

# EXPERIMENTAL STUDY ON EFFECTS OF HUMIDITY AND CHEMICAL COMPOSITION UPON IMAGINARY PART OF REFRACTIVE INDEX OF AEROSOL PARTICLES\*

LI Zihua(李子华)<sup>1\*\*</sup>, DONG Zhen(董真)<sup>2</sup>, YANG Jun(杨军)<sup>3</sup>  
and HUANG Shihong(黄世鸿)<sup>4</sup>

1) Shenyang Research Institute of Atmospheric Environment, Shenyang 110016

2) Nanjing Broadcasting and TV University, Nanjing 210048

3) Nanjing Institute of Meteorology, Nanjing 210044

4) Nanjing University, Nanjing 210093

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## ABSTRACT

Experimental study is made of the relationship between the imaginary part of refractive index (IRI) of atmospheric aerosol particles and relative humidity, and between IRI and chemical element through the assay of chemical constituents of the particles. Evidence suggests that atmospheric humidity and aerosols' chemical ingredients have great effects on the IRI's and they should thus be considered in the research of the radiation properties of the particles in the atmosphere.

**Key words:** aerosols, radiation properties, relative humidity, chemical composition

## I. INTRODUCTION

Investigation of aerosols in air has been receiving ever-increasing concern in atmospheric and environmental sciences, and their radiation properties are regarded as a crucial factor to climate and weather, in particular. Atmospheric aerosols affect directly weather and climate through absorption and scattering of solar and infrared radiation and indirectly cloud physiochemical processes and properties.

The imaginary part of refractive index (IRI) of aerosols denoted by  $n_i$  represents an important parameter of determining their absorptive performance but their radiation properties bear an intimate relation to their chemical constituents, atmospheric moisture and chemical conditions, indicating that the particles of dissimilar chemical ingredients have differing ability to absorb radiation. Measurement shows that the IRIs of aerosol particles as those from carbonaceous coal (Twitty and Weimam 1971), sea salt (Patterson et al. 1977) and sandstorm (Patterson 1980) are likely to differ by two orders of magnitude. But due to their inhomogeneous structure we have not yet reached the relationship of dissimilar elements to the IRI. Atmospheric environment humidity has direct impacts on their physiochemical properties. The rise of atmospheric humidity leads to the increase in their scale and complication of the complex refractive index on the

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\*\* The corresponding author is a professor of Nanjing Institute of Meteorology, now working as a guest researcher in the Shenyang Research Institute of Atmospheric Environment.

strength of the mixture of considerable water with solute from enlarged-size aerosols and possible liquid-phase chemical reactions. And in this respect Hänel (1976) and Sun (1983) made encouraging studies. To date, however, no reliable experimental evidence is available as concerns the variation in the IRIs with atmospheric humidity.

To investigate in depth the radiation properties of such aerosols we sampled in 23 observing sites all the country over and measured the IRIs at different relative humidity levels by use of Hogan (1984) technique to make approach to the relationship of the IRI to relative humidity, made use of sampled TSP (total suspended particles) and FP (floating particles) obtained to explore the IRIs in relation to the chemical content and size distribution of aerosols, and performed chemical assay of 111 samples to reveal the correlativity of the IRI with some of the elements contained.

## II. IRI MEASUREMENT OF ATMOSPHERIC AEROSOLS

To get the optical absorptive properties of atmospheric aerosols a simple particle sampling scheme developed by Hogan (1984) was used, with the results comparable to those of this kind presented by authors at home and abroad. Compared to Luan and Mao (1986), the Hogan technique makes use of opaque filters to measure reflective light from "colored" surfaces whereas the Luan-Mao scheme makes use of semi-transparent filters to measure the transmissivity of light through differently colored surfaces. In comparison, the former is superior because of simplicity.

Assume three opaque filters to be color-coded, viz., white (or blank), gray (a surface covered with particles, which is a specimen) and black, from all of which light is reflected from a fixed source, with the reflection intensity denoted, respectively, by  $I_{wr}$ ,  $I_{sr}$  and  $I_{br}$ , sensed by a photocell. In these intensities included is the reflection from the filter's accessories. In view of these filters having the identical structure and accessories, their reflection intensities differ only in "color-coded" effects of the surfaces. With constant source light intensity,  $I_{wr}$  and  $I_{br}$  are constant but  $I_{sr}$  (specimen) depends solely upon the amount of particles from a sampler exposed to air. Consequently,  $I_{sr} = I_{wr}$  for filters surfaced by no aerosols and  $I_{sr} = I_{br}$  when the specimen surface is totally covered with aerosol particles. Accordingly, following Hogan (1984) the portion of reflected light from the specimen filter is expressed by

$$R = \frac{I_{sr} - I_{br}}{I_{wr} - I_{br}} \quad (1)$$

And the loss of light from the surface is given by

$$E = 1 - R, \quad (2)$$

indicating that the loss, or extinction due to light absorption by the particles, denotes the bulk ability for aerosols to absorb visible light.

Set aerosol sampling rate to be  $V$  ( $\text{m}^3/\text{min}$ ), sampling time to be  $t$  and filter's area to be  $A$  and we define the linear absorption coefficient of visible light as

$$E_L = \frac{1 - R}{V \cdot t/A} = \frac{I_{wr} - I_{sr}}{(I_{wr} - I_{br})V \cdot t/A}, \quad (3)$$

where  $I_{wr}$  and  $I_{br}$  are constant,  $V$ ,  $t$  and  $A$  are assumed, leading to  $E_L$  only associated with  $I_{sr}$ , with its dimension of  $\text{M}^{-1}$ .

The measurement of the linear absorption coefficient of aerosols is more difficult on a technical basis and no equipment generally accepted is available. For this reason, the instrument developed by Hogan (1984) was utilized that contained all components of the

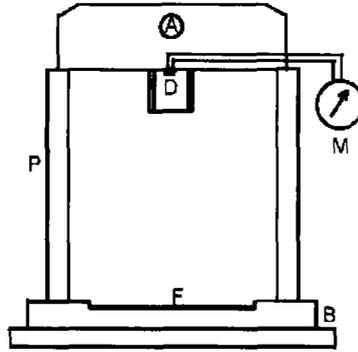


Fig. 1. Schematic diagram of the device for linearly absorbing coefficient  $E_L$  (for the meanings of the letters thereof refer to text).

prototype except for a single lamp used instead of two, with the configuration given in Fig. 1. A cylinder P with its 25-cm internal diameter slightly larger than that of the filters and its inside blackened is mounted on a rectangle-form receptacle B. A groove cut is made in the middle thereupon to allow a plate (F) to be moved freely therein. The sliding plate has three horizontal circular holes, each 0.1 cm in depth, for fixing filters. A photocell (D) is placed at the top, with the output connected to an outside microammeter (M). Just over the cylinder is a lamp (A) as a white diffuse source, whose spectrum is basically close to the solar equivalent. In measurement the filters for  $I_{wr}$ ,  $I_{sr}$  and  $I_{br}$  each were put into the holes, in order, to be directly under the illuminator (A) to get their readings from the microammeter (A). Values of  $V$ ,  $t$  and  $A$  set before exposure were used to calculate  $E_L$  by Eq. (3). The filters were the type of US Whatman 41.

Aerosols'  $E_L$  and their optical depth  $\tau$  satisfy

$$\tau = E_L \cdot \chi = \frac{I_{wr} - I_{sr}}{I_{wr} - I_{br}}, \tag{4}$$

in which  $\chi$  denotes the height of an air column,  $\chi = \frac{V \cdot t}{A}$  and by scattering theory we find

$$E_L = \int_0^\infty Q_{abs}(n_r, n_i, r, \lambda) \pi r^2 n(r) dr, \tag{5}$$

where  $Q_{abc}$  stands for the efficiency factor for wavelength  $\lambda$  light absorbed by aerosols with the real part  $n_r$  and imaginary part  $n_i$  of optical refractive index, and  $n(r)$  for the size distribution given. And for not too big a difference in size parameter  $\alpha$  between particles and always  $n_i \ll n_r$ , the absorbing efficiency factor has its approximate form

$$Q_{abs} = f(n_r) \cdot n_i \cdot \alpha \tag{6}$$

with  $\alpha = \frac{2\pi r}{\lambda}$  and  $f(n_r) = \frac{24n_r}{(n_r^2 + 2)^2}$ .

Combination of Eqs. (4)–(6) (Hu et al. 1991) leads to

$$n_i = \frac{2\tau \cdot A \cdot \lambda \cdot \rho}{3\pi \cdot f(n_r) \cdot M} \tag{7}$$

or

$$n_i = \frac{2E_L \cdot \lambda \cdot \rho \cdot V \cdot t}{3\pi \cdot f(n_r) \cdot M} = \frac{2(I_{wr} - I_{sr}) \cdot A \cdot \lambda \cdot \rho}{3\pi(I_{wr} - I_{br}) \cdot f(n_r) \cdot M}, \tag{8}$$

where  $A$  signifies the filter's area,  $\lambda$  the wavelength of light in use,  $\rho$  the density of particles and  $M$  the mass of sampled aerosols. And from Eq. (8) we get IRI, or  $n_i$  value.

Based on the precision of measured light flux, the relative error (RE) of  $\tau$  is  $< 10\%$ . At the accuracy level of  $10 \mu\text{g}$  given by an electron balance with its precision of  $10^{-5} \text{g}$  the RE of weighted particles is less than  $5\%$ . The RE of the filter's area  $A$  is about  $1\%$ . In calculation  $n_r$  is set to be 1.50 and if actual  $n_r$  values range between 1.45 and 1.55, then  $f(n_r)$  has its RE of  $< 4\%$ . The density of aerosols  $\rho$  is assumed to be  $1 \text{g/cm}^3$ , leading to its RE of  $20\%$ . The RE of galvanometer measurements is  $< 10\%$ . Following the principle of error propagation the RE of the imaginary part values  $n_i$  calculated from Eq. (8) is less than  $25\%$ .

### III. RELATIONSHIP BETWEEN IRI AND ENVIRONMENTAL RELATIVE HUMIDITY

Effects of environmental moisture on the optical properties of atmospheric aerosols lie in that 1) the particle grows hygroscopically, resulting in the increase of size and thus shifting the distribution towards larger-scale end and 2) aerosol is combined with water, thereby modifying their refractive index, in which may be included the change in IRI from liquid-phase chemical reactions that result in the alteration of chemical constituents.

In experiments with the impacts of environmental humidity upon the IRI it is of paramount importance to accurately control relative humidity ( $RH$ ). Utilized in our work was a closed glass container placed in a constant temperature chamber with silica-gel desiccant and hygroscopic paper inside and the regulation of the amounts of the desiccant and paper would lead to steady  $RH$  readings. We left the sample of aerosols in the chamber with constant  $RH$  for at least 2 h so as to make the specimen and environment at identical  $RH$ . Next to the closed chamber was the measuring device for immediately putting the sample into the latter after it was taken from the former and completing the measurement of  $n_i$  in half a minute. The  $RH$  was set to 10 values of  $10\%$ ,  $24\%$ ,  $32\%$ ,  $40\%$ ,  $48\%$ ,  $60\%$ ,  $65\%$ ,  $75\%$ ,  $80\%$  and  $97\%$ .

The experimental samples of aerosols had their size ranges 1) over  $0-100 \mu\text{m}$  for TSP taken by the samplers of KB series and 2) over  $0-2 \mu\text{m}$  for FP as the fine mode collected by the Anderson sampler. All the specimens were gathered at Nanjing Institute of Meteorology around 1997, of which the TSP ones consisted of TSP # 2, # 10 and # 12 and the three FP samples were FP # 23, # 27 and # h taken at Xishuangbanna, Yunnan Province of SW China, with their variations in  $n_i$  versus  $RH$  shown in Fig. 2.

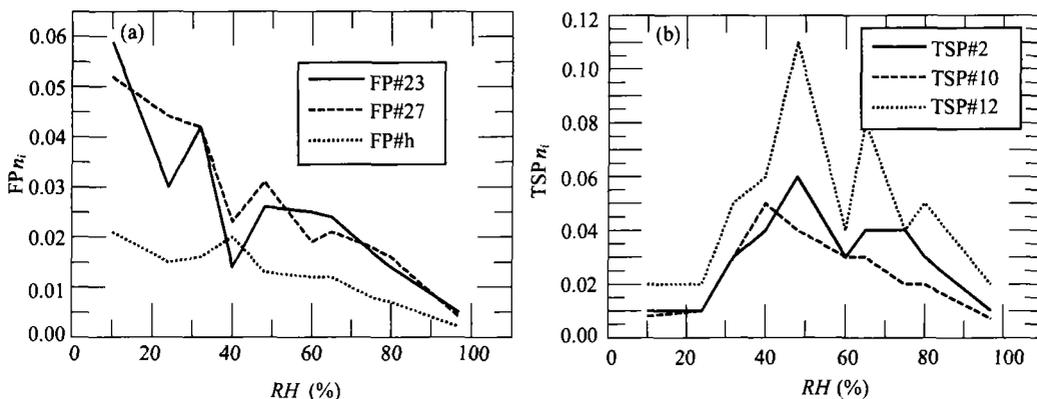


Fig. 2.  $RH$  dependent variations in IRIs of FP (a) and TSP (b) under the illumination of visible light.

It is seen from Fig. 2a that the  $n_i$  values of FP sizes reduce with increased  $RH$  and diminish from 0.05–0.06 at 10% to 0.005 at 97%  $RH$ , differing by one order of magnitude. From Fig. 2b we see that the  $n_i$  values of TSP aerosols increase with  $RH$  in the range of 5%–50% but decrease versus increasing  $RH$  of 50%–100%, a result that is consistent with that of Takamura et al. (1984) and Horvath and Trier (1993). The  $n_i$  values of FP and TSP aerosols changing with  $RH$  in different ways are due mainly to the sizes and chemical constituents. Analysis of element enrichment shows that very small-sized FP particles contain more sulphur, falling into sulfate substances of high moisture absorption. These particles grow substantially when  $RH$  increases and because the relative weight of water in aerosols rises quickly, the refractive indexes approach those of water. We take for example the FP #h specimen collected from Xishuangbanna, whose  $n_i$  value is rather small and changes more smoothly as  $RH$  rises. This is because the chemical elements of the particles are of crust's origin, with small amount of artificial release of sulphur, leading to weaker absorption of water. For larger TSP, in contrast, they are composed largely of soil and sand particles having lower hygroscopic growth, and while their sizes grow with moisture, the water content of an aerosol shows more noticeable increase only when  $RH$  exceeds a certain value (say, 50%), leading to their  $n_i$  values approaching that of water, but for  $RH < 50\%$  the change of  $n_i$  is as yet unexplainable. The possible interpretation may be the difference in chemical composition between TSP and FP particles and with increasing  $RH$  a large aerosol absorbs less water, in which probably occur complicated liquid-phase chemical reactions that lead to change in chemical constituents, making  $n_i$  augmented, but with  $RH$  in excess of 50%, a large aerosol takes in more water, making for the rise of water content and thus its  $n_i$  value close to that of water, a problem that needs to be further investigated.

From the foregoing results, we see that the  $n_i$  value of a aerosol is different either from that of a dry particle or that of water but determined jointly by a dry particle with its absorbed water; the  $n_i$  value depends upon chemical ingredients for its magnitude. Consequently, for a hygroscopic aerosol its  $n_i$  is required to be corrected.

#### IV. RELATION OF $n_i$ TO AEROSOL CONCENTRATION AND SIZE DISTRIBUTION

To understand in depth the properties of atmospheric aerosols across the country, Chinese researchers began in 1987 to gather the samples from 23 measuring sites including Zetang of Tibet Region and Xishuangbanna of Yunnan Province. All the specimens were collected solely in spring and autumn except those from Qinzhou of Guangxi in summer, and Harbin of Heilongjiang, Dalian of Liaoning, Xishuangbanna of Yunnan and Zetang of Tibet in winter. The total areal fluxes of sampled air were limited to the range 0.6–1.0  $\text{m}^3/\text{cm}^2$ . If the flux was too small, so was the number of aerosols attached to the filter, leading to unidentifiable  $I_{wr}$  and  $I_{sr}$  of Eq. (3). In the opposite case too big a flux would result in measurement error because of superimposed aerosols on the filter. Moreover, one specimen was sampled for 5–6 h a day and at least five samples were taken from one of the sites. In view of the fact that all the sites were mostly in urban areas under the influence of sources discharging pollutants, the content of aerosols was bound to be higher compared to that in rural segments, leading to higher  $E_L$  of urban specimens. Nevertheless, the obtained urban  $E_L$  is of significance as the upper limit of a regional magnitude.

Figure 3 depicts atmospheric aerosols'  $n_i$  distribution over China. On the whole,  $n_i$

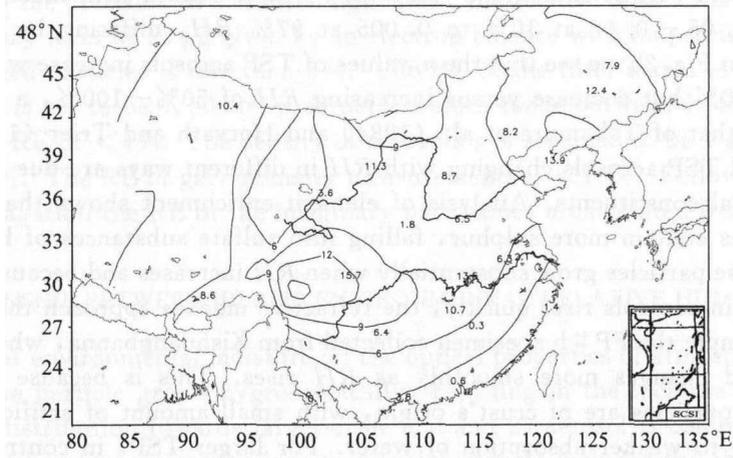


Fig. 3. Distribution of  $n_i$  values ( $\times 10^{-2}$ ) over China.

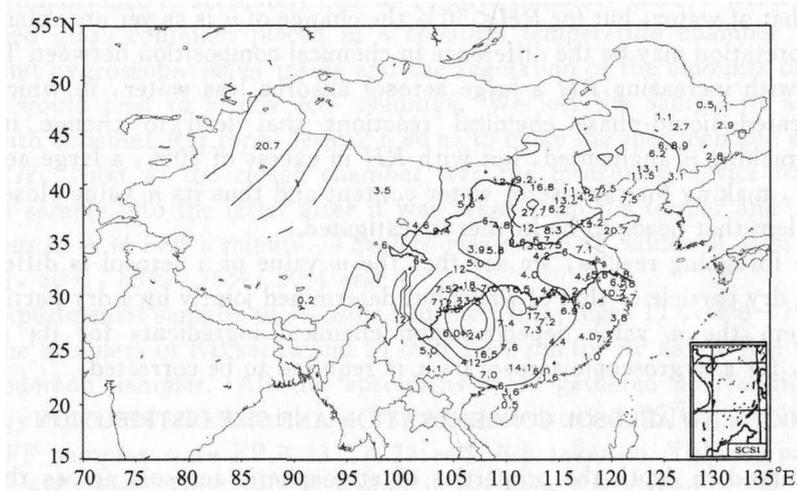


Fig. 4. Distribution of daily mean TSP isopleths ( $0.1 \text{ mg/m}^3$ ) for 1990–1995 across China.

values range over  $0.001 - 0.157$ , differing by two orders of magnitude, with values mostly of  $> 0.050$  ( $< 0.050$ ) distributed in the north (south). A relatively high core is in the Sichuan Basin and Guizhou, with the maximum  $n_i$  of  $0.157$  at Chengdu in contrast to  $0.001$  at Xishuangbanna. Figure 4 is the plot of daily averaged atmospheric TSP ( $0.1 \text{ mg/cm}^3$ ) in 1990–1995 published by the National Environment Monitoring Center of China. By and large, the distribution is marked by high (low) values in the north (south) but the high and low value cores of  $n_i$  and TSP do not coincide with each other, revealing no apparent correlativity between them. Yet, the correlation is close between  $E_L$  and TSP concentration.

Figure 5 delineates the absorption coefficient  $E_L$  in relation to the content of aerosols of 129 samples taken from Nanjing and Xi'an, indicating that  $E_L$  increases with the concentration and is in highly positive correlation with the content, reaching a coefficient of  $0.74$  at  $0.001$  significance level.

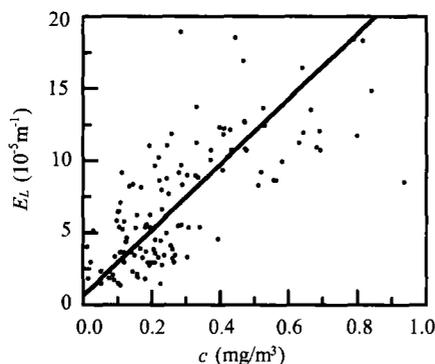


Fig. 5. Correlations between the absorption coefficient  $E_L$  and aerosol concentration  $c$ .

Because of the different source and chemical composition for different aerosol mode, there is a certain correlation between  $n_i$  and concentration changing with different sizes of particles. Therefore, we sampled simultaneously aerosols, 0–100  $\mu\text{m}$  (TSP) and 0–2  $\mu\text{m}$  in diameter (FP), by using samplers of KB series and Anderson type at some sites to measure  $n_{i,\text{TSP}}$  and  $n_{i,\text{FP}}$ . To compare the  $n_i$  values in the two ranges, we define

$$a = \frac{n_{i,\text{FP}}}{n_{i,\text{TSP}}},$$

where  $a$  denotes the relative contribution to  $n_i$  of FP aerosols. Table 1 presents the calculations of  $a$  based on data sampled at a few sites, showing that all calculations exceed 5, which reveals that the contribution of small-size aerosols to the visible light absorption is greater than that of larger ones because of their difference in chemical composition. Following James (1978), particles with  $\geq 1 \mu\text{m}$  in diameter are dominantly of soil origin and submicro-size aerosols are composed chiefly of substances with  $\text{NH}_4^+$  as the component and carbon particles. Obviously, the latter differ greatly from soil particles in absorbing visible light.

**Table 1.** Ratios of  $n_{i,\text{FP}}$  to  $n_{i,\text{TSP}}$  for Some Measuring Sites

Site	Nanjing Inst. Meteorology	Nanjing University	Xishuang- banna	Zetang, Tibet
samples	9	4	2	3
$a$ value	12.49	9.50	10.21	8.71

## V. RELATION OF $n_i$ TO CHEMICAL COMPOSITION OF ATMOSPHERIC AEROSOLS

The optical absorption properties of the aerosols depend not only upon their size distribution and environmental humidity but their chemical composition as well. It is generally recognized that the optical absorption of aerosols relies on the content of element carbon, whereupon the absorption coefficient is computed. Studies show  $n_i$  to be in positive correlation with the content of carbon, with  $r=0.658$  (Guo et al. 1996). Beyond the content of carbon little is known about studies on the correlativity with any of the

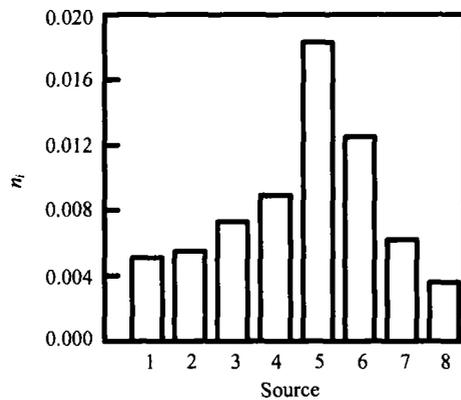


Fig. 6. Values of  $n_i$  of aerosols from the eight sources mentioned in text.

other elements.

In investigating the dependence of optical absorption properties upon chemical composition we experimentally made research of the  $n_i$  values of different kinds of atmospheric aerosols released from 8 single sources. The specimens Nos. 1, 2 and 8, were collected from the suburb of Xi'an, Yinchuan and Xishuangbanna, respectively, No. 3 for soot of the thermal power plant, No. 4 from building sites, No. 5 for metallurgical powder, No. 6 from secondary raised dust (the 4 kinds were all from Xi'an) and No. 7 from the accumulated cover of sand and dust from Nedong County of Tibet. These source specimens were sieved for the sizes 0–100  $\mu\text{m}$  and put on the filter for experiment, leading to their  $n_i$  values at 40%–50% RH. The  $n_i$  values are shown in Fig. 6 where we see the maximum given by No. 5 sample and minima by Nos. 8, 1 and 2, with the others in between, revealing the dependence of  $n_i$  values of aerosols from different origins upon their chemical composition and the crucial effects of artificial, especially industrial, discharge, on the  $n_i$  values.

To further investigate the correlation between  $n_i$  and a related element, we have dealt with  $n_i$  values of multiple specimens, and some of the elements were assayed. Specimens were collected in four months representative of the corresponding season, i. e., August of 1997, January, April and October of 1998 for Xi'an and October of 1998, January, April and July of 1999 for Nanjing. In both regions each had three sampling sites, viz., a piece of grassy land, 37 km from the city, a business quarter and an industrial area in the case of Xi'an, and a business zone, a heavy traffic area and a famous scenic resort in Nanjing. Five samples were gathered from each of the sites, totaling in 120 specimens. Removing nine contaminated samples we had 111 specimens with which to construct a sequence of representative data for statistical analysis. Calculation was accomplished of  $n_i$  of each sample and assay of its contents of chemical elements by X-ray fluorescence spectrometer (XRF), with the elements including Si, Al, Ca, Fe, K, Mg, Mn, Na, Ti, S, P, Zn and Pb. The  $n_i$  values ranged over 0.005–0.14, covering two orders of magnitude, a domain that is the same as that given in the compilation of Gerber and Hindman (1981). Below given are the correlations between  $n_i$  values and each of the related elements.

**Table 2.** Correlation ( $r$ ) between  $n_i$  and Chemical Elements of Atmospheric Aerosols

Element	Si	Al	Ca	Fe	K	Mg	Mn	Na	Ti	S	P	Pb	Zn
$r$	0.2	0.1	-0.1	0.1	0.1	0.1	0.1	0.1	0.25	0.36	0.21	0.3	-0.02

It is seen therefrom that  $n_i$  values are positively correlated with contents of eleven of the elements except negatively of Ca and Zn, but with small correlation coefficients of  $-0.1$  and  $-0.02$ , respectively. On the other hand,  $n_i$  values, although positively related to the contents of Al, Fe, K, Mg, Mn and Na, have small-value correlations of  $r < 0.15$ . As a result, it is impossible to identify whether the effect is available of the elements or their compounds upon  $n_i$  values. The correlations of Si, Ti and P to  $n_i$ , while giving bigger coefficients ( $0.21-0.25$ ), show that their values to be less than  $0.254$  as the reference value for the tests at  $0.01$  significance level, leading to no apparent correlativity with  $n_i$  values. Only S ( $r=0.36$ ) and Pb ( $r=0.30$ ) are valid in this case ( $r > 0.254$ ), indicating remarkable positive correlation of the contents of S and Pb to  $n_i$  values, which suggests that the  $n_i$  values are definitely in positive correlation with the content of sulphur, lead and their compounds, so that they make noticeable contributions to the optical absorption of atmospheric aerosols.

Besides, separate sequences of 54 and 57 samples from Xi'an and Nanjing, respectively, were constructed for further investigating the correlation between values of  $n_i$  and sulphur, revealing the correlation coefficients  $r=0.534$  and  $0.281$ , in order, the former being nearly twice as high. This indicates that the Xi'an positive correlation is more apparent compared to Nanjing equivalent, as discerned in Fig. 7. Dissimilarity arises between the cities owing to the difference in the content of sulphur of aerosol. The study reveals that the ratio of the content of sulphur to the total of 13 elements of aerosol is higher in Xi'an than in Nanjing, arriving at  $1.29:1$ , which is responsible for the higher correlation of Xi'an S to  $n_i$ . This demonstrates that the  $n_i$  value bears a relation to the origin of atmospheric aerosols, indicating that particles from different sources differ in chemical composition.

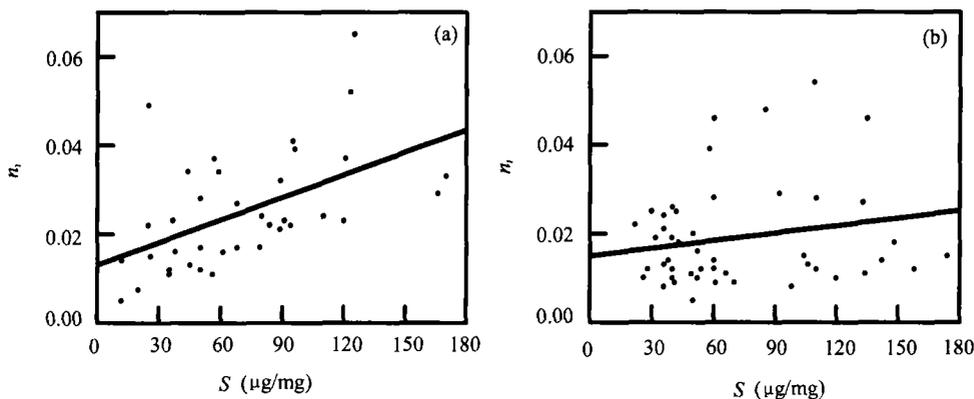


Fig. 7. Correlation between  $n_i$  value and the content of sulphur ( $\mu\text{g}/\text{mg}$ ) in aerosol for Xi'an (a) and Nanjing (b).

## VI. CONCLUSIONS

Based on the foregoing analysis we come to the following.

(1) Atmospheric moisture affects the  $n_i$  value of aerosol, which, for fine particles, drops with increased  $RH$  (relative humidity) but does the same for larger particles only with  $RH > 50\%$ .

(2) The  $n_i$  value has no distinct correlation with the concentration of aerosols except for its variation with their sizes. A fine particle makes significantly greater contribution to the visible light absorption compared to a larger one.

(3) Atmospheric aerosol's chemical composition contributes to its  $n_i$  value. In addition to the positive correlation of  $n_i$  with the content of carbon, the  $n_i$  value has a remarkable correlativity with contents of sulphur and lead.

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