1, 2	85.1 % (s.d. 5.5%)	85.3 % (s.d. 6%)
1, 3, 4	84 % (s.d. 6.4%)	86.4 % (s.d. 5.8%)
3, 4	83.2 % (s.d. 7.4%)	85.3 % (s.d. 8%)
2, 4	79.7 % (s.d. 6.8%)	87.9 % (s.d. 7.1%)
2	84.0 % (s.d. 6.1%)	83.1 % (s.d. 6.4%)
0, 1, 2, 3	75.0 % (s.d. 6.6%)	91.9 % (s.d. 5.4%)
1, 2, 4	79.2 % (s.d. 7.4%)	87.6 % (s.d. 8%)
0, 1, 2, 3, 4	74.9 % (s.d. 7.3%)	91.7 % (s.d. 6.6%)
0, 2, 3, 4	74.6 % (s.d. 7.5%)	90.8 % (s.d. 7.4%)
0, 1, 2	71.9 % (s.d. 5.6%)	93.4 % (s.d. 4.6%)
0, 2, 3	72.2 % (s.d. 5.8%)	92.6 % (s.d. 5.9%)
0, 1, 3, 4	77.4 % (s.d. 6.9%)	86.7 % (s.d. 6.1%)
0, 1, 2, 4	73.1 % (s.d. 7.6%)	90.9 % (s.d. 6.9%)
0, 1, 3	74.0 % (s.d. 6.8%)	89.6 % (s.d. 5.3%)
0, 2	69.9 % (s.d. 5.5%)	93.0 % (s.d. 5.2%)
0, 2, 4	71.8 % (s.d. 6.8%)	90.9 % (s.d. 7%)
1	82.6 % (s.d. 6.5%)	78.7 % (s.d. 8.2%)
0, 1	70.8 % (s.d. 7.4%)	89.3 % (s.d. 5.9%)
1, 4	79.0 % (s.d. 6.7%)	80.7 % (s.d. 8.3%)
0, 3, 4	78.1 % (s.d. 6.5%)	81.3 % (s.d. 7.7%)
0, 1, 4	75.9 % (s.d. 7.8%)	82 % (s.d. 8.5%)
0, 3	74.9 % (s.d. 8.5%)	82.8 % (s.d. 8.8%)
4	75.7 % (s.d. 13.2%)	78.3 % (s.d. 11.9%)
0, 4	78 % (s.d. 7.1%)	75.3 % (s.d. 8.4%)
0	68.8 % (s.d. 9.6%)	79.8 % (s.d. 10.9%)

**Table ESI-3.** Inner cross-validation performances of *V. negundo* models constructed using different combinations of MSP-TLC profile sectors. Selected sector model is highlighted in yellow.  
 (Selection criteria: highest combination of sensitivity and specificity, minimum sensitivity or specificity of 70%)

Sector Combinations	% Sensitivity	% Specificity
1, 2, 4	69.0 % (s.d. 7.4%)	80.8 % (s.d. 6.9%)
0, 2	68.0 % (s.d. 6.4%)	81.5 % (s.d. 7.3%)
2, 4	69.7 % (s.d. 6.6%)	79.8 % (s.d. 5.8%)
1, 2	63.7 % (s.d. 9.2%)	84.9 % (s.d. 9.5%)
2, 3	70.4 % (s.d. 5.7%)	77.2 % (s.d. 7.7%)
1, 2, 3	70.2 % (s.d. 6.9%)	77.3 % (s.d. 9.7%)
0, 1, 2	64.7 % (s.d. 6.9%)	82.3 % (s.d. 8.9%)
1, 2, 3, 4	69.2 % (s.d. 6.9%)	77.2 % (s.d. 8.1%)
2	65.8 % (s.d. 11.9%)	80 % (s.d. 11.4%)
0, 1, 2, 3	66.4 % (s.d. 7.6%)	79.1 % (s.d. 8.1%)
1, 4	70.9 % (s.d. 9%)	74.5 % (s.d. 9.6%)
1	68.3 % (s.d. 11.7%)	76.6 % (s.d. 15.4%)
0, 2, 3	67.2 % (s.d. 5.7%)	77.6 % (s.d. 7.4%)
1, 3	76.0 % (s.d. 8.1%)	68.3 % (s.d. 11.3%)
1, 3, 4	71.0 % (s.d. 7.4%)	73.0 % (s.d. 8%)
0, 1, 2, 4	62.5 % (s.d. 7.7%)	81.5 % (s.d. 7.5%)
2, 3, 4	65.3 % (s.d. 6.7%)	78.0 % (s.d. 7.1%)
0, 2, 4	62.4 % (s.d. 7.1%)	80.2 % (s.d. 7.5%)
0, 1	66 % (s.d. 10.5%)	75.8 % (s.d. 12.8%)
0, 1, 2, 3, 4	64.7 % (s.d. 7.3%)	77.1 % (s.d. 7.7%)
0, 1, 3	71.1 % (s.d. 6.6%)	70.5 % (s.d. 10.6%)
0, 2, 3, 4	63.6 % (s.d. 6.3%)	76.8 % (s.d. 7.2%)
3, 4	67.4 % (s.d. 6.8%)	71.8 % (s.d. 8.5%)
0, 1, 4	63.9 % (s.d. 10.6%)	74.9 % (s.d. 11.7%)
0, 1, 3, 4	67.4 % (s.d. 6.8%)	71.2 % (s.d. 8.6%)
4	69.2 % (s.d. 12.1%)	68.9 % (s.d. 11.5%)
0, 3	72.3 % (s.d. 7.5%)	65.6 % (s.d. 10.1%)
0, 3, 4	66.3 % (s.d. 7.7%)	69.6 % (s.d. 8.9%)
0	62.9 % (s.d. 18.4%)	71.1 % (s.d. 18.3%)
0, 4	64.5 % (s.d. 12.5%)	69.5 % (s.d. 13.3%)
3	70.5 % (s.d. 10.1%)	61.9 % (s.d. 12.3%)

	Within-Specifications Samples			Off-Specifications Samples			% Selectivity	% Specificity
	BB01-A	BB01-B	BB01-C	BB01-D	BB01-E	BB01-F		
BB02-A	BB02-B	BB02-C	BB03-A	BB03-B	BB04-A	BB04-B	BB04-C	BB04-D
BB05-A	BB05-B	BB05-C	BB06-A	BB06-B	BB06-C	BB07-A	BB07-B	BB07-C
BB08-A	BB08-B	BB08-C	BB09-A	BB09-B	BB10-A	BB10-B	BB11-A	BB11-B
BB12-A	BB12-B	BB12-C	BB13-A	BB13-B	BB13-C	BB14-A	BB14-B	BB14-C
BB15-A	BB15-B	BB15-C	BB16-A	BB16-B	BB16-C	BB17-A	BB17-B	BB17-C
BB18-A	BB18-B	BB18-C	BB19-A	BB19-B	BB19-C	BB20-A	BB20-B	BB20-C
BB21-A	BB21-B	BB21-C	BB22-A	BB22-B	BB22-C	BB23-A	BB23-B	BB23-C
BB24-A	BB24-B	BB24-C	BB25-A	BB25-B	BB25-C	BB26-A	BB26-B	BB26-C
BB27-A	BB27-B	BB27-C	BB28-A	BB28-B	BB28-C	BB29-A	BB29-B	BB29-C
BB30-A	BB30-B	BB30-C	BB31-A	BB31-B	BB31-C	BB32-A	BB32-B	BB32-C
BB33-A	BB33-B	BB33-C	BB34-A	BB34-B	BB34-C	BB35-A	BB35-B	BB35-C
BB36-A	BB36-B	BB36-C	BB37-A	BB37-B	BB37-C	BB38-A	BB38-B	BB38-C
BB39-A	BB39-B	BB39-C	BB40-A	BB40-B	BB40-C	BB41-A	BB41-B	BB41-C
BB42-A	BB42-B	BB42-C	BB43-A	BB43-B	BB43-C	BB44-A	BB44-B	BB44-C
BB45-A	BB45-B	BB45-C	BB46-A	BB46-B	BB46-C	BB47-A	BB47-B	BB47-C
BB48-A	BB48-B	BB48-C	BB49-A	BB49-B	BB49-C	BB50-A	BB50-B	BB50-C
BB51-A	BB51-B	BB51-C	BB52-A	BB52-B	BB52-C	BB53-A	BB53-B	BB53-C
BB54-A	BB54-B	BB54-C	BB55-A	BB55-B	BB55-C	BB56-A	BB56-B	BB56-C
BB57-A	BB57-B	BB57-C	BB58-A	BB58-B	BB58-C	BB59-A	BB59-B	BB59-C
BB60-A	BB60-B	BB60-C	BB61-A	BB61-B	BB61-C	BB62-A	BB62-B	BB62-C
BB63-A	BB63-B	BB63-C	BB64-A	BB64-B	BB64-C	BB65-A	BB65-B	BB65-C
BB66-A	BB66-B	BB66-C	BB67-A	BB67-B	BB67-C	BB68-A	BB68-B	BB68-C
BB69-A	BB69-B	BB69-C	BB70-A	BB70-B	BB70-C	BB71-A	BB71-B	BB71-C
BB72-A	BB72-B	BB72-C	BB73-A	BB73-B	BB73-C	BB74-A	BB74-B	BB74-C
BB75-A	BB75-B	BB75-C	BB76-A	BB76-B	BB76-C	BB77-A	BB77-B	BB77-C
BB78-A	BB78-B	BB78-C	BB79-A	BB79-B	BB79-C	BB80-A	BB80-B	BB80-C
BB81-A	BB81-B	BB81-C	BB82-A	BB82-B	BB82-C	BB83-A	BB83-B	BB83-C
BB84-A	BB84-B	BB84-C	BB85-A	BB85-B	BB85-C	BB86-A	BB86-B	BB86-C
BB87-A	BB87-B	BB87-C	BB88-A	BB88-B	BB88-C	BB89-A	BB89-B	BB89-C
BB90-A	BB90-B	BB90-C	BB91-A	BB91-B	BB91-C	BB92-A	BB92-B	BB92-C
BB93-A	BB93-B	BB93-C	BB94-A	BB94-B	BB94-C	BB95-A	BB95-B	BB95-C
BB96-A	BB96-B	BB96-C	BB97-A	BB97-B	BB97-C	BB98-A	BB98-B	BB98-C
BB99-A	BB99-B	BB99-C	BB100-A	BB100-B	BB100-C	BB101-A	BB101-B	BB101-C

**Fig. ESI-5.** Predicted classifications of *B. balsamifera* samples from SIMCA outer cross-validation loops.

Model predictions are aggregated to yield a final classification by majority 2/3 vote. Within-specifications = 1, Off-specifications = -1, Abstain = 0

**Fig. ESI-6.** Predicted classifications of *V. negundo* samples from SIMCA outer cross-validation loops.

Model predictions are aggregated to yield a final classification by majority 2/3 vote. Within-specifications = 1, Off-specifications = -1, Abstain = 0

**Table ESI-4.** Comparison of classification performances of MSP-TLC system, and the adapted TLC and standard Pharmacopeia TLC methods reported in ref. [1]

Plant Species	Performance Parameter	MSP-TLC system	Adapted TLC Method in ref. [1]	Pharmacopeia TLC Method in ref. [1]
<i>B. balsamifera</i>	Sensitivity	95.1%	90.2%	100%
	Specificity	91.7%	86.2%	100%
	Classification of mixtures	5% (w/w) <i>V. negundo</i> misclassified as acceptable	5-60% (w/w) <i>V. negundo</i> misclassified as acceptable	5-50% (w/w) <i>V. negundo</i> misclassified as acceptable
<i>V. negundo</i>	Sensitivity	74.4%	81.4%	100%
	Specificity	73.1%	92.0%	100%
	Classification of mixtures	5-30% (w/w) <i>B. balsamifera</i> misclassified as acceptable	5-10% (w/w) <i>B. balsamifera</i> misclassified as acceptable	5-80% (w/w) <i>B. balsamifera</i> misclassified as acceptable

Reference:

- 1 S. M. Sibug-Torres, I. D. Padolina, P. Cruz, M. R. Yabillo, F. C. Garcia, M. J. Garrovillas and E. P. Enriquez, *Anal. Methods*, 2019, **11**, 693–862.