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428 three haze levels suggests that it is harmful to stay in naturally ventilated buildings during  
429 the haze period. Consequently, appropriate mitigation measures need to be developed and  
430 implemented to decrease human exposure to PM<sub>2.5</sub> mass concentrations and related health  
431 effects during haze episodes.

### 432 ***Size-fractionated PM***

433 Since the carcinogenic health risk or ELCR values estimated for PM<sub>2.5</sub> exceeded the  
434 threshold limit, ELCR values were calculated for size-fractionated PM to get further  
435 useful insights. Results obtained from this analysis for the three haze levels for total  
436 health risk are illustrated in Fig. 6. Again, an increase in health risk was observed with  
437 the increase in haze intensity. Overall, health risk due to exposure to coarse particles was  
438 found to be within the threshold limit while submicron sized particles (accumulation and  
439 q-UF) showed higher health risk than the threshold value, except for the accumulation  
440 mode during light-haze. Furthermore, it is important to note that the health risk was the  
441 highest for q-UF particles, followed by accumulation mode particles and then coarse  
442 particles. Since these submicron-sized particles are found in higher concentration during  
443 haze episodes, their effective removal indoors through mitigation methods such as air  
444 filtration is warranted. Suitable mitigation measures such as the use of suitable air  
445 cleaners indoors and respirators outdoors can be employed for effective air filtration  
446 during smoke haze episodes. However, using effective air cleaners in homes is  
447 considered to provide greater health benefits than using respirators outdoors as reported  
448 by Qi *et al.* (2017).

449 ***Uncertainty analysis for carcinogenic health risk***

450 To refine the health risk assessment and perform extreme value analysis, the uncertainty  
451 analysis was conducted with variable exposure parameters for potential carcinogenic  
452 health risk assessment for PM<sub>2.5</sub> as well as size-fractionated aerosols. For this purpose,  
453 five input exposure parameters i.e. EF, ED, AT, IR and BW were considered as variables  
454 as mentioned before in the methodology section. Moreover, only carcinogenic health risk  
455 was considered in the uncertainty analysis as it was found to exceed the threshold limit.

456 ***PM<sub>2.5</sub>***

457 The analysis was conducted for the three haze levels for PM<sub>2.5</sub> for total carcinogenic  
458 health risk values at 50<sup>th</sup> and 95<sup>th</sup> percentiles, as shown in Fig. 7. Overall, it was observed  
459 that the 50<sup>th</sup> percentile values for health risk exceeded the threshold limit by 2.7, 4.3 and  
460 5.1 times for light-haze, moderate-haze and severe-haze levels, respectively. These values  
461 are quite close to the ones obtained in previous section for PM<sub>2.5</sub> since mean values of  
462 parameters distribution were assumed to be the same as constant values assumed in an  
463 earlier section. The values calculated for the 95<sup>th</sup> percentile were found to be 4.3, 9.2 and  
464 11.5 times higher than the threshold limit; all these values are significantly higher than  
465 the allowed threshold. The values associated with the 95<sup>th</sup> percentile showed extreme  
466 values and are important for policy making. These results further indicate heightened  
467 concerns about public health impacts associated with exposure to PM<sub>2.5</sub> during smoke  
468 haze episodes.

469 ***Size-Fractionated PM***

470 In addition to  $PM_{2.5}$ , the uncertainty analysis in health risk was also conducted for size-  
471 fractionated PM, and the obtained results are shown in Fig. S14. The results showed that  
472 50<sup>th</sup> and 95<sup>th</sup> percentiles health risks were elevated by several folds for the three particles  
473 modes under the three levels of haze. Again, the patterns showed the highest health risk  
474 for q-UF particles followed by accumulation mode and then coarse mode particles. It is  
475 noteworthy that the 95<sup>th</sup> percentile health risk was observed to be higher than the  
476 threshold limit for the severe-haze case in coarse mode, indicating that coarse mode  
477 particles can also cause deleterious health effects in the case of extreme values analysis.  
478 The uncertainty analysis provides more realistic evaluation of health risk, highlighting the  
479 need for implementation of effective mitigation efforts to reduce exposure of occupants  
480 in naturally ventilated buildings to PM, especially q-UF particles during smoke haze  
481 episodes.

## 482 **CONCLUSIONS**

483 Ambient air quality in SEA was severely affected during the 2015 haze episode with a 5-  
484 fold increase in the 24 h mean  $PM_{2.5}$  concentration compared to the 24 h mean WHO  
485  $PM_{2.5}$  guidelines. Size-fractionated analysis of  $PM_{2.5}$  showed that the increment in  $PM_{2.5}$   
486 levels was mainly due to higher concentrations of quasi-ultrafine particles. The I/O ratios  
487 of size-fractionated PM showed higher values for smaller-sized particles during the haze  
488 episodes compared to larger particles. This observation indicates that exposure to such  
489 PM may pose severe health impacts since small particles contain relatively higher levels  
490 of toxic elements than coarse particles. The estimation of deposited dose of PM in the  
491 human respiratory system showed that as the particle size decreased from  $PM_{2.5}$  to

492  $PM_{<0.25}$ , the contribution from head to alveolar regions to the total deposited dose  
493 increased, suggesting that higher concentrations of ultrafine particles can lead to their  
494 deeper penetration, thereby causing harmful health impacts during haze events. Total  
495 elemental analysis was performed for the 24 selected elements. EF and PDF analyses  
496 were used to differentiate sources of PM. Results showed that Cr, Mn Fe and Ni mainly  
497 originated from crustal sources while other elements such as Cd, Cu, Pb and Zn are  
498 mainly associated with non-crustal sources. Total potential carcinogenic health risk  
499 results estimated during the severe-haze period, based on bioavailable toxic elemental  
500 concentrations in  $PM_{2.5}$ , exhibited exceedance from the threshold limit by 5.2 times for  
501 constant exposure parameters based analysis for adults. The exceedance was observed to  
502 be 11.5 times higher compared to the allowed threshold for the extreme value (95<sup>th</sup>  
503 percentile) analysis conducted for health risk considering the uncertainty or the  
504 variability associated with exposure parameters. Moreover, the results obtained for size-  
505 fractionated health risk revealed that the health risk increased with the decrease in  
506 particle size with accumulation and quasi-ultrafine particles exceeding the threshold limit.

507 Overall, it was observed that the potential health risk increases with the increase in haze  
508 intensity. However, it should be noted that the health risk exceeded from the threshold  
509 limit for the three haze levels, implying that remaining in naturally ventilated  
510 environments may not be safer during haze events. Therefore, effective mitigation of  
511 indoor exposure to PM is much needed during the occurrence of transboundary smoke  
512 haze episodes. Mitigation measures such as the use of portable air cleaners with sufficient  
513 CADR (clean air delivery rate) indoors and suitable respirators equipped with micro-  
514 ventilators in outdoor environments during haze period can help achieve good health

515 benefits. Additionally, the current study considers only toxic elements for potential health  
516 risk assessment since they make a significant contribution to the total health risk.  
517 However, organic components in PM such as polycyclic aromatic hydrocarbons (PAHs),  
518 nitrated-PAHs and quinones may also contribute toward the overall toxicity of PM and  
519 therefore should also be included in the overall health risk assessment. Consequently,  
520 more comprehensive studies focused on PM chemical speciation together with the use of  
521 simulated lung fluids containing antioxidants in place of water as the leaching agent and a  
522 comparative evaluation of various mitigation strategies are warranted to improve our  
523 understanding of the potential health effects of haze aerosols and to recommend a  
524 suitable indoor PM exposure mitigation method. Such studies will be helpful for  
525 policymakers to make appropriate decisions for maintaining good IAQ and minimizing  
526 public health impacts of haze aerosols.

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### List of Table Titles

797 **Table 1.** Total non-carcinogenic health risk estimates of PM<sub>2.5</sub> for indoors and outdoors  
798 for the three levels of haziness.

799 **Table 2.** Total carcinogenic health risk estimates of PM<sub>2.5</sub> for indoors and outdoors for  
800 the three levels of haziness.

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812 **Table 1.** Total non-carcinogenic health risk estimates for indoors and outdoors of PM<sub>2.5</sub>  
 813 for the three levels of haziness.

Element	RfD (mg kg <sup>-1</sup> day <sup>-1</sup> )	HI for PM <sub>2.5</sub>					
		Light-Haze		Moderate-Haze		Severe-Haze	
		Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
Al	1.40×10 <sup>-3</sup>	7.42×10 <sup>-5</sup>	4.97×10 <sup>-5</sup>	9.82×10 <sup>-5</sup>	6.62×10 <sup>-5</sup>	1.07×10 <sup>-4</sup>	9.47×10 <sup>-5</sup>
As	3.01×10 <sup>-4</sup>	3.26×10 <sup>-5</sup>	1.30×10 <sup>-5</sup>	5.33×10 <sup>-5</sup>	2.64×10 <sup>-5</sup>	5.45×10 <sup>-5</sup>	2.90×10 <sup>-5</sup>
Be	5.71×10 <sup>-6</sup>	1.42×10 <sup>-5</sup>	2.72×10 <sup>-6</sup>	9.77×10 <sup>-5</sup>	1.99×10 <sup>-5</sup>	1.90×10 <sup>-6</sup>	2.80×10 <sup>-6</sup>
Cd	5.71×10 <sup>-6</sup>	2.32×10 <sup>-4</sup>	7.81×10 <sup>-5</sup>	2.18×10 <sup>-4</sup>	1.03×10 <sup>-4</sup>	4.26×10 <sup>-4</sup>	3.12×10 <sup>-4</sup>
Co	5.71×10 <sup>-6</sup>	1.32×10 <sup>-4</sup>	7.46×10 <sup>-5</sup>	1.33×10 <sup>-4</sup>	7.31×10 <sup>-5</sup>	1.04×10 <sup>-4</sup>	8.37×10 <sup>-5</sup>
Cr	2.86×10 <sup>-5</sup>	1.20×10 <sup>-3</sup>	8.68×10 <sup>-4</sup>	2.02×10 <sup>-3</sup>	1.22×10 <sup>-3</sup>	2.59×10 <sup>-3</sup>	1.35×10 <sup>-3</sup>
Cu	4.02×10 <sup>-2</sup>	1.09×10 <sup>-6</sup>	6.81×10 <sup>-7</sup>	1.36×10 <sup>-6</sup>	7.67×10 <sup>-7</sup>	3.29×10 <sup>-6</sup>	2.17×10 <sup>-6</sup>
Mn	1.43×10 <sup>-5</sup>	2.99×10 <sup>-3</sup>	1.21×10 <sup>-3</sup>	4.13×10 <sup>-3</sup>	2.10×10 <sup>-3</sup>	5.05×10 <sup>-3</sup>	3.47×10 <sup>-3</sup>
Ni	2.06×10 <sup>-2</sup>	2.77×10 <sup>-6</sup>	1.62×10 <sup>-6</sup>	3.03×10 <sup>-6</sup>	1.73×10 <sup>-6</sup>	1.84×10 <sup>-6</sup>	1.61×10 <sup>-6</sup>
Pb	3.52×10 <sup>-3</sup>	2.20×10 <sup>-6</sup>	4.99×10 <sup>-7</sup>	1.73×10 <sup>-6</sup>	7.87×10 <sup>-7</sup>	2.62×10 <sup>-6</sup>	1.85×10 <sup>-6</sup>
Zn	3.00×10 <sup>-1</sup>	7.83×10 <sup>-7</sup>	3.87×10 <sup>-7</sup>	1.05×10 <sup>-6</sup>	5.16×10 <sup>-7</sup>	2.03×10 <sup>-6</sup>	1.31×10 <sup>-6</sup>
Σ=		<b>4.69×10<sup>-3</sup></b>	<b>2.30×10<sup>-3</sup></b>	<b>6.76×10<sup>-3</sup></b>	<b>3.61×10<sup>-3</sup></b>	<b>8.35×10<sup>-3</sup></b>	<b>5.35×10<sup>-3</sup></b>
<b>Total Health Risk</b>		<b>6.99×10<sup>-3</sup></b>		<b>1.04×10<sup>-2</sup></b>		<b>1.37×10<sup>-2</sup></b>	

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822 **Table 2.** Total carcinogenic health risk estimates for indoors and outdoors of PM<sub>2.5</sub> for  
 823 the three levels of haziness.

Element	SF (kg day mg <sup>-1</sup> )	ELCR for PM <sub>2.5</sub>					
		Light-Haze		Moderate-Haze		Severe-Haze	
		Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
As	15.1	1.48×10 <sup>-7</sup>	5.92×10 <sup>-8</sup>	2.42×10 <sup>-7</sup>	1.20×10 <sup>-7</sup>	2.48×10 <sup>-7</sup>	1.32×10 <sup>-7</sup>
Be	8.4	6.83×10 <sup>-10</sup>	1.30×10 <sup>-10</sup>	4.68×10 <sup>-9</sup>	9.56×10 <sup>-10</sup>	9.13×10 <sup>-11</sup>	1.34×10 <sup>-10</sup>
Cd	6.3	8.35×10 <sup>-9</sup>	2.81×10 <sup>-9</sup>	7.86×10 <sup>-9</sup>	3.70×10 <sup>-9</sup>	1.53×10 <sup>-8</sup>	1.12×10 <sup>-8</sup>
Co	31.5	2.37×10 <sup>-8</sup>	1.34×10 <sup>-8</sup>	2.39×10 <sup>-8</sup>	1.31×10 <sup>-8</sup>	1.87×10 <sup>-8</sup>	1.51×10 <sup>-8</sup>
Cr	42	1.45×10 <sup>-6</sup>	1.04×10 <sup>-6</sup>	2.43×10 <sup>-6</sup>	1.46×10 <sup>-6</sup>	3.11×10 <sup>-6</sup>	1.62×10 <sup>-6</sup>
Ni	0.84	4.79×10 <sup>-8</sup>	2.80×10 <sup>-8</sup>	5.24×10 <sup>-8</sup>	2.99×10 <sup>-8</sup>	3.18×10 <sup>-8</sup>	2.78×10 <sup>-8</sup>
Pb	0.042	3.26×10 <sup>-10</sup>	7.37×10 <sup>-11</sup>	2.56×10 <sup>-10</sup>	1.16×10 <sup>-10</sup>	3.88×10 <sup>-10</sup>	2.74×10 <sup>-10</sup>
Σ=		<b>1.67×10<sup>-6</sup></b>	<b>1.15×10<sup>-6</sup></b>	<b>2.76×10<sup>-6</sup></b>	<b>1.63×10<sup>-6</sup></b>	<b>3.43×10<sup>-6</sup></b>	<b>1.81×10<sup>-6</sup></b>
<b>Total Health Risk</b>		<b>2.82×10<sup>-6</sup></b>		<b>4.39×10<sup>-6</sup></b>		<b>5.24×10<sup>-6</sup></b>	

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### List of Figure Captions

835 **Fig. 1.** Haze map and backward trajectory analysis using HYSPLIT model for severe-  
836 haze level on 23 October 2015. The haze map is reproduced from weblink  
837 <http://asmc.asean.org/home/> (with permission from ASEAN Specialized Meteorological  
838 Centre (ASMC)).

839 **Fig. 2.**  $PM_{2.5}$  mass concentration under different levels of haziness for indoors and  
840 outdoors with its comparison to the WHO guideline. B, C, D and E are stages in PCIS  
841 which refer to  $PM_{1.0-2.5}$ ,  $PM_{0.5-1.0}$ ,  $PM_{0.25-0.5}$  and  $PM_{<0.25}$ , respectively.

842 **Fig. 3.** Indoor and outdoor PM concentrations in various sizes under different levels of  
843 haze as per PSI values. A, B, C, D and E are  $PM_{>2.5}$ ,  $PM_{1.0-2.5}$ ,  $PM_{0.5-1.0}$ ,  $PM_{0.25-0.5}$  and  
844  $PM_{<0.25}$ , respectively.

845 **Fig. 4.** Average I/O ratios in various sizes. A, B, C, D and E are  $PM_{>2.5}$ ,  $PM_{1.0-2.5}$ ,  $PM_{0.5-1.0}$ ,  
846  $PM_{0.25-0.5}$  and  $PM_{<0.25}$ , respectively.

847 **Fig. 5.** Deposited dose in the three zones of respiratory system for size-fractionated PM  
848 under three haze levels for indoors and outdoors. L, M and S refer to light-haze,  
849 moderate-haze and severe-haze levels, respectively.

850 **Fig. 6.** Total carcinogenic human health risk for indoors and outdoors for size-  
851 fractionated aerosols (coarse, accumulation and q-UF) for adults and its comparison with  
852 threshold value for different levels of haziness.

853 **Fig. 7.** Uncertainty analysis for total carcinogenic human health risk for PM<sub>2.5</sub> for indoors  
854 and outdoors and its comparison with threshold value for different levels of haziness.

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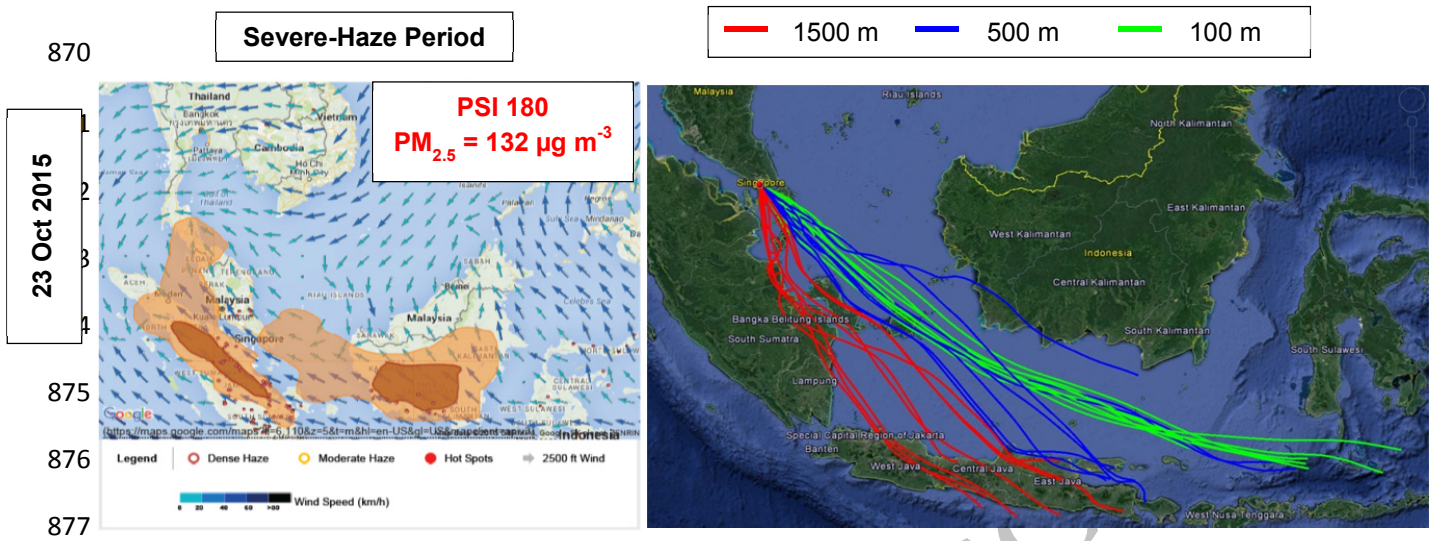
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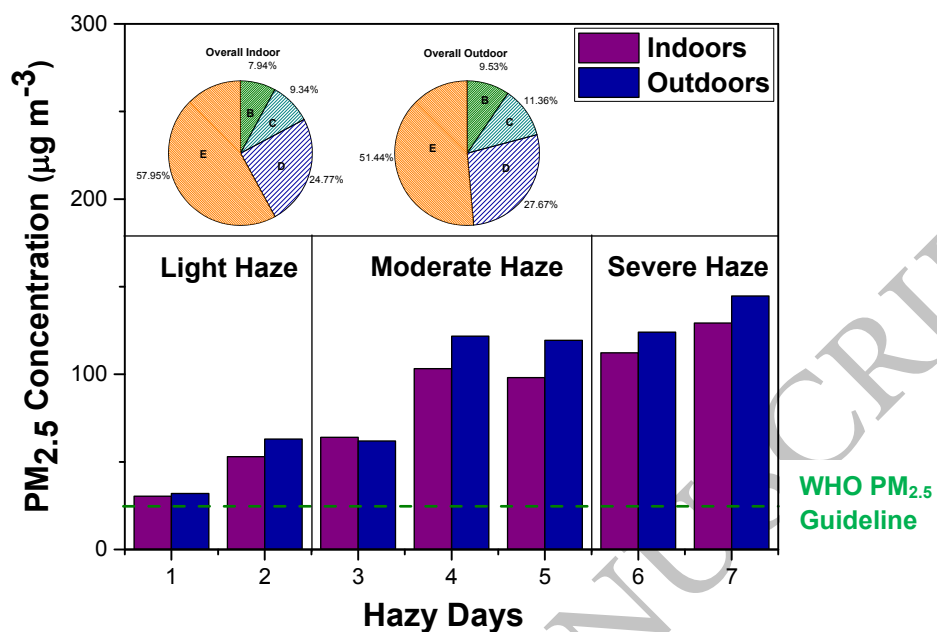
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906 **Fig. 2.** PM<sub>2.5</sub> mass concentration under different levels of haziness for indoors and  
907 outdoors with its comparison to the WHO guideline. B, C, D and E are stages in PCIS  
908 which refer to PM<sub>1.0-2.5</sub>, PM<sub>0.5-1.0</sub>, PM<sub>0.25-0.5</sub> and PM<sub><0.25</sub>, respectively.

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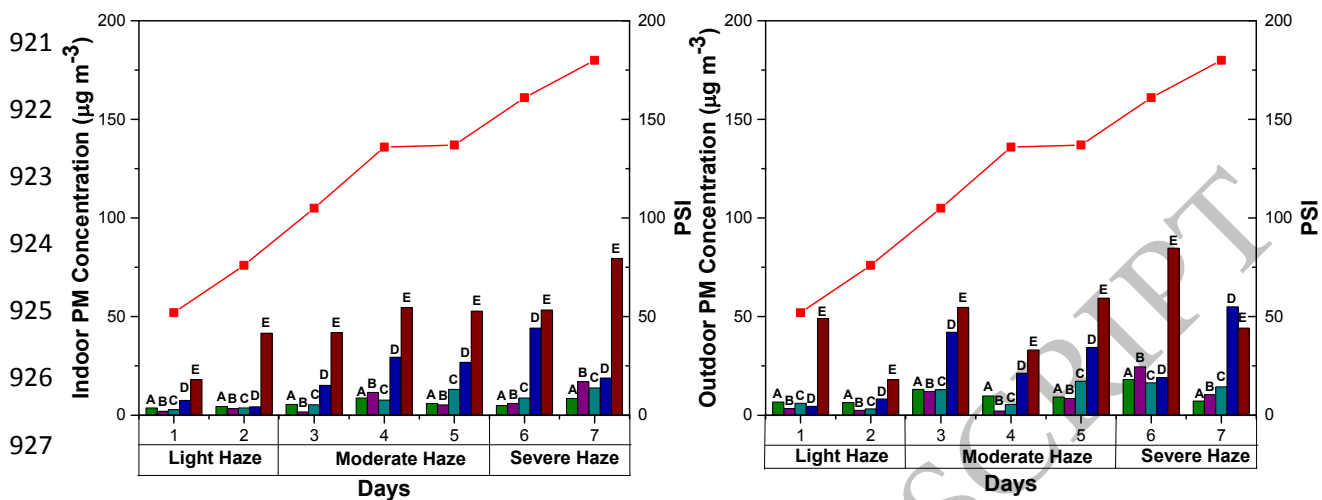
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930 haze as per PSI values. A, B, C, D and E are PM<sub>>2.5</sub>, PM<sub>1.0-2.5</sub>, PM<sub>0.5-1.0</sub>, PM<sub>0.25-0.5</sub> and  
931 PM<sub><0.25</sub>, respectively.

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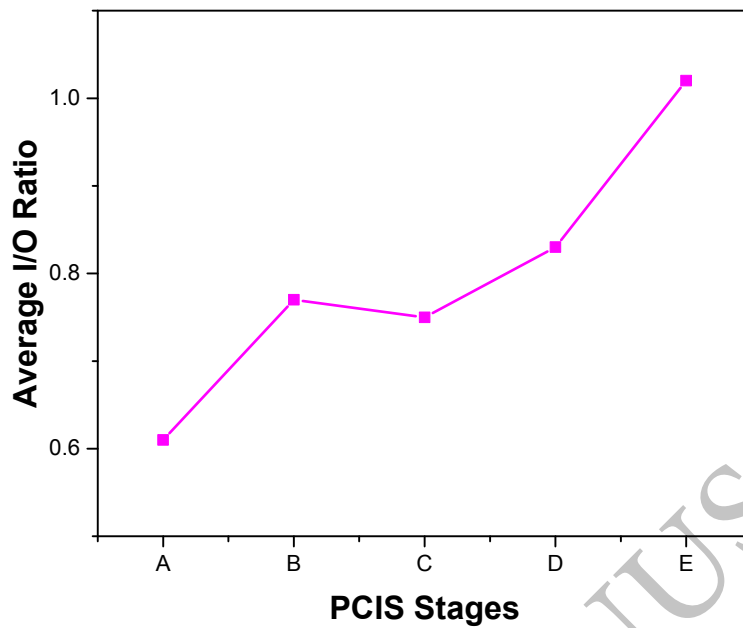
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**Fig. 4.** Average I/O ratios in various sizes. A, B, C, D and E are  $PM_{>2.5}$ ,  $PM_{1.0-2.5}$ ,  $PM_{0.5-1.0}$ ,  $PM_{0.25-0.5}$  and  $PM_{<0.25}$ , respectively.

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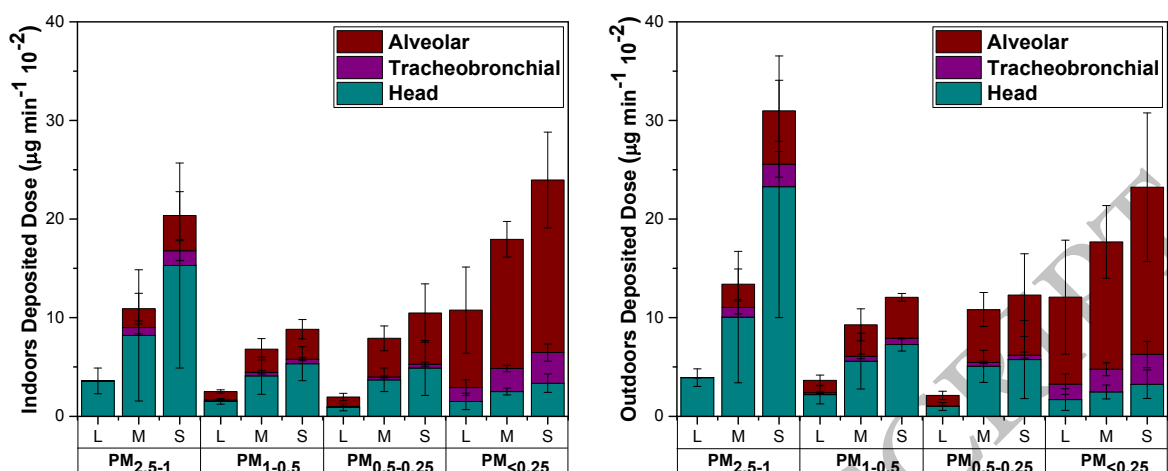
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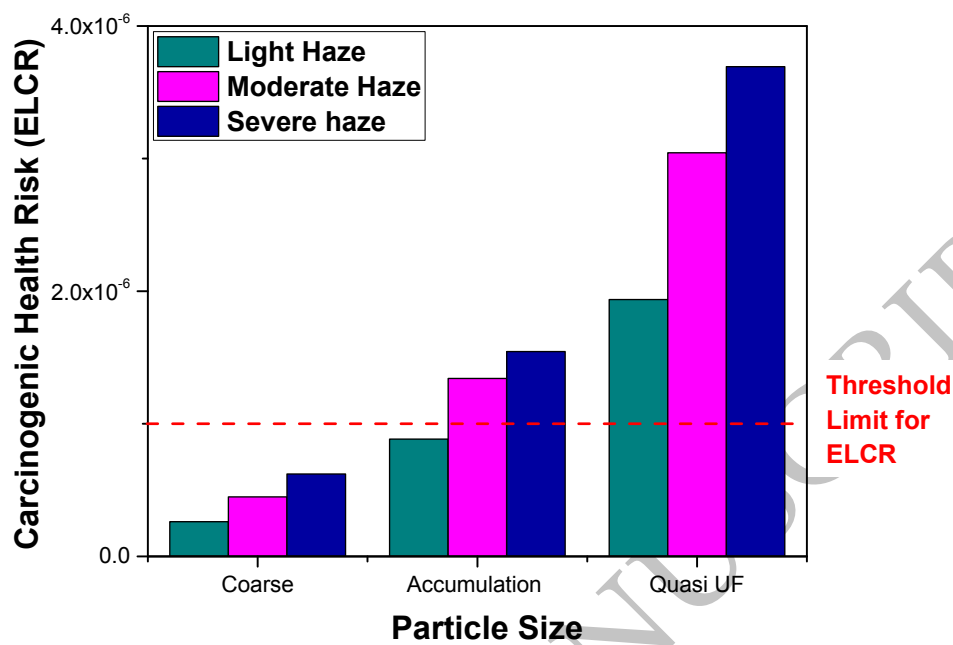
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**Fig. 6.** Total carcinogenic human health risk for indoors and outdoors for size-fractionated aerosols (coarse, accumulation and q-UF) for adults and its comparison with threshold value for different levels of haziness.



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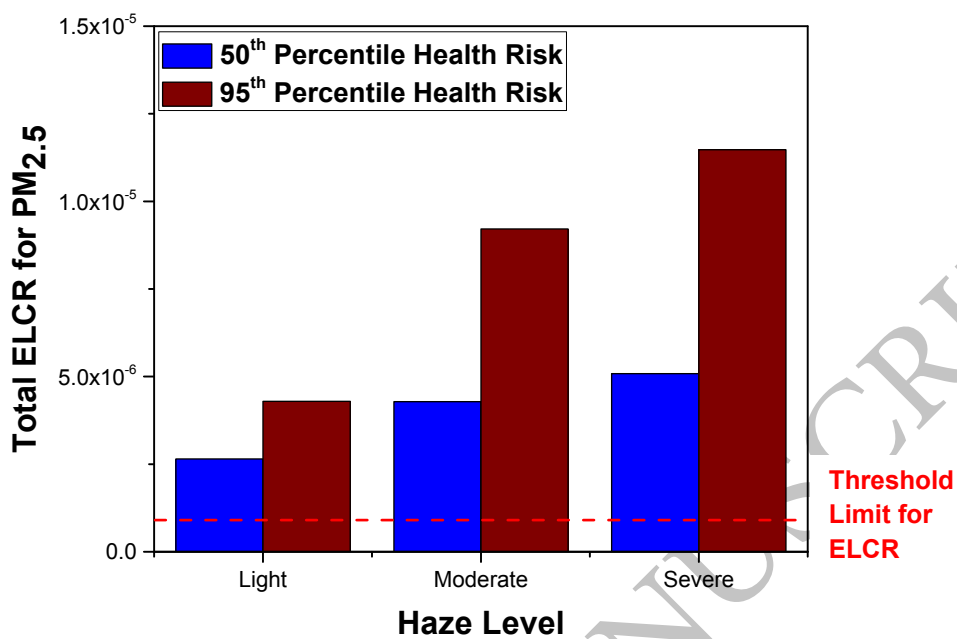
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