

268 shading), and NO_x ; (c) CO; (d) THC; (e) PM; and (f) PN EFs for each machine type under different
269 operating modes. Error bars indicate standard deviation.

270 **3.2 Speciated PM emissions**

271 The speciated PM collected and analyzed in all of the modes, and the analyzed results
272 are representing the whole operation process. The fractional mass contributions of main
273 category of various PM chemical components are shown in Fig. 3 (a); the detailed fractions
274 of speciated PM are summarized in Table 4. The PM filter for EX_2 was contaminated
275 during the sampling process, so PM data is not available for this machine. OM and EC
276 were the main PM components in the diesel construction machinery exhaust. OM
277 accounted for $41 \pm 17\%$, $52.3 \pm 15\%$, 50.9% , and 46.6% of the total PM mass for the
278 excavator, loader, bulldozer and roller, respectively, while EC contributed $47.2 \pm 8\%$, 34.5
279 $\pm 15\%$, 19.6% , and 37.1% .

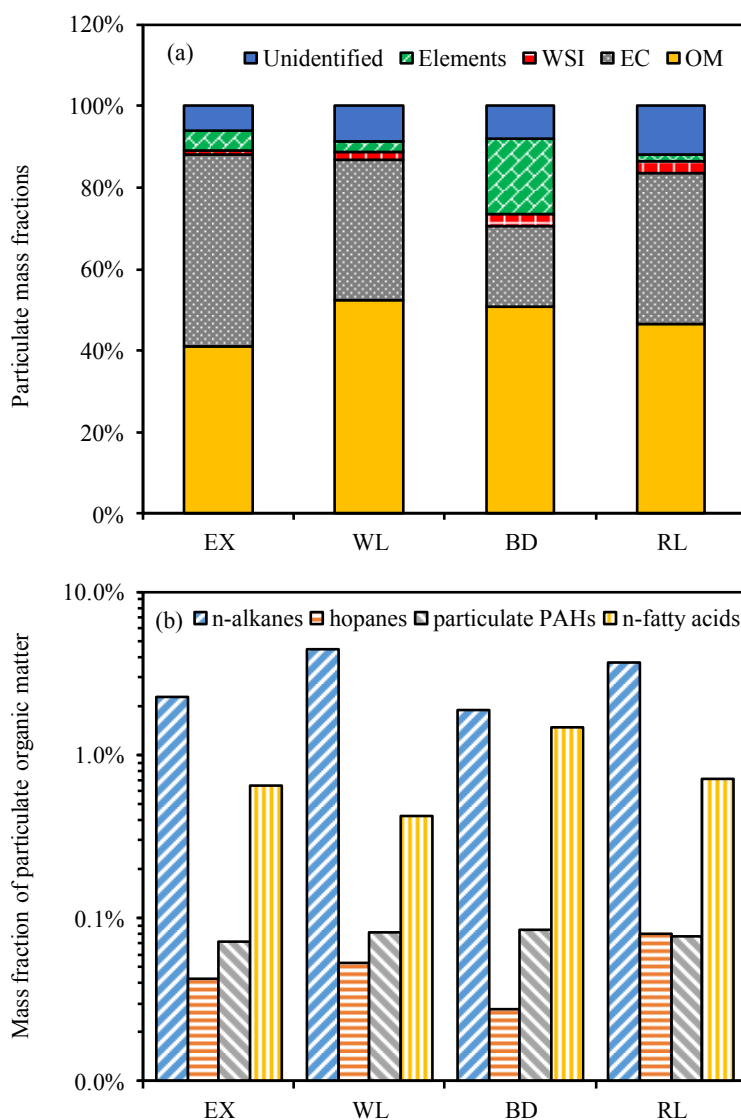
280 The contributions of WSIs to PM mass were relatively lower, with an average of $1.9 \pm$
281 1% . WSIs from the test machines consisted primarily of NO_3^- , which accounted for $1.3 \pm 1\%$
282 of the total PM mass, followed by SO_4^{2-} . These results are consistent with Ma et al. (2018).
283 However, SO_4^{2-} was the dominant contributor to PM mass, far outweighing NO_3^- , in diesel
284 vehicle and machinery exhaust in previous studies in China (Zhang et al., 2015; Cui et al.,
285 2017); these differences may be attributed to the diesel fuel sulfur content in these studies,
286 which reached up to 500 and 1300 ppm, respectively. The quality of diesel fuel has
287 improved in recent years; diesel sulfur content is currently controlled to < 50 ppm, which
288 generally reduces the fraction of SO_4^{2-} in PM emissions in diesel exhaust.

289 The elemental component fractions varied between the different types of machinery.

290 Elements constituted $4.9 \pm 2\%$, $2.6 \pm 0.9\%$, and 1.8% , respectively of the excavator, wheel
291 loader, and roller emissions; Si was the dominant elemental component, accounting for 3.5
292 $\pm 1.9\%$, $1.2 \pm 1.1\%$, and 1.1% of the total PM mass. The elemental fraction was much
293 higher for the bulldozer than for the other machine types, reaching 18.5% of the total PM
294 mass. The major elements emitted from the bulldozer were Si, Ca, Al, and Fe, which
295 accounted for 5.1% , 5.1% , 2.4% , and 1.8% , respectively, of the total PM mass. These
296 elements are generally derived primarily from diesel fuel impurities (Wang et al., 2003).
297 Thus, the inferior quality of the diesel fuel used in the test bulldozer may explain the high
298 elemental emissions. The PM SO_4^{2-} fraction emitted from the test bulldozer, which
299 measured up to 1.2% , was much higher than the SO_4^{2-} fractions emitted from the other
300 machinery types ($0.1\% - 0.3\%$).

301 The fractional contributions of various OM species to total PM organic mass are shown
302 in Fig. 3 (b). Altogether, 21 *n*-alkanes, 10 hopanes, 21 particulate PAHs, and 21 *n*-fatty
303 acids were quantified in the PM samples (see Table 4). OM species contributed
304 approximately $2.4\% - 6.4\%$ of the total PM mass. *n*-Alkanes were the dominant organic
305 component, accounting for $2.26 \pm 0.2\%$, $4.48 \pm 2\%$, 1.87% , and 3.71% of the total PM
306 mass from the excavators, wheel loaders, bulldozer, and roller, respectively; *n*-fatty acids
307 were the second most abundant category, contributing $0.65 \pm 0.4\%$, $0.42 \pm 0.4\%$, 1.47% ,
308 and 0.71% of the total PM mass. The average PM mass fractions of hopanes and particulate
309 PAHs were $0.049 \pm 0.03\%$ and $0.078 \pm 0.02\%$, respectively. Cui et al. (2017) reported
310 *n*-alkane and particulate PAH fractions reaching 5.14% and 0.098% , slightly higher than
311 the proportions found herein; this may be attributed to the use of high-sulfur fuel in that

312 study. The *n*-alkanes found herein consisted primarily of C18-C23 compounds, except for
 313 those emitted from the bulldozer, which contained more high-carbon components
 314 (C20-C24), as shown in Table 4. Hopanes can be used as organic tracers for oil combustion
 315 processes. 17a(H)21β(H)-hopane (C30H) and 17a(H)21β(H)-30-norhopane (C29H) were
 316 the most abundant hopanes, contributing $0.016 \pm 0.01\%$ and $0.009 \pm 0.004\%$ of the total
 317 PM mass, respectively, which is similar to the hopane composition of PM emissions from
 318 diesel vehicles and ships (Schauer et al., 1999; Sippula et al., 2014; Cui et al., 2017).



319
 320 **Fig. 3.** (a) Fractional contributions various species to total PM mass, and (b) fractional contributions of

321 organic matter species to total organic mass.

322 **Table 4.** Fractional chemical and organic matter of PM composition for each test machine (%).

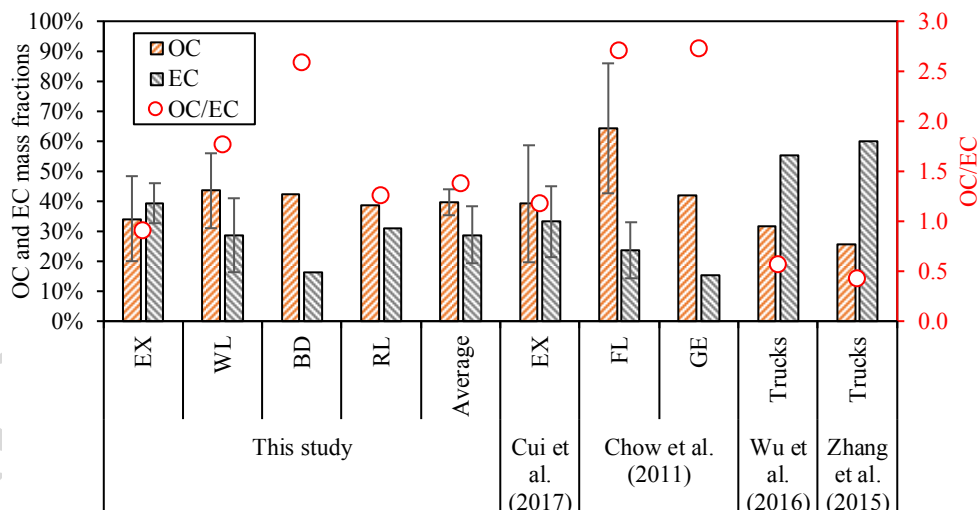
Species	EX_1	EX_3	EX_4	WL_1	WL_2	WL_3	BD_1	RL_1
OC	19.6	47.9	35	57.9	37.9	34.9	42.4	38.8
EC	47.1	36.2	34.7	19.7	43	23.6	16.4	30.9
SO ₄ ²⁻	0.1	0.2	0.2	0.3	0.1	0.2	1.2	0.3
NO ₃ ⁻	0.5	0.0	1.6	1.8	1.8	1.4	1.3	2.1
Cl ⁻	0.0	0.1	0.1	0.4	0.2	0.0	0.3	0.2
NH ₄ ⁺	0.0	0.0	0.0	0.1	0.0	0.1	0.0	0.2
ΣWSIs	0.6	0.3	1.9	2.6	2.1	1.7	2.9	2.7
Na	0.1	0.0	0.2	0.2	0.2	0.4	1.2	0.0
Mg	0.5	0.0	0.3	0.0	0.1	0.0	0.0	0.0
Al	0.8	0.5	0.8	0.4	0.5	0.0	2.4	0.3
Si	3.6	1.5	5.3	0.4	2.5	0.6	5.1	1.2
K	0.1	0.0	0.0	0.0	0.0	0.3	1.0	0.0
Ca	0.1	0.1	0.0	0.2	0.1	0.6	5.1	0.2
Ti	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0
V	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cr	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mn	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0
Fe	0.1	0.1	0.0	0.1	0.1	0.2	1.8	0.0
ΣElements	5.3	2.4	7.0	1.7	3.5	2.5	18.5	1.8
C ₁₆ H ₃₄	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
C ₁₇ H ₃₆	0.1	0.0	0.1	0.3	0.1	0.0	0.0	0.0
C ₁₈ H ₃₈	0.2	0.1	0.3	0.7	0.2	1.2	0.0	0.3
C ₁₉ H ₄₀	0.4	0.3	0.5	1.1	0.4	1.6	0.1	0.7
C ₂₀ H ₄₂	0.4	0.6	0.4	1.2	0.4	1.1	0.2	0.9
C ₂₁ H ₄₄	0.4	0.5	0.3	0.9	0.3	0.7	0.4	0.7
C ₂₂ H ₄₆	0.3	0.3	0.2	0.6	0.2	0.4	0.4	0.4
C ₂₃ H ₄₈	0.2	0.2	0.1	0.4	0.1	0.2	0.3	0.3
C ₂₄ H ₅₀	0.1	0.1	0.1	0.2	0.1	0.1	0.3	0.1
C ₂₅ H ₅₂	0.1	0.0	0.1	0.1	0.0	0.1	0.1	0.1
C ₂₆ H ₅₄	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.0
C ₂₇ H ₅₆	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₂₈ H ₅₈	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₂₉ H ₆₀	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₃₀ H ₆₂	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₃₁ H ₆₄	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₃₂ H ₆₆	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₃₃ H ₆₈	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Species	EX_1	EX_3	EX_4	WL_1	WL_2	WL_3	BD_1	RL_1
C ₃₄ H ₇₀	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₃₅ H ₇₂	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
C ₃₆ H ₇₄	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Σn-alkanes	2.5	2.2	2.2	5.5	1.9	6.0	1.9	3.7
Ts	0.002	0.002	0.005	0.004	0.002	0.009	0.002	0.005
Tm	0.002	0.001	0.004	0.003	0.002	0.006	0.002	0.005
C29H	0.006	0.006	0.015	0.009	0.006	0.014	0.005	0.010
ba-C29H	0.000	0.000	0.001	0.000	0.001	0.002	0.002	0.001
C30H	0.009	0.009	0.025	0.009	0.011	0.032	0.008	0.028
ba-C30H	0.001	0.001	0.003	0.001	0.001	0.005	0.001	0.004
C31H-S	0.003	0.003	0.006	0.005	0.000	0.011	0.003	0.010
C31H-R	0.002	0.002	0.005	0.002	0.000	0.007	0.002	0.006
C32H-S	0.002	0.002	0.004	0.002	0.000	0.009	0.002	0.006
C32H-R	0.001	0.001	0.003	0.001	0.000	0.006	0.001	0.005
Σhopanes	0.03	0.03	0.07	0.03	0.02	0.10	0.03	0.08
Phenanthrene	0.022	0.004	0.009	0.005	0.019	0.009	0.003	0.008
Anthracene	0.002	0.001	0.001	0.001	0.002	0.001	0.001	0.001
Fluoranthene	0.017	0.010	0.005	0.008	0.007	0.012	0.009	0.009
Acephenanthrene	0.004	0.002	0.001	0.002	0.002	0.003	0.001	0.002
Retene	0.002	0.002	0.001	0.002	0.001	0.002	0.001	0.001
Pyrene	0.039	0.021	0.021	0.038	0.028	0.036	0.014	0.035
Benzo[ghi]fluoranthene	0.004	0.001	0.001	0.002	0.001	0.002	0.005	0.002
Cyclopenta[cd] pyrene	0.001	0.000	0.000	0.001	0.000	0.001	0.001	0.001
Benz[a]anthracene	0.002	0.001	0.001	0.001	0.001	0.002	0.003	0.001
Chrysene	0.004	0.002	0.001	0.002	0.002	0.003	0.006	0.002
Benzo[b+k]fluoranthene	0.004	0.001	0.003	0.007	0.004	0.003	0.009	0.002
Benzo[a]fluoranthene	0.000	0.000	0.000	0.001	0.000	0.001	0.001	0.001
1,3,5-triphenylbenzene	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.001
Benzo[e]pyrene	0.003	0.001	0.001	0.002	0.003	0.003	0.006	0.002
Benzo[a]pyrene	0.002	0.001	0.002	0.002	0.000	0.002	0.003	0.002
Perylene	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000
Anthanthracene	0.001	0.000	0.000	0.001	0.000	0.002	0.003	0.001
Benzo[123-cd]pyrene	0.002	0.000	0.001	0.002	0.000	0.003	0.004	0.001
Dibenz[ah]anthracene	0.000	0.000	0.000	0.000	0.000	0.001	0.001	0.000
Benzo[ghi]perylene	0.005	0.001	0.002	0.003	0.001	0.005	0.009	0.003
Coronene	0.001	0.000	0.001	0.001	0.001	0.001	0.002	0.000
Σparticulate PAHs	0.12	0.05	0.05	0.08	0.07	0.09	0.09	0.08
Σfatty acids	0.4	1.1	0.5	0.8	0.4	0.1	1.5	0.7

323 **3.3 OC and EC emissions**

324 On average, the OC and EC measured in the test machine emissions herein contributed

325 39.3 ± 11% and 37.7 ± 13% of the total PM mass, as shown in Fig. 4. The average OC/EC
 326 ratio was 1.24 ± 0.7, which is similar to the average excavator OC/EC ratio (1.18)
 327 measured by Cui et al. (2017), lower than the ratios measured for diesel fork lifts (FL, 2.71)
 328 and generators (GE, 2.73) by Chow et al. (2011), and higher than the ratios (0.43-0.57)
 329 measured in diesel truck exhaust in China (Zhang et al., 2015; Wu et al., 2016). Previous
 330 studies have indicated that engine load affects OC and EC emissions considerably (Liu et
 331 al., 2005; Wu et al., 2016; Cui et al., 2017); OC/EC ratios are much higher under low
 332 engine load, such as during idling, than under high engine load, as OC is generated
 333 predominantly at low temperatures in fuel-rich zones. The diesel trucks evaluated by Zhang
 334 et al. (2015) and Wu et al. (2016) were operated at high speed (high load), resulting in
 335 relatively low OC/EC ratios compared with construction machinery in this and previous
 336 studies (Chow et al., 2011; Cui et al., 2017).

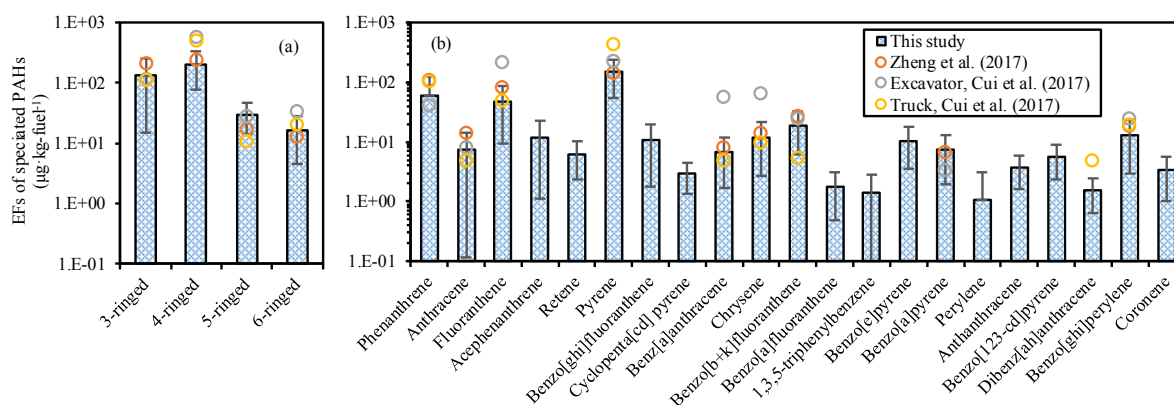


337
 338 **Fig. 4.** OC and EC mass fractions and OC/EC ratios for emissions from different types of machinery in
 339 this study and in previous studies on construction machinery and diesel trucks in China.

340 3.4 PAH emissions

341 The fuel-based EFs of the speciated PAHs were classified by the number of benzene

342 rings, as shown in Fig. 5(a). The average fuel-based EF for the test machinery was $382 \pm$
 343 $264 \text{ g kg-fuel}^{-1}$. Compounds with 3 or 4 benzene rings were dominant, accounting for 34.9%
 344 and 52.3%, respectively, of the total particulate PAHs. Pyrene, phenanthrene, and
 345 fluoranthene were the most abundant particulate PAH compounds emitted from the test
 346 machines, constituting 37.1%, 12.6%, and 12.2%, respectively, of the total PAHs. Other
 347 important compounds included benzo[b+k]fluoranthene (BbkF), benzo[ghi]perylene
 348 (BghiP), chrysene (Chr), benzo[e]pyrene (BeP), and benzo[ghi]fluoranthene (BghiF). The
 349 distributions of individual particulate PAH species, as well as compounds with given
 350 numbers of rings, were consistent with previous studies on diesel excavators (Cui et al.,
 351 2017) and trucks (Cui et al., 2017; Zheng et al., 2017) in China, as shown in Fig. 5.



352
 353 **Fig. 5.** Fuel-based EFs for speciated PAHs classified by (a) the number of benzene rings and (b) the
 354 speciated components, as compared with previous studies in China.

355 Molecular diagnostic ratios (MDRs), which consist of ratios of two individual PAHs,
 356 have been used as organic markers for various anthropogenic sources (Katsoyiannis et al.,
 357 2011) and can also be used in primary organic aerosol source apportionment (Shrivastava et
 358 al., 2007). MDRs consisting of common PAHs (e.g., Fluoranthene/(Pyrene+Fluoranthene):
 359 Flu/(Pyr+Flu); Anthracene/(Phenanthrene+Anthracene): Ant/(Phe+Ant);

360 Benzo[a]anthracene/(Chrysene + Benzo[a]anthracene): BaA/(Chr+BaA);
361 Benzo[a]pyrene/Benzo[ghi]perylene: BaP/BghiP) have been used to estimate source
362 contributions from diesel exhaust, as shown in Table 5. The overall Flu/(Pyr+Flu) ratio in
363 this study was 0.25 ± 0.07 , similar to values reported by Shah et al. (2005) and Pakbin et al.
364 (2009), but lower than those reported by Schauer et al. (1999), Cui et al. (2017), and Zheng
365 et al. (2017). Katsoyiannis et al. (2011) suggested that Flu/(Pyr+Flu) MDRs between 0.40
366 and 0.50 represent pyrogenic sources (e.g., fuel combustion). Yunker et al. (2002) and
367 Ravindra et al. (2008) found that Flu/(Pyr+Flu) MDRs of 0.39 ± 0.1 and > 0.5 , respectively,
368 could be used to distinguish diesel combustion sources. Our measurements indicate much
369 lower thresholds than those recommended by previous studies, which may introduce
370 substantial uncertainties in the use of these ratios to infer diesel combustion sources. The
371 average Ant/(Phe+Ant) and BaA/(Chr+BaA) ratios in this study were 0.12 ± 0.02 and 0.36
372 ± 0.02 , which are both similar to the recommended MDR lower limits (0.1 and 0.35) used
373 to identify fuel combustion sources in Katsoyiannis et al. (2011). The MDR results herein
374 are similar to measurements in Zheng et al. (2017), but lower than those reported by Pakbin
375 et al. (2009), Liu et al. (2015), and Cui et al. (2017). The overall BaP/BghiP ratio in this
376 study was 0.63 ± 0.5 on average, comparable to the MDR (> 0.6) suggested for traffic
377 emission sources by Katsoyiannis et al. (2011) and the range (0.5-0.6) suggested by
378 Ravindra et al. (2008). The organic markers emitted from diesel machinery and vehicles
379 varied widely with vehicle type, fuel quality, operating mode, and control technology, as
380 shown in Table 5. In addition, the markers recommended in previous studies cannot be used
381 to distinguish on-road emissions from non-road diesel exhaust. Therefore, we suggest that

Source	Vehicle type	Flu/(Pyr+Flu)	Ant/(Phe+Ant)	BaA/(Chr+BaA)	BaP/BghiP
Measured MDRs from diesel combustion					
This study	Excavator	0.27 ± 0.07	0.12 ± 0.02	0.34 ± 0.01	1.01 ± 0.5
	Wheel loader	0.21 ± 0.03	0.11 ± 0.01	0.38 ± 0.01	0.33 ± 0.3
	Bulldozer	0.38	0.15	0.35	0.30
	Roller	0.20	0.13	0.38	0.72
Schauer et al. (1999)	Diesel vehicle	0.39	0.19	0.33	N/A
Shah et al. (2005)	Diesel vehicle	0.28 (creep),	0.07 (creep),	0.53 (creep),	N/A
		0.26 (transit), and 0.26 (cruise)	0.05 (transit), and 0.03 (cruise)	0.50 (transit), and 0.51 (cruise)	
Pakbin et al. (2009)	Diesel vehicle	0.22 (UDDS)	0.20 (UDDS)	0.44 (UDDS)	N/A
		and 0.26 (cruise)	and 0.21 (cruise)	and 0.41 (cruise)	

382 organic markers be applied carefully during diesel emissions source apportionment until
383 additional real-world PAH emissions data can be obtained for on- and non-road diesel
384 vehicles.

385 **Table 5.** Molecular diagnostic ratios (MDRs) for particulate PAHs from diesel construction machinery
386 and trucks in this and previous studies.

Liu et al. (2015)	Non-road diesel engine	0.19	0.16	0.44	0.81
Cui et al. (2017)	Excavator	0.48	0.17	0.47	0.13
Zheng et al. (2017)	Diesel vehicle	0.40	0.10	0.33	0.35
Recommended MDRs for source apportionment					
Katsoyiannis et al. (2011)	Fuel combustion	0.40-0.50	> 0.1	> 0.35	
	Traffic emission				> 0.6
Yunker et al. (2002)	Diesel combustion	0.39 ± 0.1			
Ravindra et al. (2008)	Diesel combustion	> 0.5			
	Traffic emission				0.5-0.6

387 4 Conclusions

388 This study presents detailed EFs and PM chemical composition measurements from an
389 on-board emission measurement system for nine diesel construction machines in China.
390 NO_x, CO, THC, PM, and PN EFs and fractional contributions of PM components
391 (including OC, EC, WSIs, elements, and OM) were determined for each test machine under
392 real-world conditions, including idling, moving, and working. The particulate and gaseous
393 EFs were generally consistent with previous studies. The machines possessed much higher
394 FC rates, but slightly lower fuel-based EFs, when moving and working than they did while
395 idling. Overall, emissions were prominent under moving and working conditions.
396 Compared with newer machinery (under more stringent emissions standards), diesel
397 construction machines used in China produced relatively high EFs due to a lag in emissions
398 standards. OC was the most abundant particulate component, contributing 39.3 ± 11% of
399 the total PM mass, followed by EC, which accounted for 37.7 ± 13% on average. The
400 average proportions of WSIs and elements emitted from the test machines were 1.9 ± 1%
401 and 5.3 ± 6%. NO₃⁻ was the most abundant WSI, while Si dominated the elemental content.
402 Fuel quality had an adverse impact on WSI and element emissions; SO₄²⁻ and elements
403 (e.g., Si, Ca, Al, and Fe) were emitted in higher quantities from machinery fueled with
404 inferior diesel fuel. Organic matter species in PM, including *n*-alkanes, hopanes, particulate

405 PAHs, and *n*-fatty acids, contributed approximately 2.4 - 6.4% of the total PM mass.
406 Compounds with 3 or 4 aromatic rings, including pyrene, phenanthrene, and fluoranthene,
407 were dominant, accounting for 62% of the total particulate PAHs. Other major compounds
408 included benzo[b+k]fluoranthene, benzo[ghi]perylene, chrysene, benzo[e]pyrene, and
409 benzo[ghi]fluoranthene. The overall ratios of Flu/(Pyr+Flu), Ant/(Phe+Ant),
410 BaA/(Chr+BaA), and BaP/BghiP released from the test machines were 0.25 ± 0.07 , $0.12 \pm$
411 0.02 , 0.36 ± 0.02 , and 0.63 ± 0.5 , respectively. The ratio of Flu/(Pyr+Flu) was not
412 consistent with the MDRs suggested by previous studies, while the other ratios were
413 comparable to the suggested ones. More measurements are needed to identify organic
414 markers from non-road diesel machinery.

415 **Acknowledgments**

416 This work was supported by the National Key R&D Program of China (Grant No.
417 2016YFC0201501), the National Natural Science Foundation of China (Grant No.
418 21777101), the Science and Technology Commission of Shanghai Municipality Fund
419 Project (Grant No. 16dz1206704).

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