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Particle Size Distribution in Ambient Air of Delhi and Its Statistical Analysis

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Abstract Particle size distribution in ambient air has been studied in an urban city, Delhi. Different activity sites namely; kerbside, industrial and residential were selected for the study. The statistical analysis was carried out to study the frequency distribution and sources of different particle size fractions. The dominance of coarse particles attributed to local activities was observed at all the sites. It was observed that at kerbside sites, up to 52% of the particles were lower respiratory tract and up to 47% of the particles were upper respiratory tract particles. At residential and industrial sites, up to 40% and 31% were lower and upper respiratory tract particles, respectively. Factor analysis results indicated auto-exhaust as the dominant source of particulate matter at two of the kerbside sites. Resuspended dust was dominant at remaining two kerbside and residential sites. It was inferred using geometric standard deviation of particle size fractions that these were from different sources at residential and industrial site and from similar sources at three of the kerbside sites.

Keywords Size fraction distribution · Elemental composition · Source apportionment

The concerns about health impacts of fine particulate matter in ambient air urge for identifying its sources and variations over time and space. The increase in particulate matter levels causes serious atmospheric pollution problem that affects adversely to human health. Several studies have reported association of particulate matter and increased mortality and morbidity rates (Dockery and Pope 1994). The health effects of particulate matter mainly depend on particulate size, surface properties and composition pattern. The association has been observed to be stronger with exposure to fine particles than to coarse particles (Schwartz 1993). Several studies have therefore been initiated to examine the particle size distribution and its variation over time and space (Kikas and Tamm 1996; Lonati and Giugliano 2006). In India, particulate matter problem is significant due to the large number of vehicles plying on the road, number of power plants, combustion processes, dust storms and domestic emissions. In spite of the importance of fine particles, very few studies on particle size distribution have been reported for Indian cities (Balchandran et al. 2000; Gokhale and Patil 2004; Yadav and Rajamani 2006). In this study, the particle size distribution was therefore studied at six sites in Delhi. Different activity zones namely; kerbside, industrial and residential were considered to represent the various activities of an urban area. The statistical analysis was performed to gain insight into the frequency distribution and sources of particulates. For this, varimax rotated factor analysis was applied on the chemical composition of particulate matter of size less than 10 micron (PM10) to determine the sources at various sites in Delhi. Due to non-availability of the chemical composition of different size ranges, geometric standard deviation was used to infer the sources of particle size fractions.

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Materials and Methods

Six sampling locations namely; residential (Res), industrial (Ind) and four kerbside (KS1, KS2, KS3, KS4) were

Table 1 Description of sampling sites and respective total dust (PM10) concentration (unit: μg m^{-3})

Site	Site classification	Description	Avg	SD
1	Kerbside (KS1)	Ring road near by Maharana Pratap interstate bus terminal (ISBT)	324.9	116.9
2	Kerbside (KS2)	Ashram chowk near Mathura, road with heavy traffic of all type of vehicle	231.1	104.5
3	Kerbside (KS3)	Heavy traffics, commercial area, unpaved roads and DDA park on Loni road	1605.4	122.4
4	Kerbside (KS4)	Gazipur-karkari road near traffic signal road no 56, heavy traffic and commercial activity	161.4	101
5	Industrial (Ind)	Downwind of industrial activities and heavy traffic on GT High way	401.3	107.3
6	Residential (Res)	K.U. Black residential area with commercial, domestic and traffic activities	342.6	128

selected to carry out size fractionation study in Delhi (Table 1). The sites were named based on their prevalent activities. As high levels of pollutant concentrations were observed at traffic junctions, number of kerbside sites was more than other sites. The sampling was carried out during September 2005 to January 2006 twice in a week. Measurements were obtained using Anderson 1 ACFM NonViable Ambient sizing samplers in a eight-stage cascade impactor (Model No. 20-800, Thermo Anderson, 500 Technology Count Smyrno, GA 300082) with back-up filter (Anderson 1992). The stages have the size ranges of particulate diameter ≥9.0, 5.8–9.0, 4.7–5.8, 2.1–3.3, 1.1–2.1, 0.7–1.1, 0.4–0.7, and 0.0–0.4 μm. The sampler was operated at flow rate of 28 L min^{-1}. Glassfibre filters (Whatman Glass Microfibre, GF/A) were used as the collection substrate. During pre and post sampling, weighing of filter paper was done by equilibration procedure. The exposed filter was kept for 24 h in a desiccator. Sample filters were weighed on a Sortorius balance. The particulate concentration was then determined for each size range. Total PM10 was obtained by the addition of mass concentration of each fraction stage of the impactor**.** In order to find the inhalation pattern of particles, the particle fractions were further divided into sub categories as; <2.1 μm—lower respiratory tract; 2.1–5.8 μm—upper respiratory tract and >5.8 μm—coarse particles (Balachandran et al. 2000). For source apportionment, PM10 samples were further characterized for carbon, cations, anions and toxic metals. The samples for the metals such as Zn, Pb, Ni, Fe, Cr, Cd, Cu, B, Ba and Mn were analyzed by ICP-AES instrument (Make: Jobin Yuan, France). In the case of anions (Cl^-, SO_4^-, NO_3^-) and cations (Na^+, K^+, Ca^{++}, NH_4^+ and Mg^{++}), PM10 samples were extracted/ultrasonicated in doubled distilled water and analyzed by Ion Chromatography.

During the sample analysis, standard solutions were repeatedly aspirated to ensure calibration of the instrument within the limit of the sample concentrations for precise and accurate results (Katz 1977). Elemental Carbon (EC) and Organic Carbon (OC) were determined by Carbon Analyzer.

Results and Discussion

It can be observed from Table 2 that the total dust concentration was lower at KS4 than that at other sites. The total dust at other kerbside stations was 1.4–10 times higher than that at KS4. At Res and Ind sites, total dust concentration was 2.12 and 2.48 times higher than that at KS4. These values exceeded the standards prescribed by Central Pollution Control Board, New Delhi (60 μg m^{-3}). PM10 concentrations given in Table 2 were comparable with the levels observed in other studies e.g. 285.6–491.92 μg m^{-3} in Delhi reported by Khillare et al. (2004). At KS3 abruptly high concentration of total dust was observed, which may be due to unpaved road dust and loose soil. The mean size fraction distribution is given in Fig. 1. At KS1 and KS2, fine and coarse particles were significantly present. A bimodal size distribution was observed at KS1 and KS2 with two peaks. For KS1 the peaks were observed at 1.1–2.1 and ≥9.0 μm. For KS2, the peaks were observed at 0–0.4 and ≥9.0 μm. Fine size particle contribution (0.0–1.1 μm) was also significant at KS1 (∼35%). These two kerbside sites were traffic exposed sites with the dominance of diesel vehicles and two wheelers, which may be the major cause of presence of fine particles. At KS3, bimodal size distribution was observed with peaks at 0.7–1.1 and ≥9.0 μm. The percentage of coarse particle concentration was higher at this site, which could be attributed to presence of loose soil and unpaved road dust. At KS4, the majority of the particles were from 1.1–3.3 and ≥9.0 μm range. A bimodal frequency distribution with peaks at 1.1–2.1 and ≥9.0 μm mainly due to prevalence of soil dust was observed at this site. At Res, the maximum contribution to the total dust was from 1.1–2.1 and >4.7 μm size ranges. This site has the influence of two-wheeler traffic, domestic emissions and resuspension of road dust. Similar to Res site, the peaks were observed at 1.1–2.1 and ≥9.0 μm size at Ind site. Here the medium and coarse particles were significantly present in the total dust. In a study at a commercial site in Delhi by Balachandran et al. (2000), fine particles were reported to be dominant and at industrial and residential sites, the particle size range

Table 2 Elemental composition of PM10 (μg m^{-3}) at six sites in Delhi

Site	KS1		KS2		KS3		KS4		Ind		Res	
Variable	Avg	SD	Avg	SD	Avg	SD	Avg	SD	Avg	SD	Avg	SD
PM10	324.9	116.9	231.1	104.5	1605.4	122.4	161.4	101	401.3	107.3	342.6	128
Zn	0.036	0.003	0.030	0.004	0.025	0.003	0.018	0.002	0.014	0.002	0.046	0.004
Fe	0.074	0.009	0.042	0.008	0.021	0.002	0.048	0.006	0.018	0.002	0.025	0.004
Cr	0.006	0.001	0.003	0.001	0.003	0.002	0.008	0.001	0.008	0.001	0.014	0.002
Cu	0.015	0.002	0.012	0.001	0.011	0.002	0.020	0.003	0.009	0.001	0.012	0.001
Mn	0.006	0.001	0.025	0.005	0.006	0.001	0.006	0.001	0.013	0.002	0.016	0.002
Ni	0.004	0.002	0.004	0.002	0.005	0.002	0.005	0.003	0.007	0.004	0.020	0.008
Cd	0.005	0.003	0.005	0.003	0.004	0.002	0.009	0.002	0.004	0.001	0.009	0.001
B	0.004	0.002	0.011	0.008	0.005	0.003	0.005	0.001	0.010	0.001	0.009	0.002
Ba	0.010	0.001	0.018	0.002	0.005	0.001	0.018	0.002	0.014	0.001	0.021	0.003
Pb	0.068	0.03	0.083	0.010	0.039	0.005	0.087	0.014	0.079	0.008	0.068	0.01
EC	21.21	9.647	37.85	14.536	14.45	5.114	22.67	11.77	17.75	4.351	27.70	8.355
OC	5.211	2.485	5.550	2.174	3.850	2.277	11.83	2.746	4.650	2.498	4.250	1.074
Cl^-	8.233	4.000	7.595	4.445	10.72	5.288	11.12	1.15	13.52	5.679	15.06	6.42
SO_4^-	16.74	9.338	17.79	5.641	23.27	9.207	17.39	6.166	25.98	13.63	14.92	5.097
NO_3^-	15.13	6.994	18.11	12.429	15.89	8.206	18.63	9.80	26.04	17.61	16.34	8.828
Na^+	5.768	2.152	7.656	1.671	6.766	2.731	7.112	2.988	6.900	2.021	5.582	1.414
K^+	4.111	2.257	5.377	2.657	5.889	2.367	5.658	2.278	4.595	1.412	4.571	2.390
Ca^{++}	6.824	2.765	9.197	3.777	11.15	5.236	7.470	2.464	6.944	2.925	5.415	1.859
NH_4^+	6.067	4.431	7.192	5.652	6.273	3.957	9.328	5.005	9.370	4.995	8.771	5.548
Mg^{++}	1.305	0.714	1.396	0.640	1.931	1.151	1.515	0.548	1.367	0.656	1.024	0.482

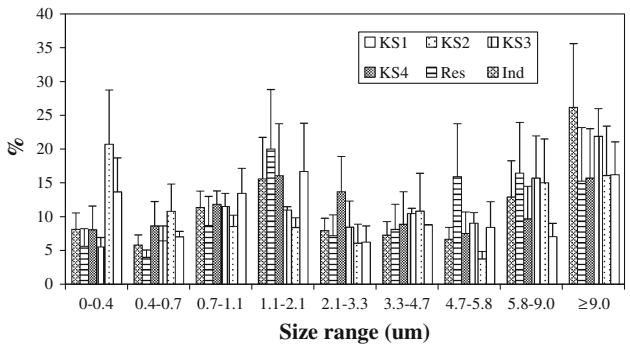

Fig. 1 Distribution of particle size fractions at six sites in Delhi

of 2.1–5.8 μm was observed to be significant. The bimodal distribution has also been reported in other studies (Kao and Friedlander 1995). The above analysis indicated that at KS1, lower respiratory tract particles (<2.1 μm) were dominating. At KS1 and KS2, approximately 52% and 47% were lower respiratory tract particles. At KS4, 25% of the particles were coarse (>5.8 μm), 31% of the particles were upper respiratory tract (2.1–5.8 μm) and 44% of the particles were in the lower respiratory tract range. At Res, 31% of the particles were coarse in nature, whereas 31% and 38% were in the upper and lower respiratory tract range, respectively. At Ind and KS3 sites, 38% of the particles were coarse. At Ind site, 40% and 22% of the particles were lower and upper respiratory tract particles, respectively. At KS3, 27% of the particles were upper respiratory tract and 35% were lower respiratory tract particles. In general at kerbside locations, up to 52% of the particles were lower respiratory tract and up to 47% of the particles were upper respiratory tract particles. At the Res and Ind sites, up to 40% and 31% were lower and upper respiratory tract particles, respectively.

The correlation between different size fractions gives an indication of similarity in the sources. The averages over three size ranges; coarse (≥5.8 μm), medium (1.1–5.8 μm) and fine (0.0–1.1 μm) were obtained and the correlation coefficient was computed between these size ranges. At KS1, KS2 and KS3, a good correlation was observed between coarse and fine particles (R^2 of 0.92, 0.94 and 0.76 respectively). An insignificant correlation ($R^2 = 0.25$) was observed between coarse and fine particles at KS4, which may be due to the influence of soil dust. At Ind site, a good correlation ($R^2 = 0.80$) was observed between fine and coarse particles. This site has the influence of coal and traffic due to its proximity to the highway and small scale industrial activities including furnace coal burning.

At Res, a good correlation ($R^2 = 0.77$) was observed between coarse and fine particles, which can be attributed

Table 3 Particle size distribution during winter and post monsoon (PM)

Site	KS1		KS2		KS3		KS4		Ind		Res	
Particle size (μm)	PM	Winter	PM	Winter	PM	Winter	PM	Winter	PM	Winter	PM	Winter
PM10	457.1	192.7	258.2	176.8	1692.0	1518.9	207.6	115.1	575.6	53.7	494.4	190.8
≥9.0	58.3	37.8	46.7	9.4	321.1	376.0	31.1	9.8	191.8	5.3	80.0	16.3
5.8–9.0	38.6	10.9	53.2	15.6	190.6	305.3	28.8	9.1	80.8	6.1	96.4	23.9
4.7–5.8	22.8	22.8	10.0	8.1	171.4	120.2	15.8	8.1	41.8	3.1	101.6	18.5
3.3–4.7	40.3	16.8	27.3	3.0	186.3	151.3	26.9	11.1	62.1	1.3	53.3	19.1
2.1–3.3	36.3	8.8	18.1	5.4	189.3	87.0	33.7	15.4	39.7	5.3	39.7	22.9
1.1–2.1	83.5	15.8	19.4	15.0	180.5	172.2	29.8	25.6	71.9	11.7	63.8	58.6
0.7–1.1	30.7	50.3	25.6	12.4	171.6	195.4	28.4	12.2	57.7	7.3	45.0	15.2
0.4–0.7	34.8	12.4	32.4	25.5	147.1	63.7	7.9	11.3	28.7	3.9	14.8	7.6
0–0.4	111.9	64.5	27.3	82.5	134.0	47.7	5.5	12.6	15.6	9.8	7.6	8.7

to the dust from domestic activities and local traffic. A good correlation was observed between coarse and medium size particles at all the sites. While analyzing the variations in particle size fractions during two seasons, it can be observed from Table 3 that the total dust concentration was higher during post monsoon at all the sites. The smaller size particle concentrations were however significant in winter at KS1, KS2, KS4 and Res sites. At KS3, the concentrations in the size range 0.0–0.7 and 1.1–5.8 μm were observed to be higher during post monsoon. It can therefore be inferred that post monsoon, more specifically the period around October was critical for particulate concentrations in all the size ranges.

For factor analysis, toxic metals (Ni, Ca^{++}, Pb, Cr, Cu, Cd, Zn, B, Ba, Mn, Fe, Mg^{++}), ions (NH_4^+, SO_4^-, NO_3^-, K^+, Na^+, Cl^-), elemental and organic carbon (EC and OC) were used. Varimax rotation technique was used to obtain the factor loading matrix. The components with eigen value greater than 1 were retained (Thurston and Spengler 1985). The average concentrations of each species over the study period are presented in Table 2. The elemental species were observed to be in the order Fe > Pb > Zn > Cu > Ba > Cr = Mn > Cd > Ni = B at KS1, Pb > Fe > Zn > Mn > Ba > Cu > B>Cd > Ni > Cr at KS2, Pb > Zn > Fe > Cu > Mn > Ni = B = Ba > Cd > Cr at KS3, Pb > Fe > Zn = Ba > Cu > Cd > Cr > Mn > Ni = B at KS4, Pb > Fe > Zn > Ba > Mn > B>Cu > Cr > Ni > Cd at Ind and Pb > Zn > Fe > Ba > Ni > Mn > Cr > Cu > Cd = B at Res site. The anions were observed in the order $SO_4^- > NO_3^- > Cl^-$ at KS1 and KS3; $NO_3^- > SO_4^- > Cl^-$ at KS2, KS4 and Ind; and $NO_3^- > Cl^- > SO_4^-$ at Res site. The cations were in the order, $Ca^{++} > NH_4^+ > Na^+ > K^+ > Mg^{++}$ at KS1, $Ca^{++} > Na^+ > NH_4^+ > K^+ > Mg^{++}$ at KS2 and KS3; $NH_4^+ > Ca^{++} > Na^+ > K^+ > Mg^{++}$ at KS4 and Ind; and $NH_4^+ > Na^+ > Ca^{++} > K^+ > Mg^{++}$ at Res site.

Factor analysis was performed using STATISTICA Version 5. The varimax rotated factor loadings are given in Table 4. At KS1, four factors together accounted for 86.6% of the total variance. The high loadings for EC, OC and Zn in the first factor indicated the significance of auto-exhaust emissions. The second factor correlated well with industrial species, whereas the third factor was correlated with dust originated variables. High loadings for Zn and Pb in factor 4 may be due to refuse burning at this site. At KS2, three factors together accounted for 76.82% of the total variance. The first factor which has high loadings for resuspension of road dust originated variables accounted for 44% of total variance. Auto-exhaust originated variables dominated at factor 2, whereas industrial contribution was significant in factor 3. At KS3, five factors together accounted for 79.35% of the total variance. The high loadings for Fe, Cr, Mn, K^+ showed the dominance of resuspension of road dust contribution at this site. The high correlation of factor 2 with EC and OC indicated auto-exhaust emissions, whereas industrial emission variables dominated factor 3. Factor 4 correlated well with Zn, mainly due to refuse burning (Mamame 1988). At KS4, three factors together accounted for 82.02% of the total variance. Factor 1 correlated well with EC and OC, which may be attributed to auto-exhaust emissions. Factor 2 was correlated with industrial originated species. Factor 3 has high loadings for Fe, Cu, Mn and Ni, which can be attributed to the contributions from resuspension of road dust.

At Ind, three factors together accounted for 79.15% of the total variance. At this site, industrial contributions were most dominant as can be seen from Table 4. Resuspension of road dust and vehicular emissions were other sources of PM10 at this site. At Res site, four factors together accounted for 77.25% of the total variance. Factor 1 has high loadings for resuspension of road dust emission species, which accounted for ~27% of the total variance.

Table 4 Varimax rotated factor loading matrix

Species	KS1				KS2			KS3				KS4			Ind			Res			
	F1 AE	F2 Ind	F3 RD	F4 RB	F1 RD	F2 AE	F3 Ind	F1 RD	F2 AE	F3 Ind	F4 RB	F1 AE	F2 Ind	F3 RD	F1 Ind	F2 RD	F3 AE	F1 RD	F2 AE	F3 Ind	F4 SAF
Zn	0.62			0.56	0.75						0.82		0.76			0.56			0.55		
Fe			0.93		0.51			0.86						0.66		0.60		0.69			
Cr			0.88				0.78	0.86					0.93								
Cu	0.86					0.73			0.71					0.91	0.84					0.93	
Mn			0.91		0.84			0.85						0.86		0.86		0.71			
Ni		0.87					0.83		0.94					0.84	0.89				0.57		
Cd		0.90					0.76		0.94				0.9			0.76		0.75			
B		0.73				0.80		0.57					0.7	0.61	0.93					0.93	
Ba		0.85					0.76			0.8			0.56	0.74	0.63			0.83			
Pb				0.86		0.78		0.57	0.56				0.81		0.54					0.92	
EC	0.87					0.62			0.60			0.60					0.80		0.60		
OC	0.65					0.75			0.54			0.82					0.80		0.87		
Cl^-	0.84				0.99			0.81				0.92			0.91				0.88		
SO_4^-	0.49	0.74			0.92			0.87				0.76				0.64			0.51		0.73
NO_3^-	0.74				0.87			0.92				0.98			0.92				0.84		
Na^+		0.95			0.98			0.64				0.79			0.94						0.77
K^+	0.78				0.99			0.53		0.54		0.92			0.55	0.62					
Ca^{++}	0.63	0.52			0.99			0.80				0.49	0.47	0.45		0.80		0.90			
NH_4^+	0.9								0.51					0.83		0.55					0.86
Mg^{++}		0.73			0.98			0.79					0.49	0.58		0.89		0.91			
% Var	42.4	23.3	10.7	10.2	44.1	17.8	14.9	42.5	25.1	5.9	5.7	28.9	28.06	25.06	46.2	22.8	10.2	26.9	23.7	15.9	10.8

AE, auto-exhaust, *Ind*, industry, *RD*, resuspension of road dust, *RB*, refuse burning, *SAF*, secondary aerosol formation, *Var*, variance

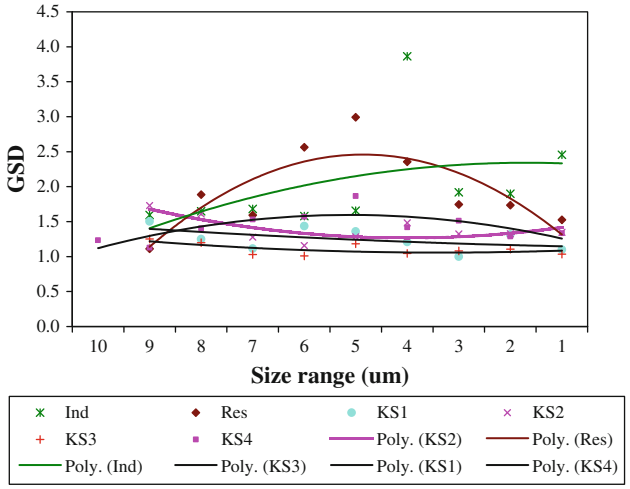

Fig. 2 Geometric Standard Deviation (GSD) of various size ranges—Poly indicates fitted polynomial curve

Factor 2 correlated well with auto-exhaust emission species such as EC and OC. Factor 3 was mainly due to the emissions from industries, whereas secondary aerosol formation species were significant in factor 4. In general, it can be observed from Table 4 that at KS1 and KS4, auto-exhaust contributed most for PM10. At KS2, KS3 and Res site resuspended dust was the dominant source, whereas at Ind site, industrial contribution was dominant. The other major sources at this site were auto-exhaust and resuspended road dust. Secondary aerosol formation contributed in a smaller fraction ($\sim$11%) at Res site. Industrial contribution was also present at kerbside and residential sites. The findings compared well with the other studies e.g. by Khillare et al. (2004), where vehicular traffic and industrial emissions were observed to be the significant contributors of particulate matter at residential, industrial and traffic exposed areas of Delhi. In another study at a residential site in Delhi by Sharma et al. (2003), motor vehicle exhaust emissions and biomass and/or refuse burning were observed to be significant contributors to organic fraction of PM10.

Geometric Standard Deviation (GSD) as suggested by Kikas and Tamm (1996) was used to infer the sources of different particulate size ranges. The polynomial lines were fitted to GSDs at various sites to examine their variations with different particle size fractions, which would help in inferring the similarity or differences in the sources. It can be observed from Fig. 2 that polynomial trend lines have a concave shape for KS1, KS2 and KS3 stations and convex shape for KS4, Ind and Res. Kikas and Tamm (1996) observed the concave shape for urban sites and convex shape of polynomials for rural sites. In the present study, it was observed that KS4, Ind and Res sites form one group and remaining sites form another group. The different shapes of polynomials indicated the presence of different processes controlling the size distribution of particles.

Considering the GSD of various size ranges, it can be seen that GSD was approximately similar for all the size ranges at KS1, KS2 and KS3 whereas at Res, it was similar for 0.0–1.1 and $\geq$4.7 μm. Another cluster was observed for the size range 1.1–4.7 μm. At KS4, two groups in the range 0.7–9.0 and 0.0–0.7 μm were observed with similar GSD. At Ind site, highest GSD was observed for the size range 3.3–4.7 μm followed by the smaller peak of $\geq$9.0 μm. This indicated that various particle size fractions at KS1, KS2 and KS3 have similar sources and at Res, KS4 and Ind sites, the sources were different for different size fractions.

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