OBSERVATIONS OF AITKEN NUCLEI AND TRACE GASES IN DIFFERENT ENVIRONMENTS IN INDIA

L. T. KHEMANI, G. A. MOMIN, MEDHA S. NAIK, R. KUMAR, and BH. V. RAMANA MURTY

Indian Institute of Tropical Meteorology, Pune 411 005, India

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Abstract. Surface measurements of Aitken nuclei have been made at a few representative environments in India. The periods of measurements have ranged from a few days to a few years depending upon the place of measurement. Aircraft measurements of Aitken nuclei were made at one of the locations during three successive monsoon periods. During part of the time simultaneous measurements were made of the trace gases SO_2, NH_3, NO_2, and O_3. The study presents the seasonal and diurnal variation of Aitken nuclei in the different environments and their association with the trace gas concentrations.

1. Introduction

Aitken nuclei (A.N) are in the size range 0.001 to 0.1 μm radius and are formed predominantly by conversion of the gas phase of certain natural trace constituents in the troposphere. Recent observations in filtered air have shown that even trace levels of certain gases can give rise to A.N by a variety of possible chemical reactions. They are removed from the atmosphere through a complex chain of processes including precipitation and deposition. Their concentration in air varies from 10 to 10^7 cm^{-3}. They play a vital role in several atmospheric processes, such as electricity, radiation balance and chemistry, and cloud phenomena. These nuclei continue to be of interest because their nature and origin remain obscure (Bigg and Turvey, 1978).

The variation of A.N concentration between air masses from continents and oceans is very large (Jaenicke *et al.*, 1971; Hogan *et al.*, 1973). The concentration over oceans has been studied more extensively than over land. The median concentration over mid-oceans has been reported in the range of 100 to 300 cm^{-3} (Blanchard and Syzdek, 1972). Extensive measurements made in Australia indicated that the median concentration in the well-mixed lower layer of continental air masses averaged only 680 cm^{-3} (Bigg and Turvey, 1978); however, this value is far lower than that found by other observers (Landsberg, 1938; Selezneva, 1966; Went, 1966; Mohnen and Hogan, 1977) over tropical lands where the concentrations varied widely and have exceeded 1000 cm^{-3}.

Since observations of A.N over the Indian sub-continent were scanty, measurements were made at a few representative environments in India during 1980-83. Also, simultaneous measurements were made of the trace gases SO_2, NH_3, NO_2, and O_3. The results of the study of the seasonal and diurnal variation of A.N in different environments and their relationship with trace gas concentrations are presented here.

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2. Observational Sites

The locations of the observational sites are shown in Figure 1.

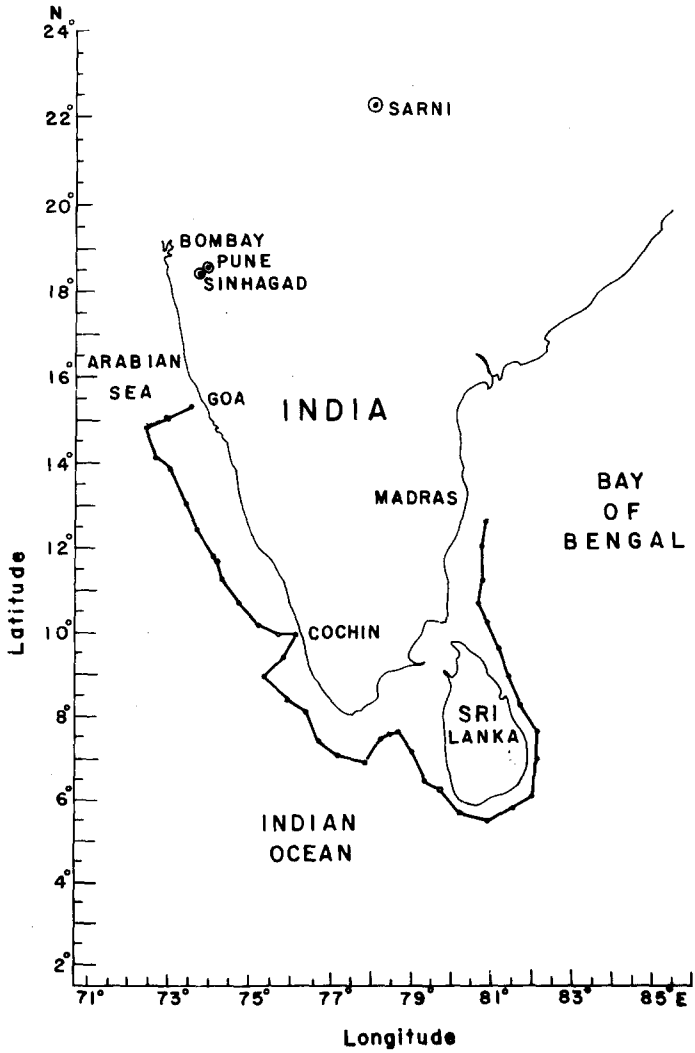

Fig. 1. Map showing the route followed by the research vessel 'Gaveshani' in the Arabian sea, Indian ocean and the Bay of Bengal regions and the locations of the observational sites.

2.1. PUNE (18° 32′ N, 73° 51′ E, 559 m amsl)

This urban environment is about 100 km from the west coast and is located on the lee-side of the western ghats. The air flow in the lower troposphere is predominantly westerly during the summer monsoon (June-September) when there is a large influx of moisture from the Arabian sea. The wind in the lower troposphere reverses with the

withdrawal of the monsoon and an easterly flow sets in from October and continental air masses, rich in nuclei of continental origin, pass over the region.

2.2. SINHAGAD (18° 21′ N, 73° 45′ E, 1313 m amsl)

This non-urban environment is located about 18 km in the south-west direction from Pune City. It is in the forest region of Maharashtra State and is sparsely populated.

2.3. SARNI (22° 07′ N, 78° 10′ E, 436 m amsl)

This thermal power plant environment is well inland in the forest area of Madhya Pradesh State of India. A 722.5 MW thermal power plant has been operating at this place without electrostatic precipitators.

2.4. SEA REGIONS

Observations over the sea have been made on board the research vessel 'Gaveshani' of the National Institute of Oceanography, Panjim, India. The cruise of the vessel in the sea regions around the Indian sub-continent is shown in Figure 1.

3. Observations

A portable expansion type counter (Rich, 1955) is used to measure A.N concentration in the size range 0.001 to 0.1 μm. For each observation, 3 to 4 readings are taken. The readings are repeatable and no significant variations have been noticed between the readings.

The nuclei counter is volume controlled and can be used for aircraft measurements in the lower atmosphere (below 800 mb). At higher levels the nuclei concentration measured by the counter is not absolute but is only indicative (Nolan and Scott, 1964). Trace gases (SO_2, NH_3, NO_2, and O_3) were measured by wet-scrubbing method (Khemani *et al.*, 1980).

Surface observations at Pune have been conducted from the terrace of the Institute's building at a height of about 12 m above the ground level during the period June 1980-May 1982. Also, simultaneous surface observations of trace gases (SO_2, NH_3, NO_2, and O_3) and A.N were made during the period March 3 to 13, 1981. Aircraft observations up to 3.3 km were made in this region during the monsoon period (June-September) in the three years 1980, 1981 and 1982.

Continuous observations on A.N and trace gases were made at Sinhagad for a period of seven days in January 1982. At Sarni, observations were made for a period of 15 days in May 1982.

Over the sea regions, continuous observations were made for a period of 10 days in May 1983.

TABLE I

Concentrations of Aitken nuclei ($N\ cm^{-3}$) and trace gases ($\mu g\ m^{-3}$) at different locations in India

Aitken nuclei / Trace gases	PUNE			SARNI			SINHAGAD			SEA REGION					
										Arabian Sea			Bay of Bengal		
	Min	Ave	Max	Min	Ave	Max	Min	Ave	Max	Min	Ave	Max	Min	Ave	Max
A.N	1300	28900	90000	1500	8800	50000	300	690	1500	300	460	600	500	730	1050
SO_2	0.5	5.5	20.5	0.6	14.1	148.7	0.7	1.2	1.5	0.6	2.6	3.8	2.5	3.2	4.4
NH_3	0.8	2.0	4.2	0.2	1.2	2.9	0.1	0.6	0.9	0.8	1.6	2.9	1.4	1.9	2.5
NO_2	0.6	1.6	5.3	1.1	3.8	12.1	0.7	2.9	3.3	0.6	1.3	3.9	1.0	1.3	1.6
O_3	0.2	13.8	29.3	34.3	38.7	79.2	40.0	93.2	154.8	–	–	–	–	–	–

4. Results and Discussion

4.1. Variation of A.N in different environments

The variations in the minimum, maximum and average concentrations of A.N and trace gases are given in Table I for different places. The average concentration of A.N is highest at Pune (an urban environment). The concentrations of A.N at Sarni, representing the thermal power plant environment, are less than those at Pune. This feature is attributed to coagulation of A.N with the giant particles of fly ash released from the stacks of the power plant at Sarni, which operates without electrostatic precipitators. The values of A.N at Sinhagad, a hill station representing a non-urban environment, are by and large, similar to those representing the marine environment.

The reaction products of terpenes or similar substances exuded by vegetation have been considered to be one of the most important sources for the formation of A.N (Rasmussen and Went, 1965; Davies, 1974; Ellsaesser, 1975). If this would have been the case for India, we should have recorded high concentrations at Sinhagad which is predominantly forested land. The main criterion for the low A.N concentration at this place appears to be the absence of significant population centers and pollution sources upwind rather than the nature and amount of vegetation. It is therefore postulated that the particle generation from vegetation is low at Sinhagad because of the low level of pollution sources. The terpenes and other such products exuded by vegetation may be an important source for generation of A.N in more polluted regions. Low concentration of A.N was also observed by Bigg and Turvey (1978) in predominantly forested areas in Australia.

The average concentration of A.N was 730 cm^{-3} over the Bay of Bengal and it was 460 cm^{-3} over the Arabian sea. The difference in the concentration between the above two oceanic regions was due to the difference in the air mass characteristics during the period of observation. The surface wind over Arabian sea was westerly and purely of maritime origin and hence the concentration of A.N was lower. The westerly surface wind over the Bay of Bengal crossed the Indian sub-continent and transported nuclei of continental origin. As a result higher concentration of A.N was observed over the Bay of Bengal than over the Arabian sea. Discussions on the variations of A.N over the oceans have been reported (Blanchard and Syzdek, 1972; Hogan *et al.*, 1973; Hogan, 1976; Halter and Robinson, 1977; Shives and Robinson, 1979). Maximum concentrations may be 4 or more times the minimum concentrations at any given location owing to changing synoptic scale meteorology (Hogan *et al.*, 1973).

Besides continental sources, chemical processes in the marine atmosphere (Vohra *et al.*, 1970; Dinger *et al.*, 1970) and release of particles from ocean surface (Blanchard, 1971) have been suggested as sources of A.N over oceans. Elliott *et al.* (1974) reported average value 700 cm^{-3} of A.N in the Indian ocean around the Indian sub-continent. Our recent observations have also recorded, by and large, the same order of value.

4.2. Seasonal variation

The variations in the monthly concentrations of A.N at Pune for the 2 yr period of measurement are shown in the form of histograms (Figure 2). As many as 25 observations have been made in each month. The average concentrations during the monsoon (June-September) and the Winter (October-March) season were 1.4×10^4 cm^{-3} and 4.8×10^4 cm^{-3}, respectively. The increase of 230% noticed during the winter over the monsoon is attributed to the transport of nuclei of continental origin by air masses traveling overland from the east during the winter. The lower concentrations of A.N during the monsoon period were due to the maritime character of the prevailing winds coupled with washout effects.

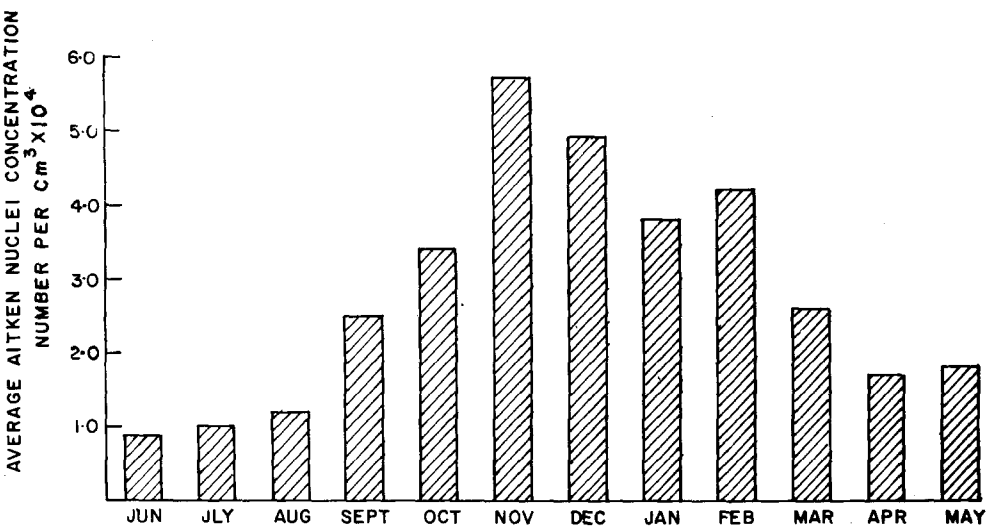

Fig. 2. The average monthly concentrations of A.N at Pune during June 1980 to May 1982.

4.3. Vertical distribution

The average vertical distribution of A.N in the cloud free air for the three monsoon seasons (1980, 1981 and 1982) is plotted in Figure 3. The data shown in the figure relate to 69 days of aircraft observations made during three summer monsoon seasons in cloud-free air. The concentration was nearly steady from 0.9 to 2.1 km and was one order of magnitude more than that at the surface. The high concentration of nuclei, which is nearly steady up to 2.1 km could be due to the thorough mixing of the layer from 0.9 to 2.1 km. As the cloud-top in the region is generally found to be around 2.1 km, the concentration decreased sharply above that level. Similar distributions were also reported by other investigators (Bigg and Turvey, 1978).

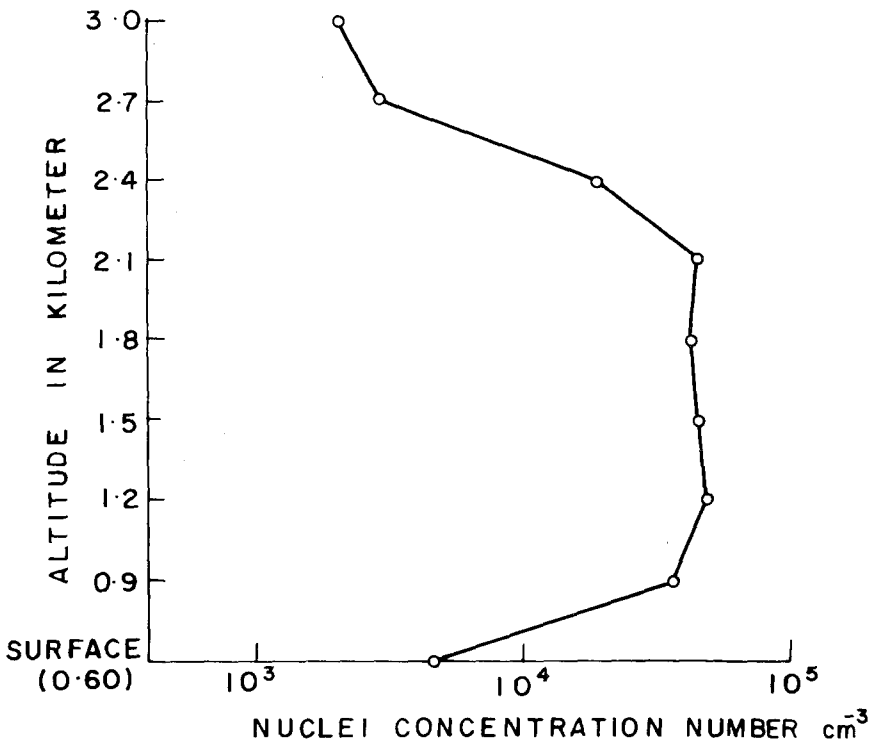

Fig. 3. Vertical distribution of A.N in Pune region based on observations made during three years of monsoon seasons from 1980–1982.

4.4. Diurnal variation

The diurnal variation of A.N based on the measurements made during March 3 to 13, 1981 at Pune is shown in Figure 4. The concentration was maximum when the solar elevation was maximum. A similar behaviour, which is not presented, was also noticed at Sinhagad.

Formation of particles from gases, or condensation of volatile vapours, has been reviewed by Bigg and Turvey (1978). Lopez *et al.* (1974) have suggested that vegetation must be an extremely important source of A.N. Sunlight and the presence of a catalyst such as NO_2 are believed to be essential for particle formation (Went, 1960, 1966). Ozone and organic vapors aided by sunlight, provide a good recipe for forming low vapor pressure products which can condense to form A.N (Vohra *et al.*, 1970). In order to study the association between A.N and trace gases, the average 6 h concentrations of SO_2, NH_3, NO_2, O_3, and A.N are given in Table II. The concentrations of trace gases SO_2, NH_3, and NO_2 were maximum during the night hours (2200 to 0400 hr) when the concentration of A.N was minimum. The gaseous concentrations were minimum during day light hours (1000 to 1600 hr) when the A.N concentration was maximum. These features bring out the influence of photochemical conversion of the trace gases, namely, SO_2, NO_2, and NH_3 on the concentration of A.N. The diurnal variation of A.N at Pune

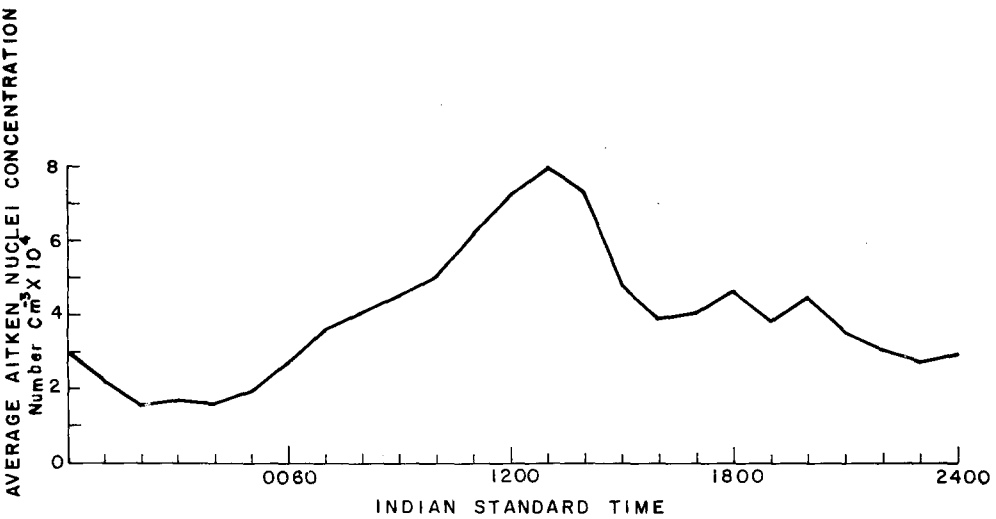

Fig. 4. Diurnal variation of A.N at Pune based on the observations made during 3–13 March, 1981.

is similar to what was noted at some continental stations (Hogan, 1968; Mohnen and Hogan, 1977) suggesting that solar radiation may be the important source for the formation of A.N in the presence of trace gases.

Both A.N and O_3 were found to be maximum during day-light hours (1000 to 1600 hr) and minimum during night hours (2200 to 0400 hr). This result is at variance with what has been reported from observations at Whiteface Mountain field station which pointed out anti-phase relationship between A.N and O_3 (Mohnen and Hogan, 1977).

The diurnal variation of A.N over the Bay of Bengal has been plotted in Figure 5. Considering the total data, the lowest concentration was observed in pre-dawn hours followed by rapid rise shortly after sunrise. Higher levels of A.N occurred throughout the day with a distinct peak at 0900 IST. The prolonged period of high day-time levels of A.N over the Bay of Bengal was apparently the result of contamination from the coasts or inland anthropogenic sources which were brought continuously by the westerly winds passing over land regions. The morning peak is attributed to the increased

TABLE II

Six hourly average concentration of trace gases in $\mu g\ m^{-3}$ and Aitken nuclei ($10^4\ cm^{-3}$) from 3–13 March, 1981.

Hours (IST)	SO_2	NH_3	NO_2	O_3	Aitken nuclei
1000–1600	2.1	1.3	1.0	22.3	5.8
1600–2200	3.6	1.8	1.8	13.9	3.9
2200–0400	10.7	2.6	2.5	6.8	2.0
0400–1000	5.8	2.3	1.1	12.0	3.3

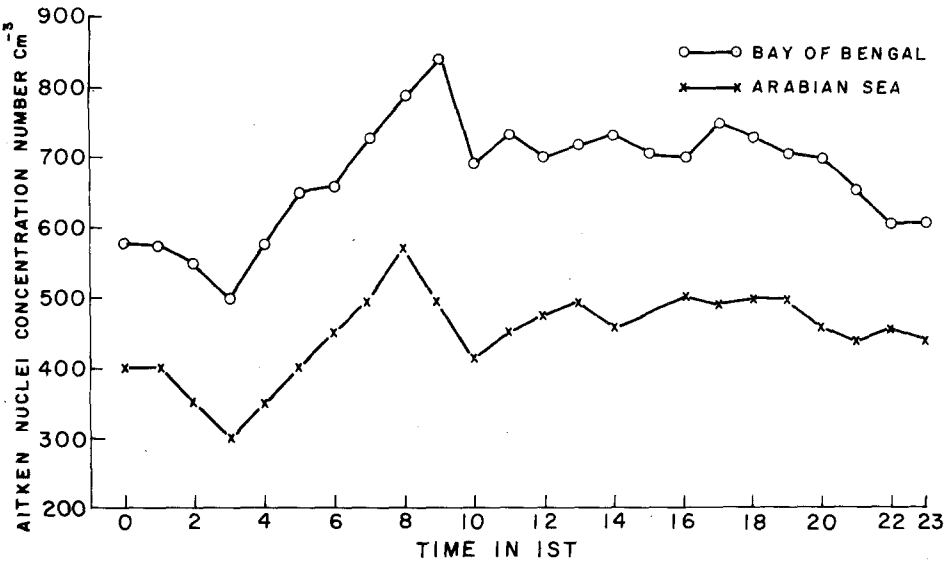

Fig. 5. Diurnal variation of A.N over the Bay of Bengal and Arabian sea regions based on the observations made during 10 days period in May 1983.

photochemical activity after sunrise. Shives and Robinson (1979) have reported similar diurnal variation for their observations at Pacific coast. The diurnal variation noticed over the Arabian sea, which resembles that over the Bay of Bengal, is also shown in Figure 5. The concentrations of A.N are markedly higher over the Bay of Bengal than over the Arabian sea. The reason for higher concentrations over the Bay of Bengal has been mentioned in Section 4.1.

4.5. Variation of A.N inside and outside cloud air

Observations of A.N were made inside stratocumulus and cumulus clouds and in the air outside the cloud at the same altitude during three monsoon seasons and the results are given in Table III. The concentration of A.N was found to be significantly higher

TABLE III

The average concentration of Aitken nuclei (10^4 cm^{-3}) inside cloud and cloud free air during three monsoon seasons at 1.5 km level. Figures in brackets denote standard deviations.

Years	No. of observations	Inside cloud	Cloud free air
1980	16	7.4	4.2
		(3.38)	(2.78)
1981	28	7.1	3.9
		(4.14)	(3.40)
1982	25	4.4	1.9
		(2.46)	(1.31)

inside the cloud than outside the cloud air. A similar feature was noticed in the measurements reported by others (Hegg *et al.*, 1980). The higher concentrations inside the cloud air were attributed to the higher gas-to-particle conversion rates inside the cloud.

5. Conclusions

Observations of A.N have been made in different environments. The concentration of A.N has varied from 1300 to 90 000 cm^{-3} for urban environment, 300 to 1500 cm^{-3} for non-urban environment, 1500 to 50 000 cm^{-3} for thermal power plant environment and 300 to 1050 cm^{-3} for marine environment.

The conversion of trace gases by solar radiation is a major influence in the diurnal variation of A.N. The formation of A.N by gas-to-particle conversion is low during night and high during day.

Low concentrations of A.N have been noticed in the forested region suggesting that, in the absence of pollution sources, vegetation alone may not be an important source for the formation of A.N.

Significant higher concentrations of A.N have been noticed in the clour air than in the cloud-free air. The implication of this feature on the microphysical properties of different cloud systems merits investigation.

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