Emission factors of $PM_{2.5}$-Bounded selected metals, organic carbon, elemental carbon, and water-soluble ionic species emitted from combustions of biomass materials for source Apportionment—A new database for 17 plant species

Siwatt Pongpiachan, Qiyuan Wang, Thaneeya Chetiyanukornkul, Li Li, Li Xing, Guohui Li, Yongming Han, Junji Cao, and Vanisa Surapipith

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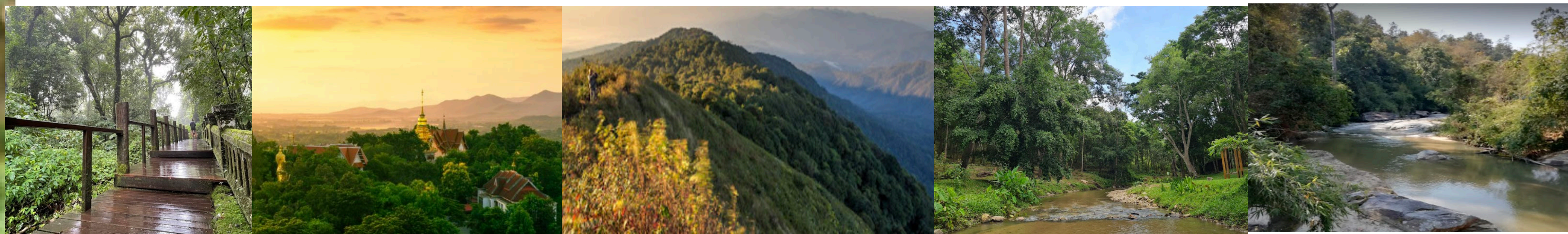

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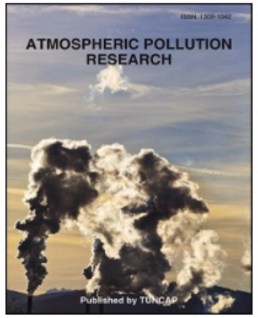

Emission factors of $PM_{2.5}$-Bounded selected metals, organic carbon, elemental carbon, and water-soluble ionic species emitted from combustions of biomass materials for source Apportionment—A new database for 17 plant species

Siwatt Pongpiachan [a,e,*], Qiyuan Wang [b,**], Thaneeya Chetiyanukornkul [c,d], Li Li [b], Li Xing [f], Guohui Li [b], Yongming Han [b], Junji Cao [b], Vanisa Surapipith [e]

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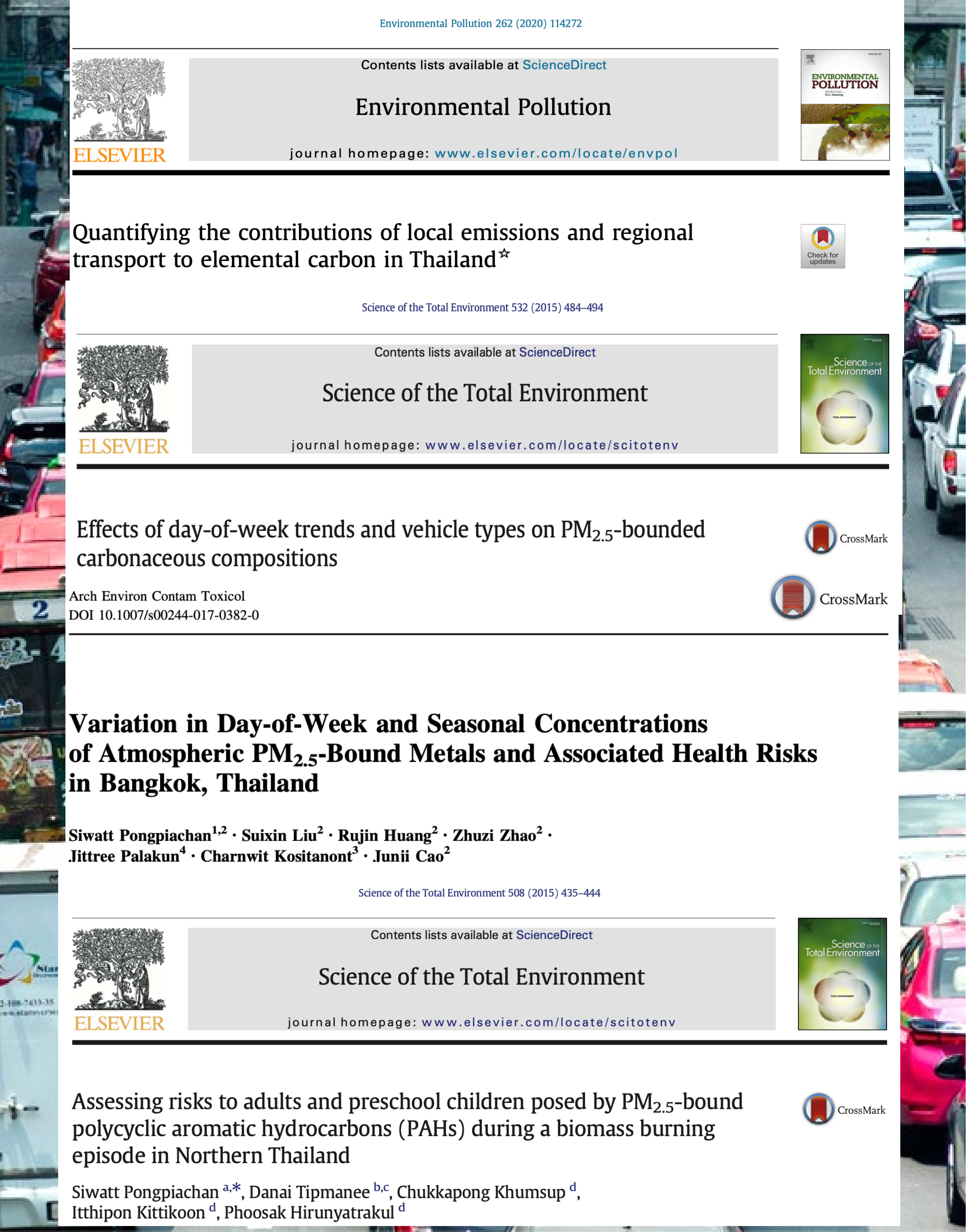

Our previous studies related to traffic emissions in ambient air

Organic Carbon/Elemental Carbon

Xing, L., Li, G., Pongpiachan, S., Wang, Q., Han, Y., Cao, J., ... & Poshyachinda, S. (2020). Quantifying the contributions of local emissions and regional transport to elemental carbon in Thailand. Environmental Pollution, 262, 114272.

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Atmospheric Environment 108 (2015) 13–19

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Enhanced PM_{10} bounded PAHs from shipping emissions

CrossMark

S. Pongpiachan [a,*], M. Hattayanone [b], C. Choochuay [a], R. Mekmok [c], N. Wuttijak [c], A. Ketratanakul [c]

Aerosol and Air Quality Research

ORIGINAL RESEARCH
https://doi.org/10.4209/aaqr.210030

Using Synchrotron Radiation X-ray Fluorescence (SRXRF) to Assess the Impacts of Shipping Emissions on the Variations of PM_{10}-bound Elemental Species

Siwatt Pongpiachan[1*], Nichada Jearanaikoon[2], Kanjana Thumanu[2], Jureerat Pradubsri[2], Ratchadaporn Supruangnet[2], Chaisri Tharasawatpipat[3], Muhammad Zaffar Hashmi[4], Ronbanchob Apiratikul[3]

Our previous studies related to shipping emissions in ambient air

- Pongpiachan, S., Hattayanone, M., Choochuay, C., Mekmok, R., Wuttijak, N., & Ketratanakul, A. (2015). Enhanced PM10 bounded PAHs from shipping emissions. Atmospheric environment, 108, 13-19.
- Pongpiachan, S., Jearanaikoon, N., Thumanu, K., Pradubsri, J., Supruangnet, R., Tharasawatpipat, C., ... & Apiratiku, R. (2021). Using Synchrotron Radiation X-ray Fluorescence (SRXRF) to Assess the Impacts of Shipping Emissions on the Variations of PM10-bound Elemental Species. Aerosol and Air Quality Research, 21, 210030.
- Pongpiachan, S., Jearanaikoon, N., Thumanu, K., Pradubsri, J., Supruangnet, R., Tharasawatpipat, C., ... & Apiratiku, R. (2021). Applying Synchrotron Radiation-based Attenuated Total Reflection-Fourier Transform Infrared to Evaluate the Effects of Shipping Emissions on Fluctuations of PM10-Bound Organic Functional Groups. Atmospheric Pollution Research, in press.

Previous studies related to the impact of forest fire & biomass burnings on air quality

Atmospheric Pollution Research xxx (2017) 1–12

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Effect of agricultural waste burning season on $PM_{2.5}$-bound polycyclic

Journal Pre-proof

Ambient $PM_{2.5}$, Polycyclic Aromatic Hydrocarbons and

Atmospheric Environment 180 (2018) 184–191

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Enhanced light absorption due to the mixing state of black carbon in fresh biomass burning emissions

Check for updates

Qiyuan Wang[a,*], Junji Cao[a,b,**], Yongming Han[a,c], Jie Tian[d], Yue Zhang[d], Siwatt Pongpiachan[e], Yonggang Zhang[a], Li Li[a], Xinyi Niu[c], Zhenxing Shen[d], Zhuzi Zhao[a], Danai Tipmanee[f],

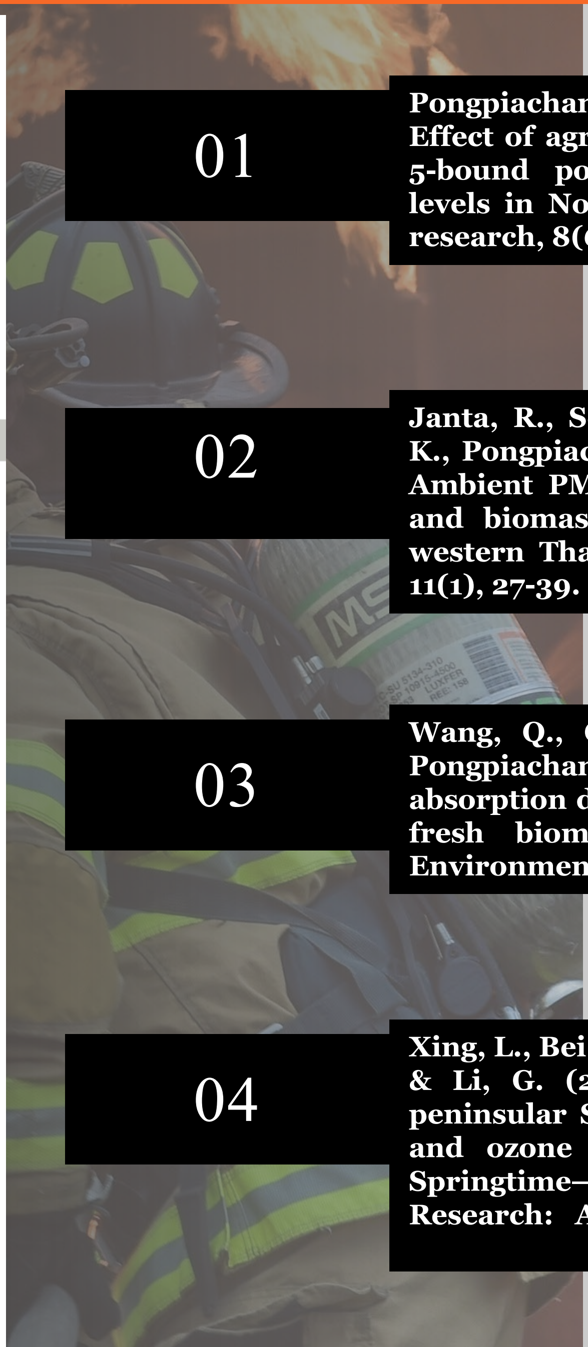
JGR Atmospheres

RESEARCH ARTICLE

10.1029/2021JD034908

Special Section:
Fire in the Earth System

Impacts of Biomass Burning in Peninsular Southeast Asia on $PM_{2.5}$ Concentration and Ozone Formation in Southern China During Springtime—A Case Study



01 Pongpiachan, S., Hattayanone, M., & Cao, J. (2017). Effect of agricultural waste burning season on PM2. 5-bound polycyclic aromatic hydrocarbon (PAH) levels in Northern Thailand. Atmospheric pollution research, 8(6), 1069-1080.

02 Janta, R., Sekiguchi, K., Yamaguchi, R., Sopajaree, K., Pongpiachan, S., & Chetiyanukornkul, T. (2020). Ambient PM2. 5, polycyclic aromatic hydrocarbons and biomass burning tracer in Mae Sot District, western Thailand. Atmospheric Pollution Research, 11(1), 27-39.

03 Wang, Q., Cao, J., Han, Y., Tian, J., Zhang, Y., Pongpiachan, S., ... & Sun, J. (2018). Enhanced light absorption due to the mixing state of black carbon in fresh biomass burning emissions. Atmospheric Environment, 180, 184-191.

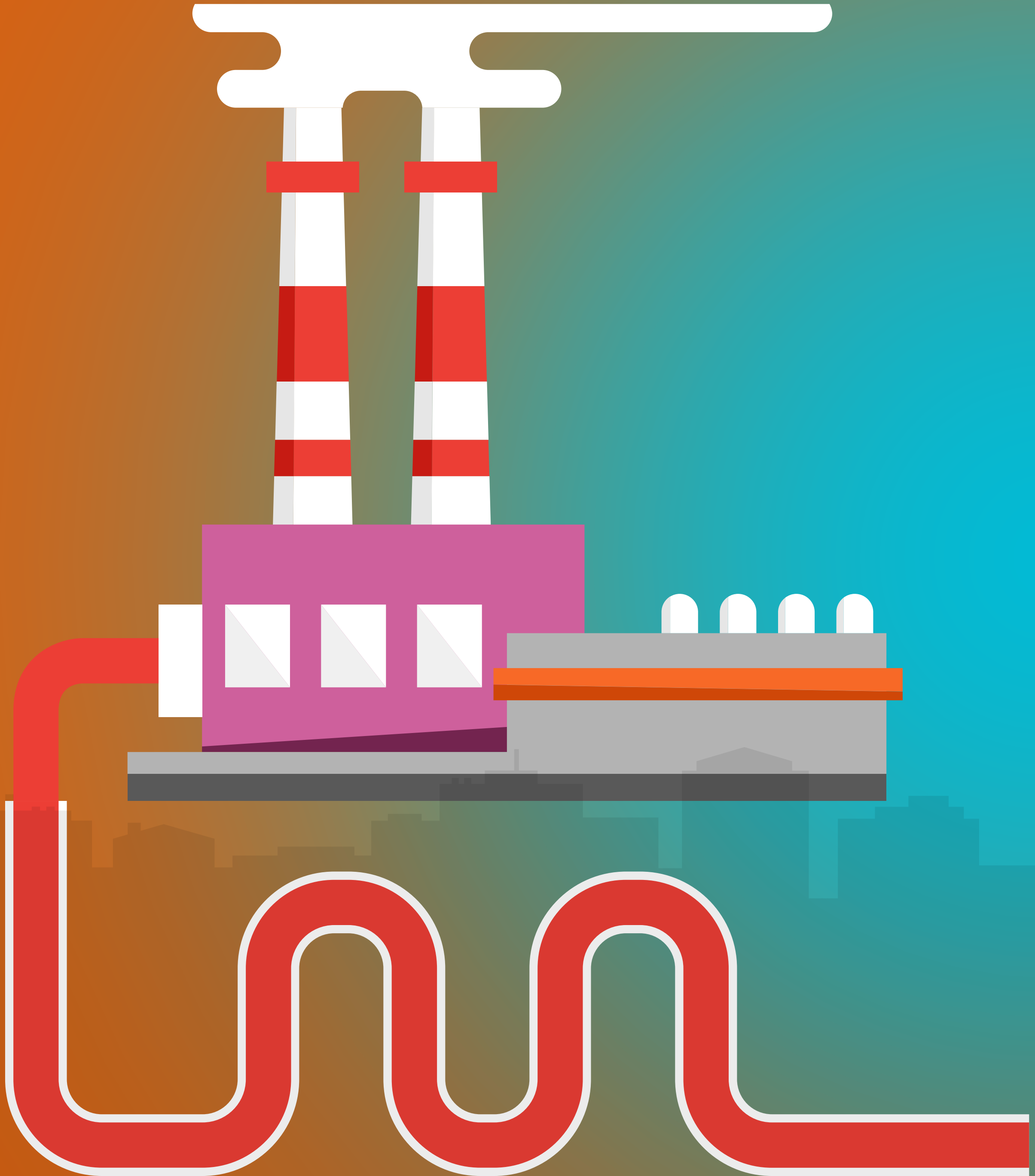
04 Xing, L., Bei, N., Guo, J., Wang, Q., Liu, S., Han, Y., ... & Li, G. (2021). Impacts of biomass burning in peninsular Southeast Asia on PM2. 5 concentration and ozone formation in Southern China During Springtime—A case study. Journal of Geophysical Research: Atmospheres, 126(22), e2021JD034908.

Receptor models can be categorized into two types, namely, Chemical Mass Balance (CMB) and multivariate models (e.g. PCA, PMF, UNMIX).

This technique has been widely applied to source apportionment of particulate pollutants, especially trace metals, and more recently, PAHs.

In urban atmosphere, which is composed by many potential and diverse sources, Multivariate models have been chosen by many workers for source apportionment.

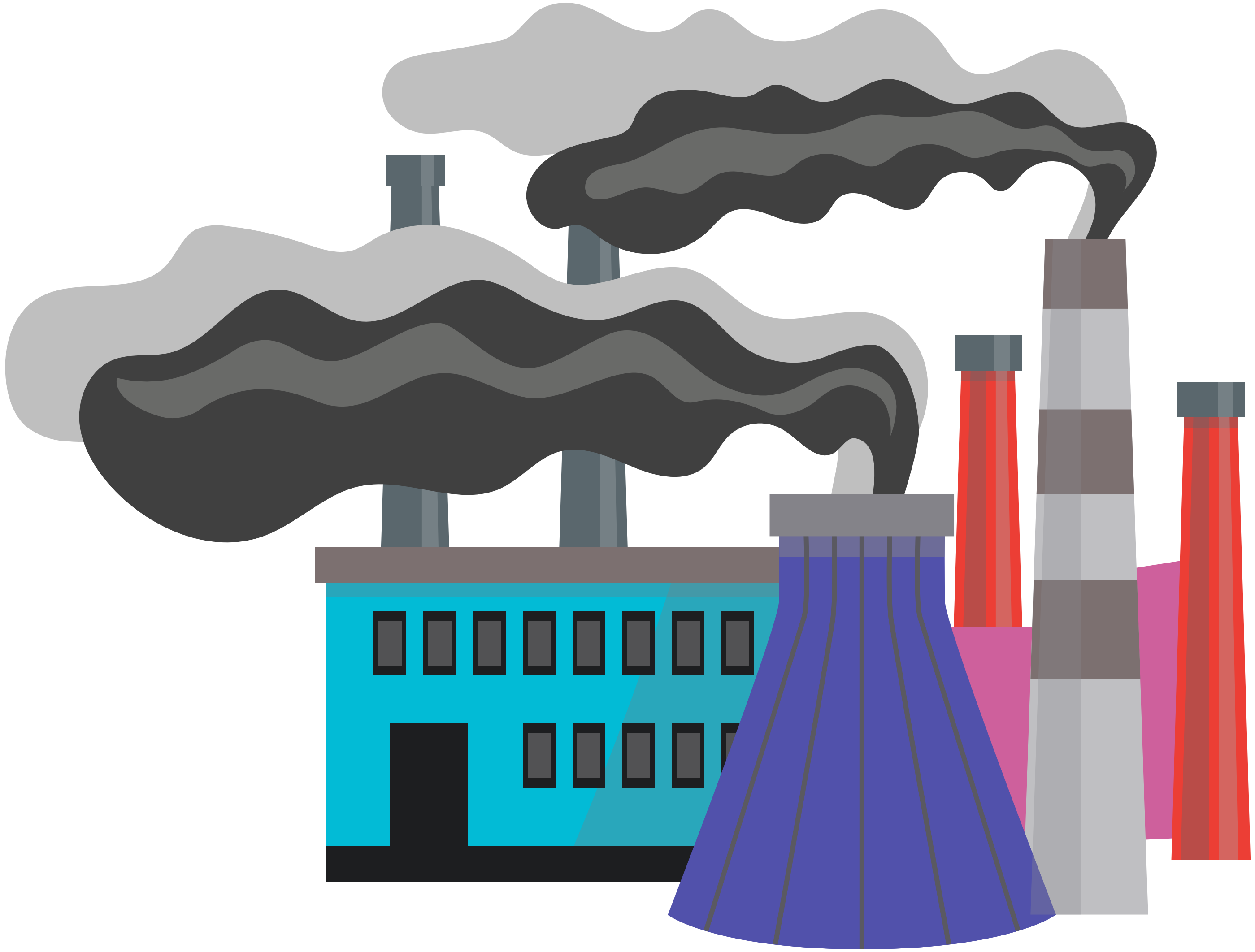
Factor analysis offers the advantages of not requiring prior knowledge of the chemical composition and size distribution of emissions from specific sources



Limitations of Multivariate Models (i.e. PCA, PMF, UNMIX)

- it can recognize at most only about eight individual source categories in any study, and poor discrimination of closely related source categories is commonly found.
- A further disadvantage of multivariate factor analysis is that large numbers of ambient air samples must be collected and analysed (usually at least 50) and the statistically independent source tracers are required for each major source type.
- It is quite common that PCA, PMF, and UNMIX showed different source apportionment results which will cause some difficulties for data interpretation.

Chemical Mass Balance (CMB) Model (Watson et al., 1990)



Model Advantages & Limitations

- The theory of CMB model is based on the basis of the law of mass conservation and thus the reliability of this model is strongly influenced by the stability of chemical compositions of the emission source profile. In reality, SVOC can be the subject of secondary formation due to atmospheric chemical reactions.

- **You need chemical source profiles of each source to run the model!!**

Watson, J. G., Robinson, N. F., Chow, J. C., Henry, R. C., Kim, B. M., Pace, T. G., ... & Nguyen, Q. (1990). The USEPA/DRI chemical mass balance receptor model, CMB 7.0. *Environmental Software*, *5*(1), 38-49.

Objectives

To chemically characterize 20 selected metals (i.e. Al, Si, S, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Sr, Ba, and Pb), OC, EC and WSIS in $PM_{2.5}$, emitted from the combustion of 17 biomass materials

To compute OC/EC and metal ratios, which have been widely employed as one of source identification techniques

To conduct the Linear Regression Analysis of OC versus EC

Biomass Sampling Sites

Doi Inthanon (482 km²)

Doi Inthanon is the highest peak of the Inthanon Range of the Thanon Thong Chai Range, a subrange of the Shan Hills in the Thai highlands stretching southwards from the Daen Lao Range.

Doi Suthep-Pui (261 km²)

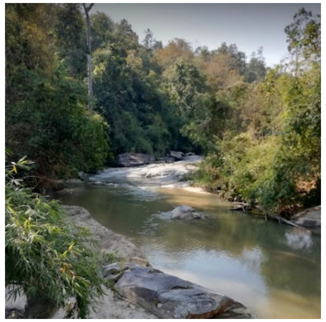
Doi Suthep–Pui National Parkis a national park in Chiang Mai Province in Thailand. It includes Wat Phra That Doi Suthep.

Doi Pha Hom Pok (524 km²)

Doi Pha Hom Pok is the northernmost national park in Thailand. It straddles Fang, Mae Ai, and Chai Prakan Districts of Chiang Mai Province. The park covers 524 km2 of the mountain area of the Daen Lao Range, at the border with Myanmar. .

Khun Khan (208 km²)

Khun Khan National Park is a national park in Thailand's Chiang Mai Province. This mountainous park is home to forests, waterfalls.



Mae Wang (120 km²)

Mae Wang National Park is located in Chom Thong District, Doi Lo District and Mae Wang District in Chiang Mai Province.

17 Biomass Species Commonly Found in Southeast Asia

Terminalia catappa

Bambusa sp.

Alstonia scholaris

Eucalyptus sp.

Saccharum officinarum

Cassia fistula

Polyalthia longifolia

Casuarina equisetifolia

Lagerstroemia sp.

Citrus hystrix

Gossypium sp

Cycas sp.

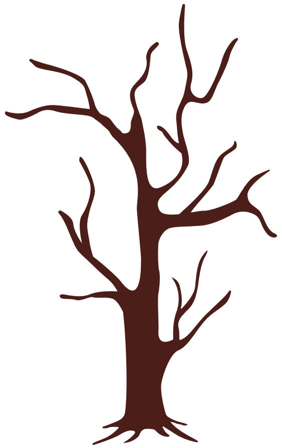
Albizia saman

Afzelia xylocarpa

Azadirachta indica

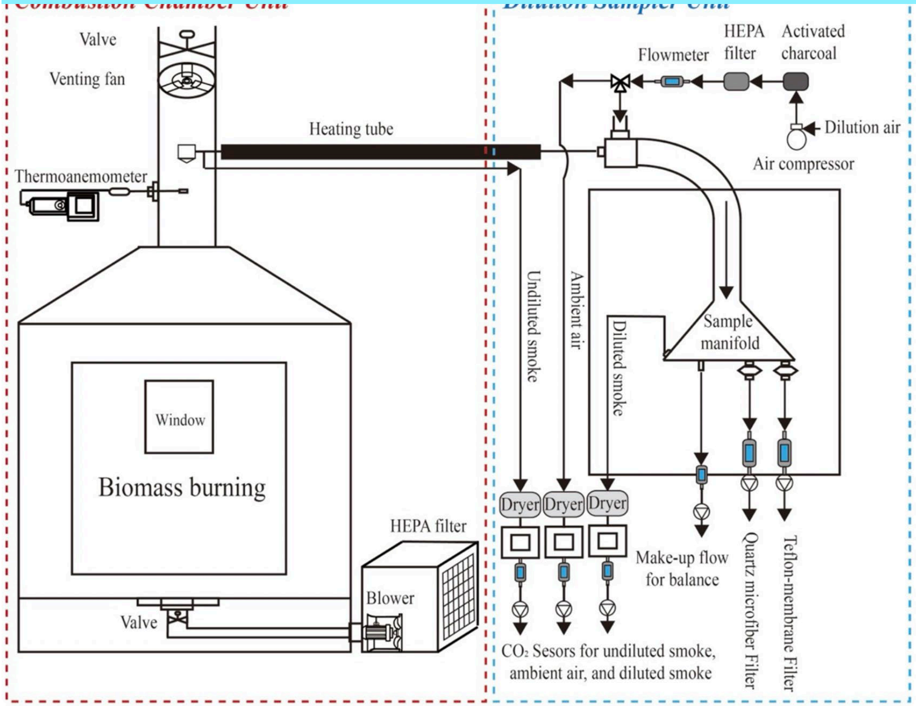
Leucaena leucocephala

Tectona grandis



50 g dry weight

The Institute of Earth Environment, Chinese Academy of Sciences (IEECAS)



*Before burning the samples were stored in a room with temperature of ~20 °C and relative humidity of 35–45% for a month.

*The volume of the combustion chamber is about 8 m^3 (1.8 m × 1.8 m × 2.2 m).

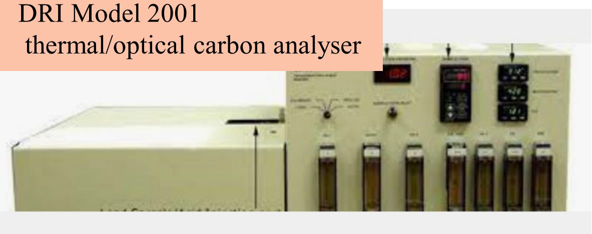
*A dilution sampler (Model 18, Baldwin Environmental Inc., Reno, NV, USA) was set followed the chamber, which used to dilute the smoke.

Epsilon 5 ED-XRF

Heavy Metals

Al, Si, S, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Sr, Ba, and Pb

DRI Model 2001 thermal/optical carbon analyser



Organic Carbon/Elemental Carbon (OC/EC)

OC (OC1, OC2, OC3, and OC4) +EC (EC1, EC2, EC3, OP)

Dionex DX-600

Water Soluble Ionic Species (WSIS)

Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+} Cl^-, NO_2^-, NO_3^-, SO_4^{2-}

Table 1

Ranking of emission factors (g compound emitted per kg dry biomass burned) of 32 chemical species (i.e. WSIS, carbonaceous aerosols, and selected metals) from combustions of 17 biomass materials.

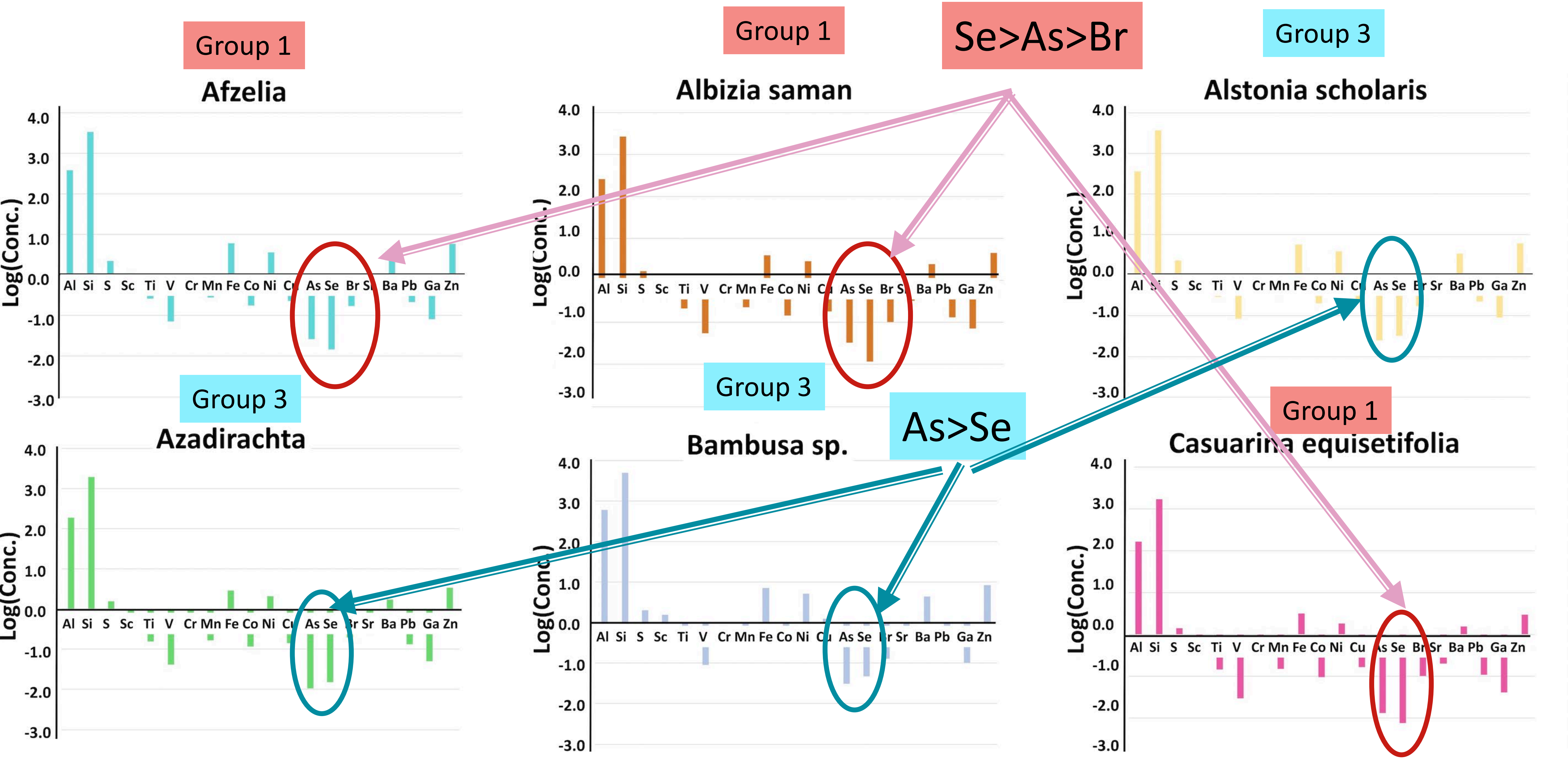
	Cl^-	NO_2^-	NO_3^-	SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	TC	OC	EC	Al	Si	S	Sc
Afzelia xylocarpa	13	5	8	10	7	10	13	11	4	6	6	6	6	9	9	7
Albizia saman	10	12	10	14	12	14	5	12	11	12	12	13	12	12	13	12
Alstonia scholaris	14	15	12	12	13	15	17	4	5	10	10	8	8	7	8	8
Azadirachta Indica	5	13	16	16	15	3	3	15	13	15	15	15	16	16	14	16
Bambusa sp.	4	9	6	9	8	4	11	1	2	11	11	12	1	3	11	3
Cassia fistula	16	1	3	4	9	11	15	16	15	1	1	1	7	6	4	6
Casuarina equisetifolia	9	10	15	17	16	17	2	13	12	17	16	16	17	17	16	17
Citrus hystrix	6	7	5	5	5	8	8	5	3	4	4	4	4	5	5	5
Cycas sp.	17	3	2	1	1	5	12	7	9	3	3	2	3	2	1	2
Eucalyptus sp.	7	16	13	13	14	16	10	10	6	7	7	11	9	8	12	11
Gossypium sp.	8	2	1	6	2	6	14	17	17	13	13	14	15	14	15	14
Lagerstroemia sp.	12	4	7	2	6	9	9	3	7	9	9	9	10	10	3	10
Leucaena leucocephala	1	8	4	3	4	7	1	9	14	16	17	10	5	4	2	4
Polyalthia longifolia	11	11	11	8	11	13	4	14	16	2	2	3	14	13	6	13
Saccharum officinarum	3	6	14	7	3	2	6	2	1	5	5	5	2	1	10	1
Tectona grandis	15	14	9	11	10	12	16	6	10	8	8	7	11	11	7	9
Terminalia catappa	2	17	17	15	17	1	7	8	8	14	14	17	13	15	17	15

	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	As	Se	Br	Sr	Ba	Pb	Ga	Total
Afzelia xylocarpa	10	9	10	11	6	11	9	11	7	12	7	10	8	10	8	**278**
Albizia saman	12	12	12	12	14	13	12	15	5	15	12	12	12	16	11	**383**
Alstonia scholaris	7	4	7	7	7	7	7	9	9	3	6	7	7	9	6	**275**
Azadirachta Indica	16	16	16	16	17	16	16	17	16	8	5	16	16	15	16	**444**
Bambusa sp.	4	3	3	3	4	3	3	1	6	1	8	3	3	2	5	**161**
Cassia fistula	6	6	6	5	8	6	6	7	13	7	10	6	6	5	7	**226**
Casuarina equisetifolia	17	17	17	17	16	17	17	16	15	17	11	17	17	17	17	**489**
Citrus hystrix	5	7	5	6	5	5	5	5	4	5	4	5	5	3	4	**161**
Cycas sp.	1	2	2	1	2	2	2	3	1	6	15	2	2	4	3	**123**
Eucalyptus sp.	8	11	8	9	9	8	8	10	10	10	3	8	9	8	10	**307**
Gossypium sp.	15	14	15	15	12	14	15	13	14	9	14	15	14	12	12	**388**
Lagerstroemia sp.	9	10	9	10	10	9	10	6	17	11	16	9	10	7	9	**281**
Leucaena leucocephala	3	5	4	4	3	4	4	4	2	4	2	4	4	6	2	**168**
Polyalthia longifolia	13	13	13	14	15	12	13	14	8	16	9	13	13	14	13	**358**
Saccharum officinarum	2	1	1	2	1	1	1	2	3	2	1	1	1	1	1	**95**
Tectona grandis	11	8	11	8	11	10	11	8	11	13	17	11	11	11	14	**341**
Terminalia catappa	14	15	14	13	13	15	14	12	12	14	13	14	15	13	15	**418**

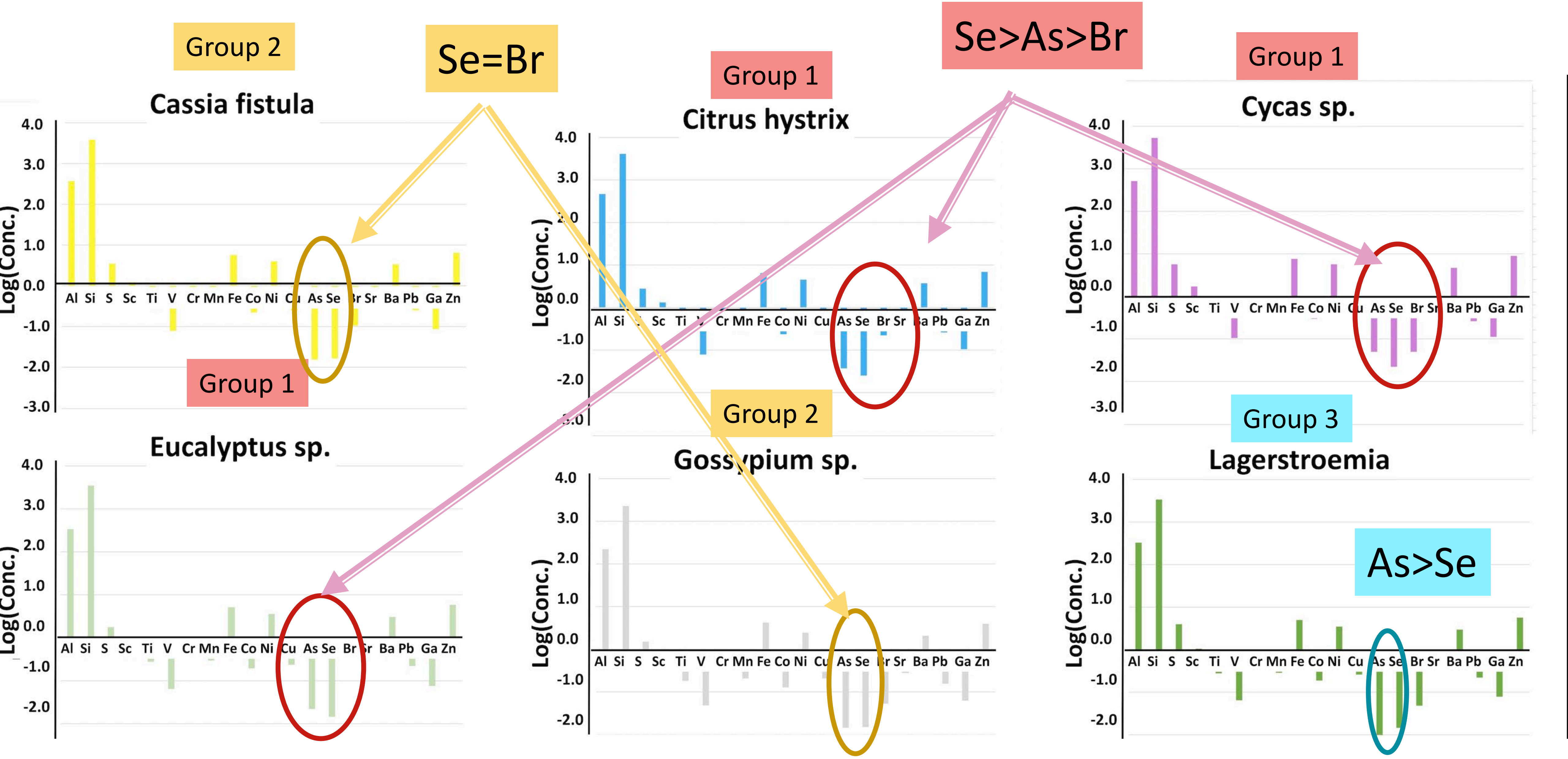
The similar distributional characteristics of logarithm profiles corresponding to the three main features.

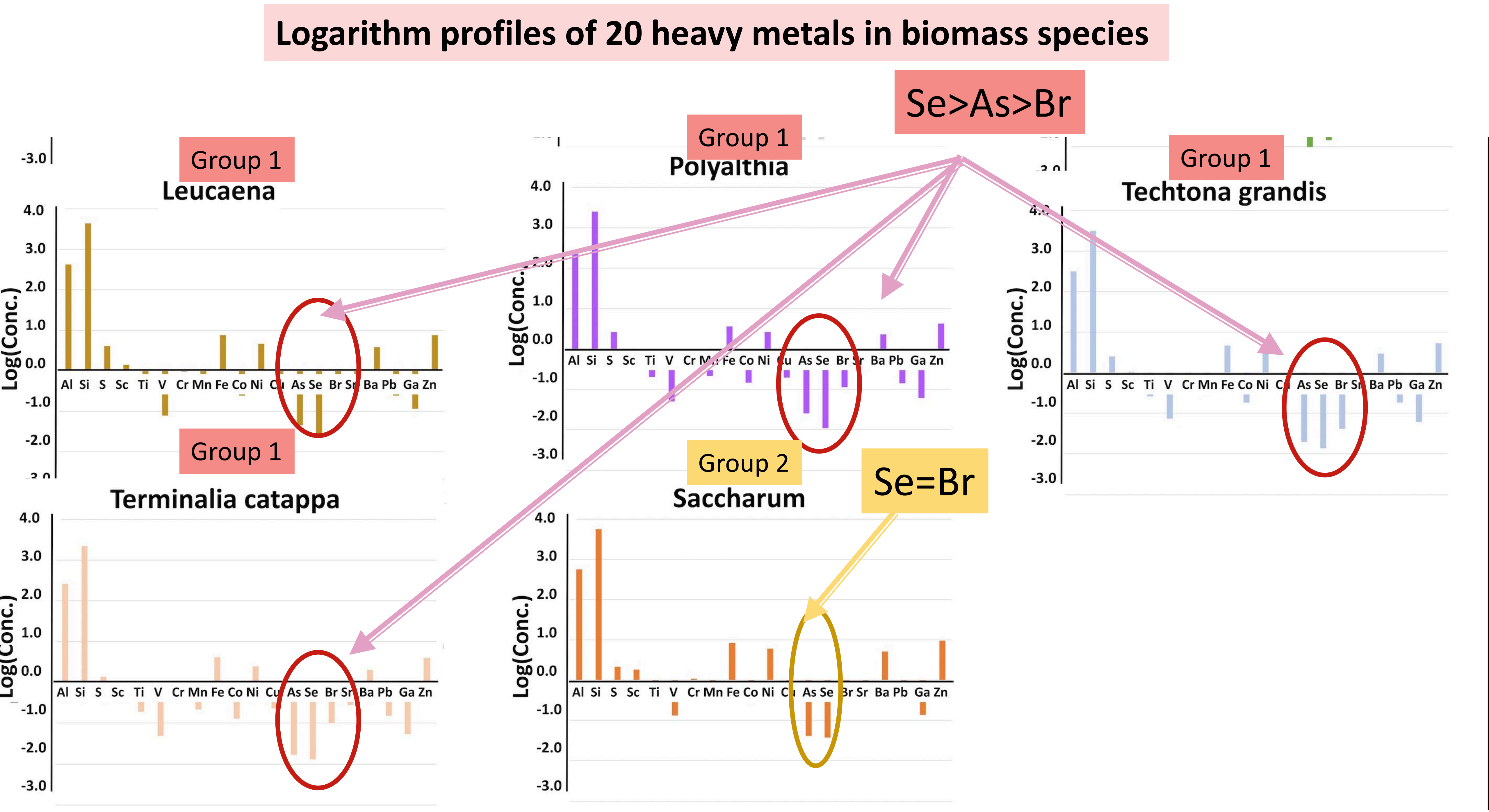
Firstly, the most abundant quantified metals in $PM_{2.5}$ were Si, Al, Fe, and Zn.
Secondly, the decreasing order of transition metals in all groups was Cr>Mn>Ti>V.
Thirdly, the descending sequence of alkaline earth metals and post-transition metals was Ba>Sr>Pb>Ga.

Logarithm profiles of 20 heavy metals in biomass species



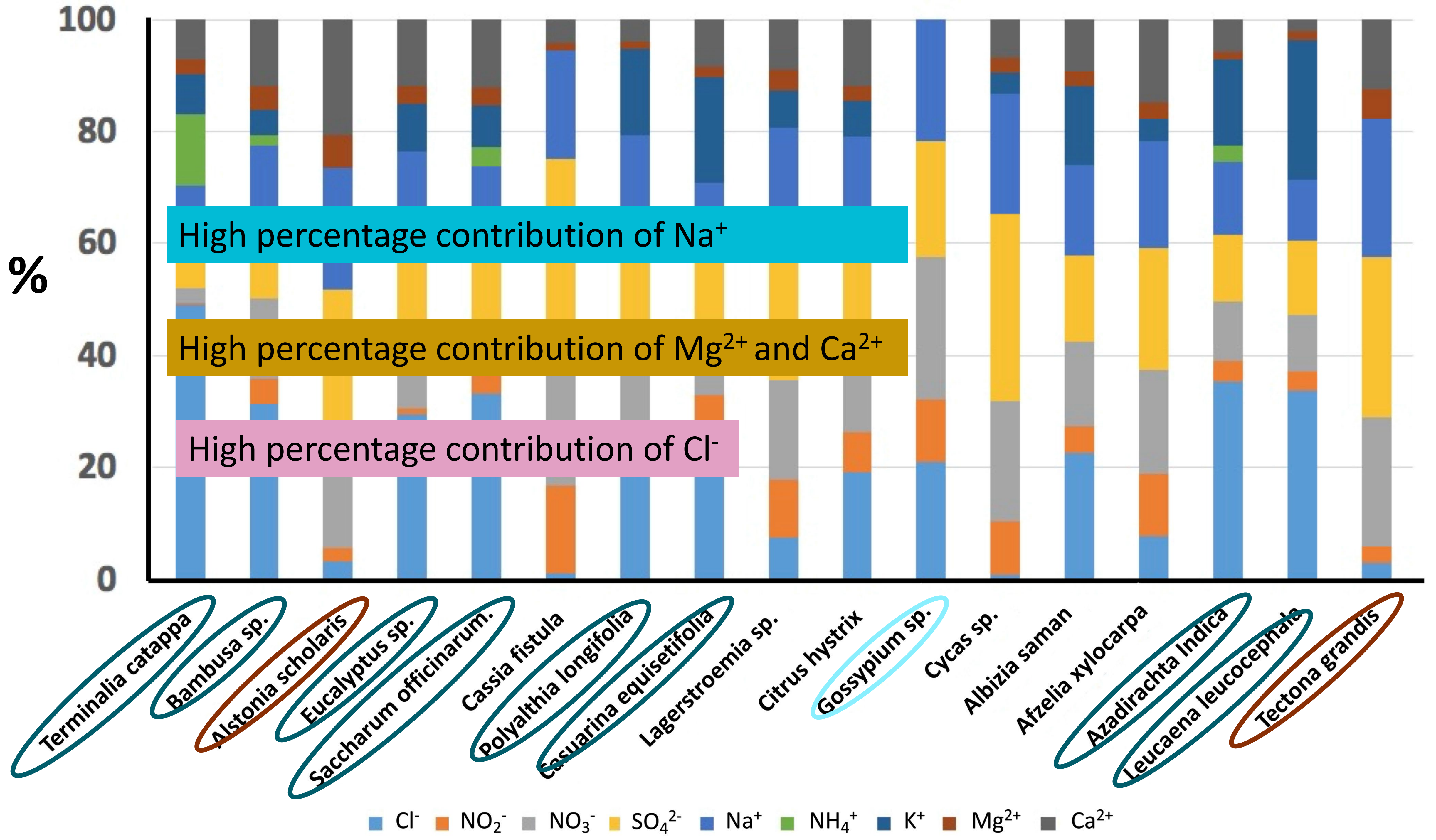
Logarithm profiles of 20 heavy metals in biomass species




Logarithm profiles of 20 heavy metals in biomass species
Se>As>Br
Group 1
Leucaena
Group 1
Polyalthia
Group 1
Techtona grandis
Group 1
Terminalia catappa
Group 2
Saccharum
Se=Br
Log(Conc.)
4.0
3.0
2.0
1.0
0.0
-1.0
-2.0
-3.0
Al Si S Sc Ti V Cr Mn Fe Co Ni Cu As Se Br Sr Ba Pb Ga Zn

In this study, the highest *EF* of highly toxic metals, such as V, Cr, Co, Ni, and Pb, were observed in $PM_{2.5}$, released from the combustion of sugarcane (i.e. *Saccharum officinarum*), with average values of 0.136±0.0634 g kg^{-1}, 1.10±0.311 g kg^{-1}, 0.318±0.108 g kg^{-1}, 6.10±1.87 g kg^{-1}, and 0.354±0.160 g kg^{-1}, respectively

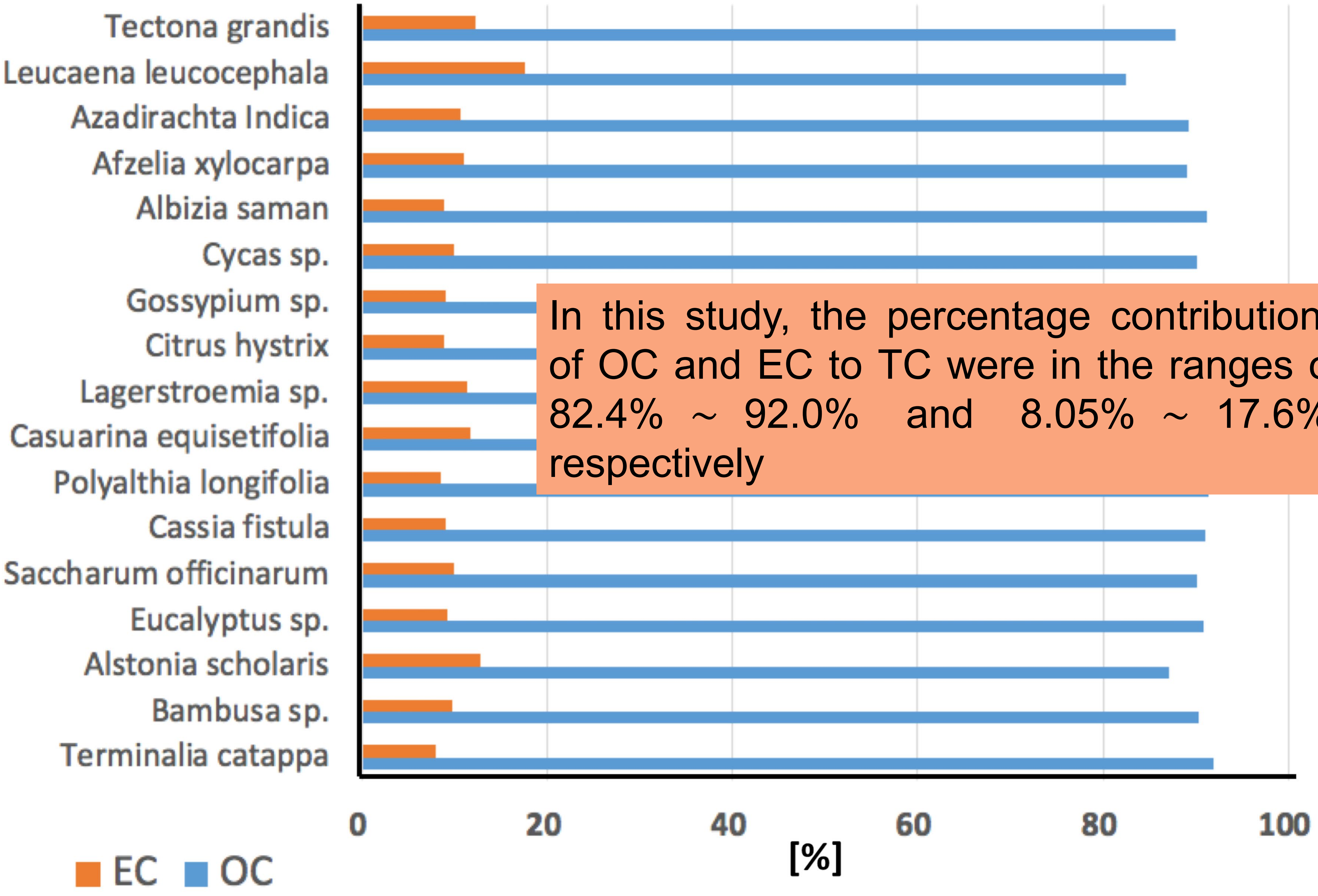
The relatively high EF values of V, Cr, Co, Ni, and Pb emitted from sugar cane burning might have raised concerns because of its comparatively high degree of toxicity.



%
100
80
60
40
20
0
High percentage contribution of Na+
High percentage contribution of Mg2+ and Ca2+
High percentage contribution of Cl-
Terminalia catappa
Bambusa sp.
Alstonia scholaris
Eucalyptus sp.
Saccharum officinarum.
Cassia fistula
Polyalthia longifolia
Casuarina equisetifolia
Lagerstroemia sp.
Citrus hystrix
Gossypium sp.
Cycas sp.
Albizia saman
Afzelia xylocarpa
Azadirachta Indica
Leucaena leucocephala
Tectona grandis
Cl-
NO2-
NO3-
SO42-
Na+
NH4+
K+
Mg2+
Ca2+

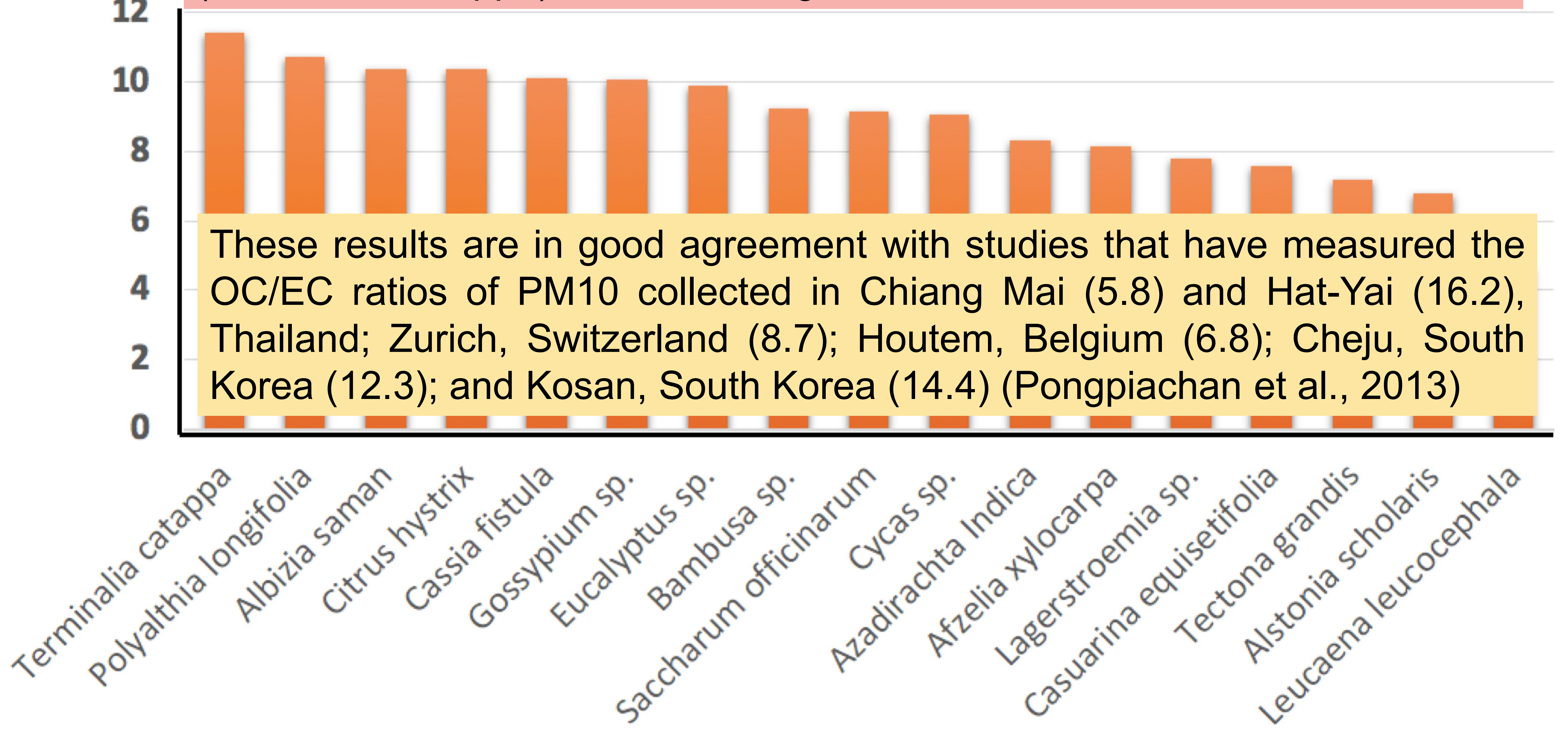
The comparatively high percentage contributions of Na^{+}, Mg^{2+} and Ca^{2+} were detected in $PM_{2.5}$, emitted from the burning of *Cycas sp*., *Bambusa sp.* and *Saccharum officinarum*. Since 83% of total crop residue burning in Thailand comes from rice and sugarcane (Kumar et al., 2020), the application of Mg^{2+} and Ca^{2+} as chemical tracers for identifying maritime aerosols need to be conducted with great caution particularly in the middle of haze episode.

Analytical measurements of K^+ were below the detection limits in the case of Alstonia scholaris, Cassia fistula, Gossypium sp., and Tectona grandis. These results indicate that the conventional method of applying K+ as a sole tracer of biomass burning for source apportionment could be problematic (Pachon et al., 2013; Yu et al., 2018). Thus, applying this tracer should be performed with careful consideration of the distribution patterns of plant species diversity in the study area.

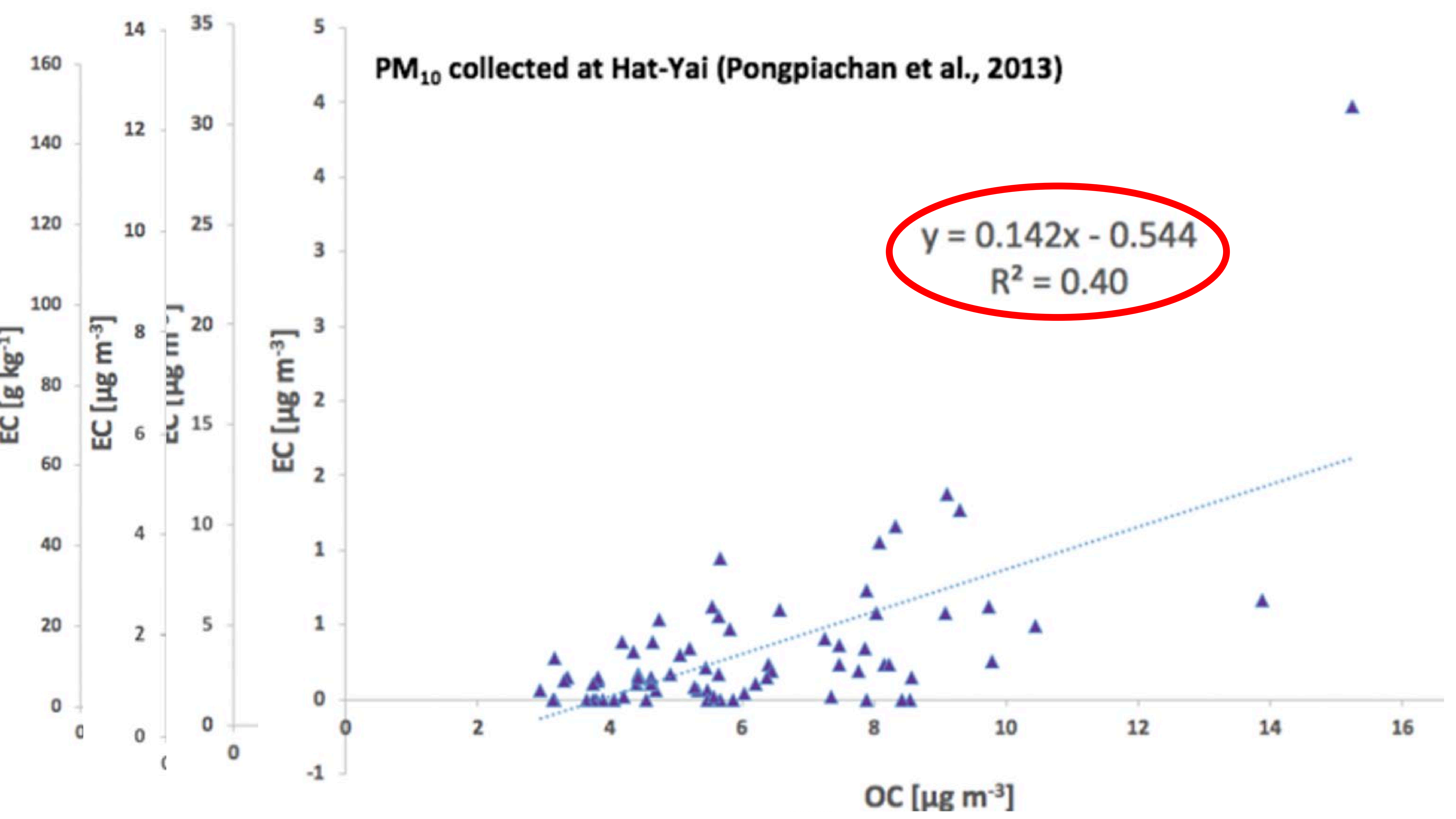


In this study, the percentage contributions of OC and EC to TC were in the ranges of 82.4% ~ 92.0% and 8.05% ~ 17.6%, respectively

The OC/EC ratios ranged from 4.68 (Leucaena leucocephala) to 11.4 (Terminalia catappa) with an average value of 8.88±1.72

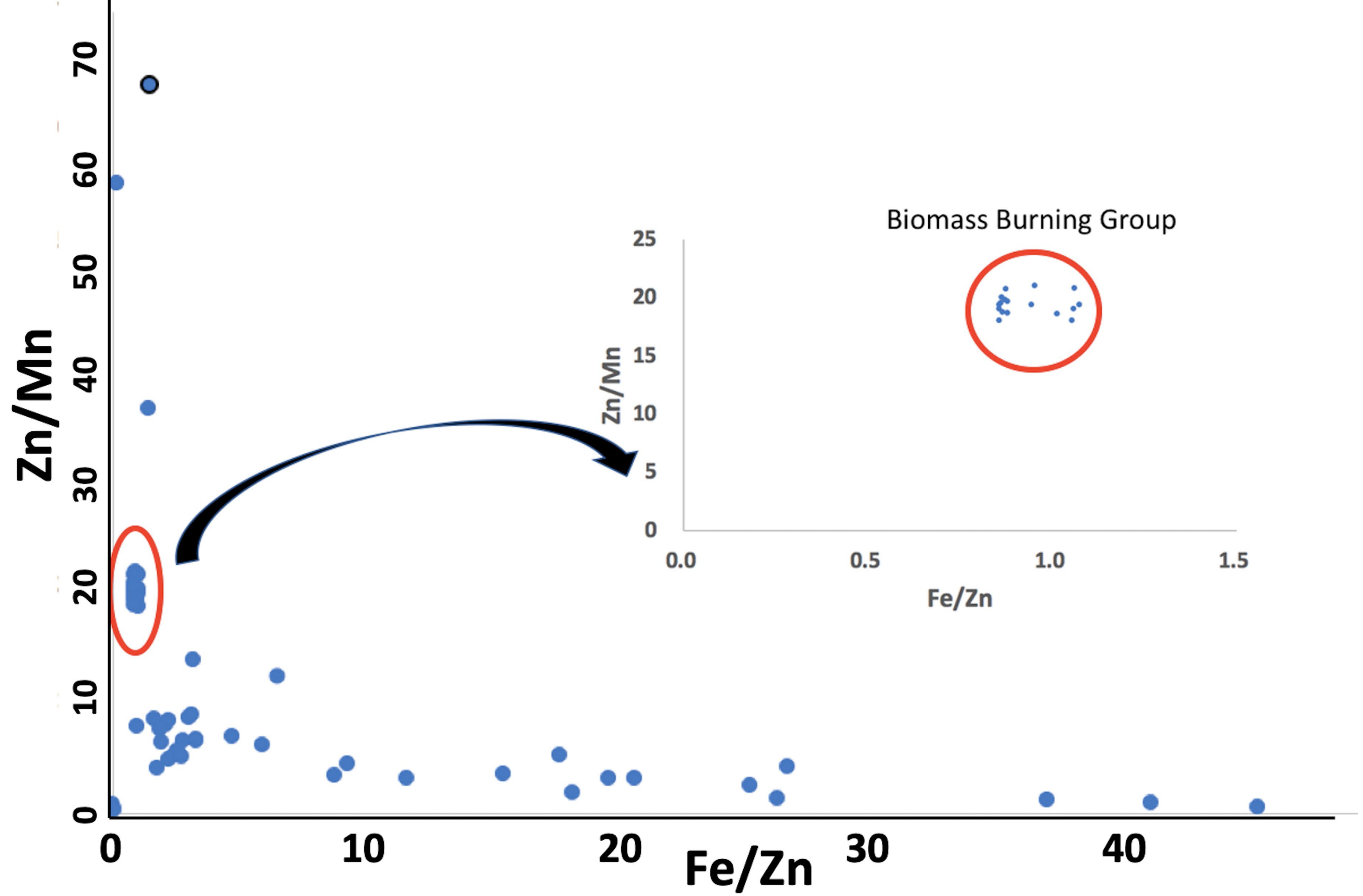


These results are in good agreement with studies that have measured the OC/EC ratios of PM10 collected in Chiang Mai (5.8) and Hat-Yai (16.2), Thailand; Zurich, Switzerland (8.7); Houtem, Belgium (6.8); Cheju, South Korea (12.3); and Kosan, South Korea (14.4) (Pongpiachan et al., 2013)

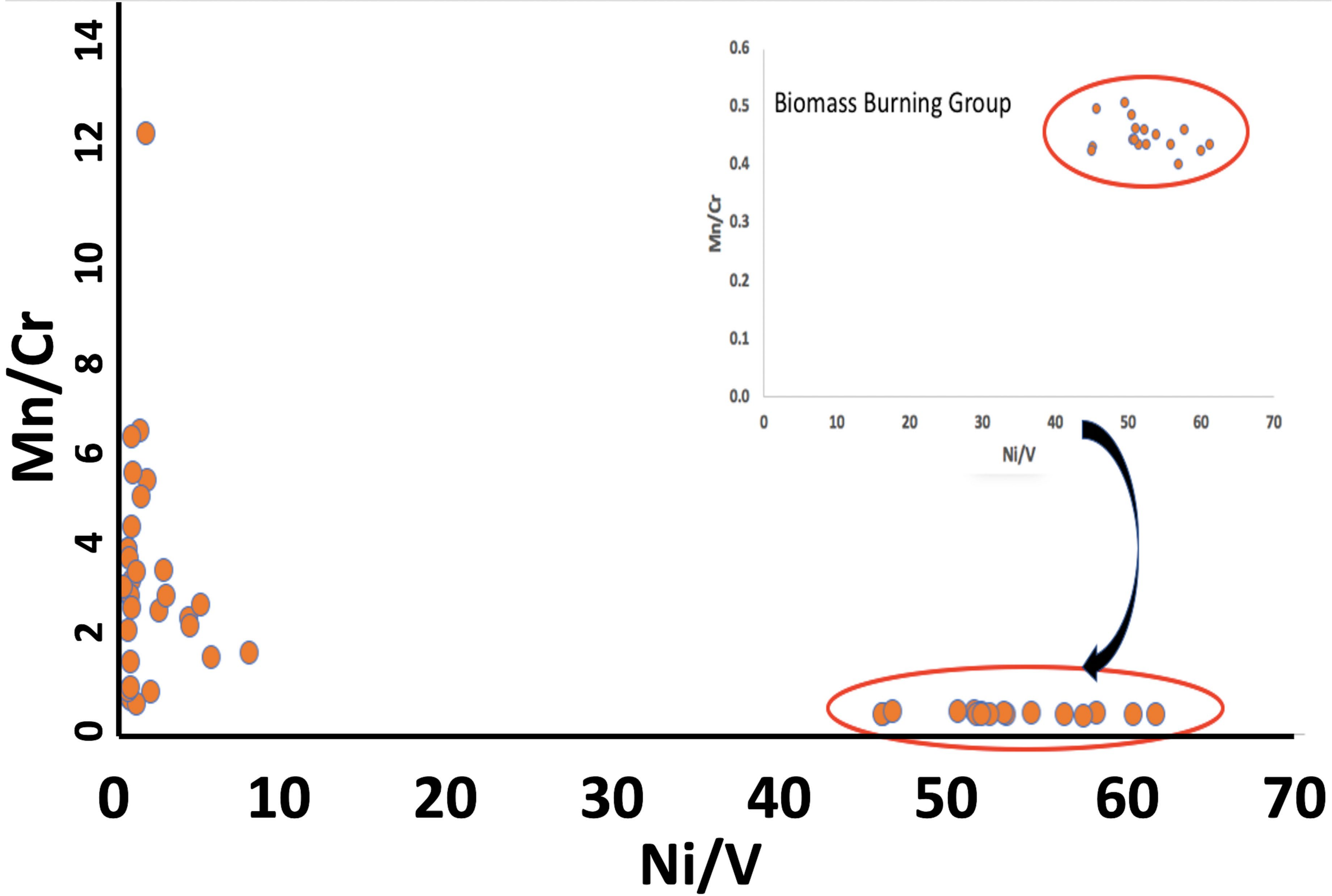


PM$_{10}$ collected at Hat-Yai (Pongpiachan et al., 2013)
y = 0.142x - 0.544
R^2 = 0.40
EC [g kg^{-1}]
EC [μg m^{-3}]
EC [μg m^{-3}]
OC [μg m^{-3}]

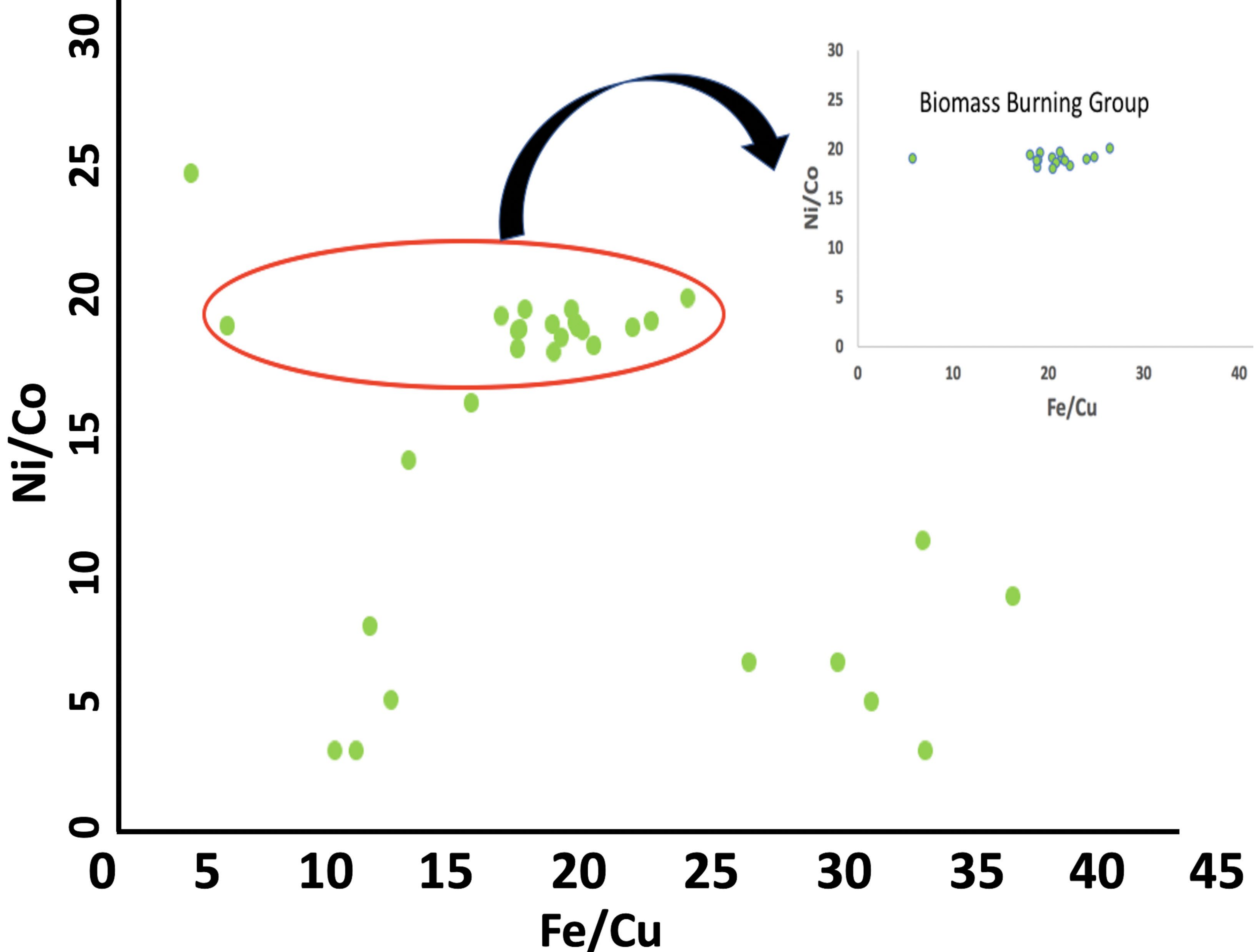
Application of Metal Ratios for Assesing the Impacts of Biomass Burning in the Ambient Air of Cities around the World



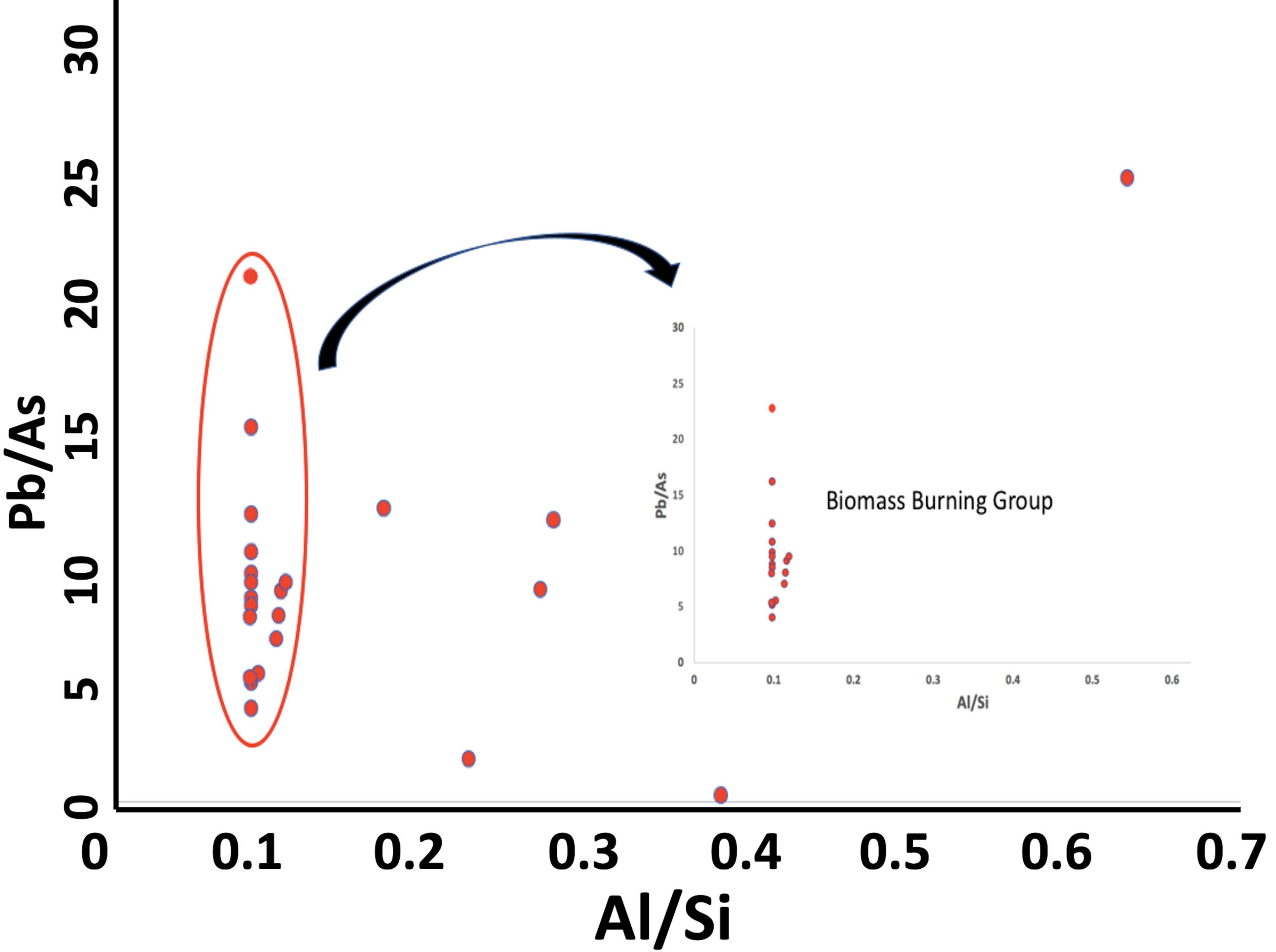
Zn/Mn
70
60
50
40
30
20
10
0
0
10
20
30
40
Fe/Zn
Biomass Burning Group
25
20
15
10
5
0
0.0
0.5
1.0
1.5
Zn/Mn
Fe/Zn



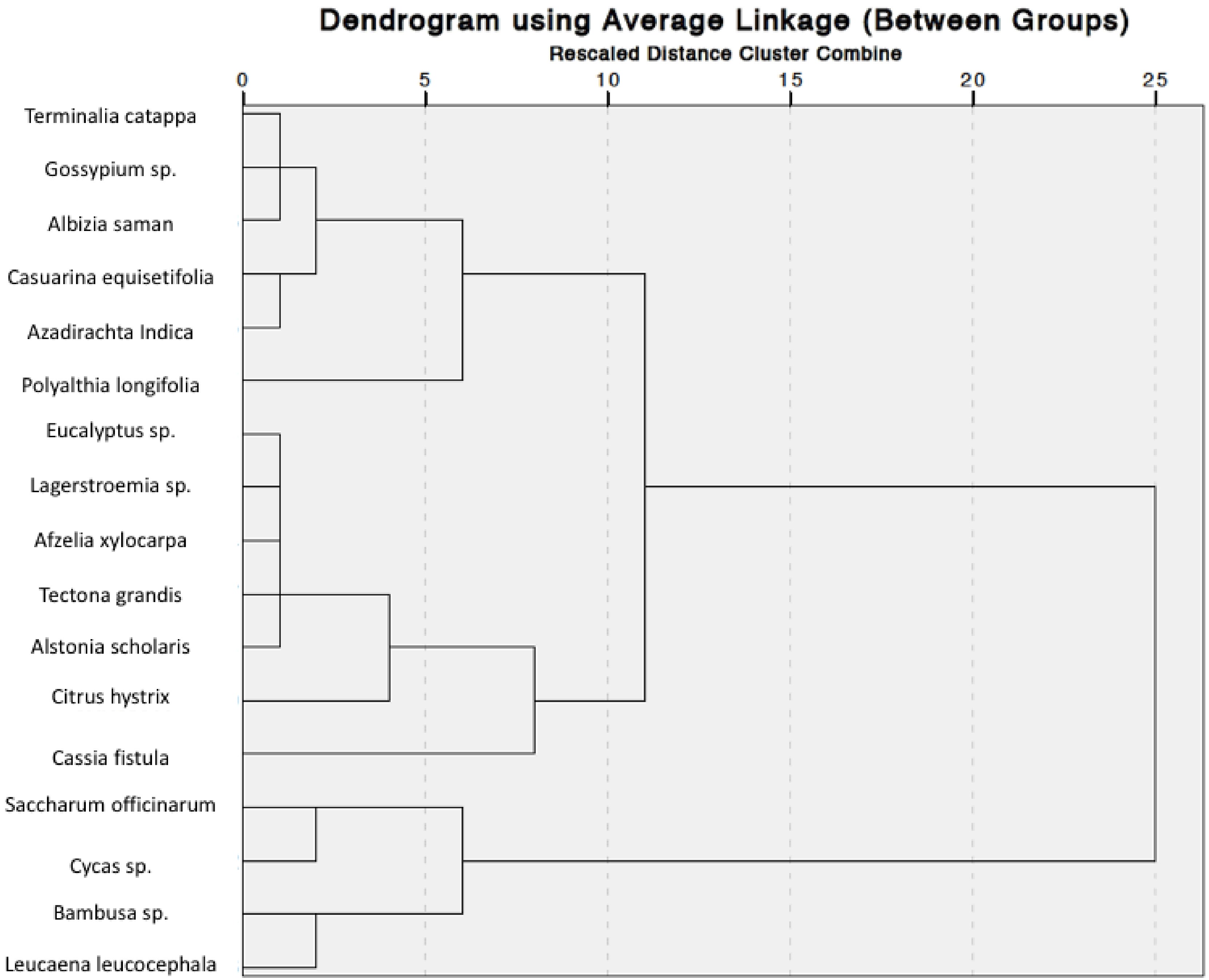
Mn/Cr
0
2
4
6
8
10
12
14
Ni/V
0
10
20
30
40
50
60
70
Biomass Burning Group
0.0
0.1
0.2
0.3
0.4
0.5
0.6



Ni/Co
Fe/Cu
0
5
10
15
20
25
30
35
40
45
Biomass Burning Group



Pb/As
Al/Si
0
5
10
15
20
25
30
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
Biomass Burning Group
Pb/As
Al/Si



Input Parameters

Al, Si, S, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Sr, Ba, and Pb

OC1, OC2, OC3, and OC4
EC1, EC2, and EC3

Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_2^-, NO_3^-, SO_4^{2-}

Conclusions

Based on three parallel channels situated downstream of the residence chamber of the dilution sampler, *EFs* of 32 chemical species (i.e. WSIS, OC, EC, and selected metals) were detected for 17 plant species.

The highest *EFs* of all chemical species for *Saccharum officinarum* (sugarcane) were obtained, followed by *Cycas sp.* (cycad), *Bambusa sp.* (bamboo), and *Citrus hystrix* (kaffir lime). In general, Si, Al, Zn, Fe, and Ni are five metals with comparatively high *EFs* for all plant species.

Conclusions

Sugarcane burning has the greatest EFs of highly toxic metals such as V, Cr, Co, Ni, and Pb; millers who purchase large amounts of burnt sugarcane should be fined.

That some plant species show a comparatively high percentage contribution of Cl^- raises some concerns over the application of this WSIS as a geochemical tracer of maritime aerosols.

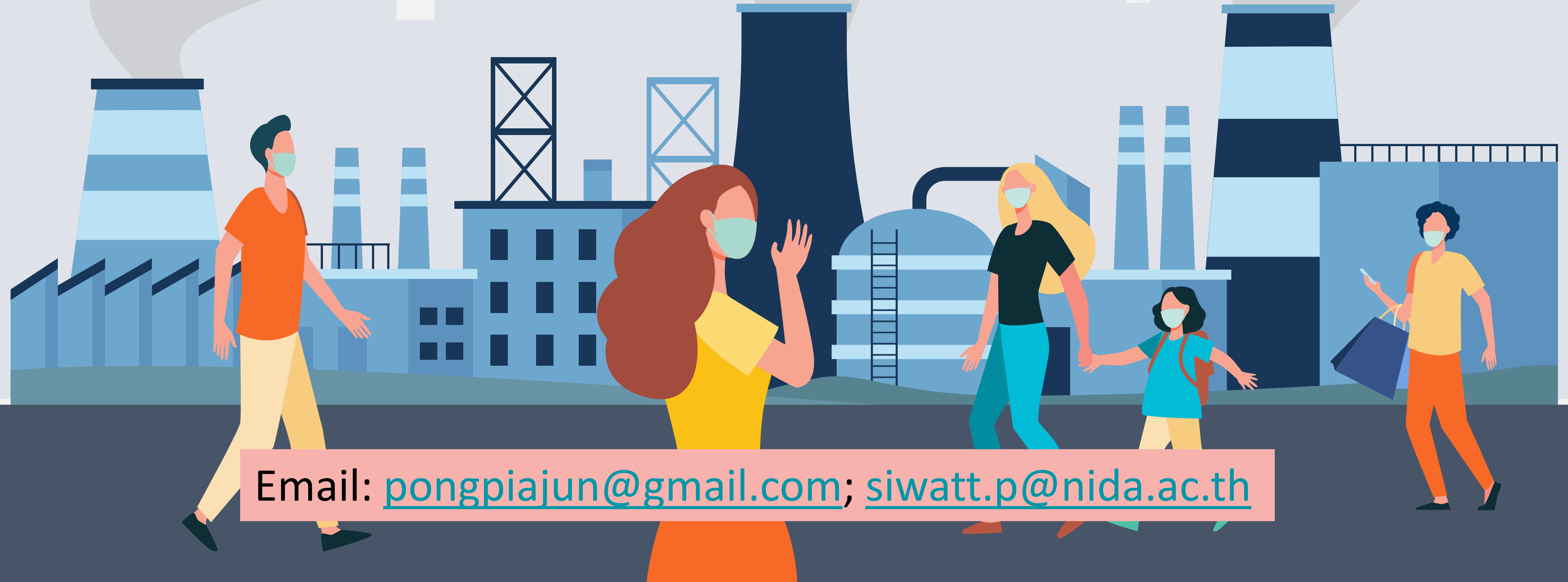
Furthermore, that the K^+ contents fell below detection limits in the case of *Alstonia scholaris*, *Cassia fistula*, *Gossypium sp*., and *Tectona grandis* suggests that the traditional protocol of employing K^+ as the sole tracer of agricultural waste burning for source identification could be questionable.

Conclusions

The relatively high OC/EC ratios observed in this study highlight the importance of biomass burning as a main contributor to carbonaceous aerosols in some regions. In addition, the comparatively high R^2 values of OC *versus* EC measured in Chiang Mai reflected an overwhelming of similar sources or by other contributors having similar OC/EC ratios, particularly during the haze episode.

Acknowledgement

This work was performed with the approval of Ministry of Higher Education, Science, Research and Innovation, Thailand and the Strategic Priority Research Program of Chinese Academy of Sciences (XDB40000000).



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