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A principal component analysis of visibility and air pollution in six California cities

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RESUMEN

Los datos de seis estaciones de calidad del aire en California fueron utilizados para hacer un análisis de los principales componentes de variables elementales, gases y variables meteorológicas. La naturaleza altamente intercorrelacionada de muchas de las variables puede producir resultados dudosos, cuando se usan técnicas de regresión ordinarias. La aplicación del análisis de componente principal al conjunto de datos produce combinaciones lineales, estadísticamente independientes, de las variables orignales. El factor que representa la materia en forma de partículas derivadas del suelo generalmente explica la mayor parte de la variabilidad observada. El factor visibilidad está presente en todas las localidades y aerosoles de tamaño intermedio conteniendo azufre (rango del tamaño del azufre 0.6 μm $< D_p <$ 3.6μm) son el mayor contribuyente de la reducción de visibilidad en todos los sitios: costa, zona urbana y valle interior. El resto de los principales factores fueron, con base en su composición química, provisionalmente atribuidos a contaminación de automóviles, quema de combustible y quema agrícola.

ABSTRACT

The data from six air quality stations in California were subjected to a principal components analysis involving elemental, gas, and meteorological variables. The highly intercorrelated nature of many of the variables can make the results of ordinary regresssion techniques uncertain. The application of principal component analysis to the data set produces statistically independent linear combinations of the original variables. The factor which represents soil-derived particulate matter usually accounts for most of the observed variability. The visibility factor is present at every location and intermediate size sulfur-containing aerosols (sulfur size mode 0.6 μm $< D_p <$ 3.6μm) are major contributors to visibility reduction at all sites: coastal, urban, and interior valley. The rest of the principal factors were tentatively assigned to automotive pollution, fuel burning, and agricultural burning on the basis of their chemical composition.

Introduction

The investigation of the basic relationships between air quality and other meteorological variables by statistical means is complicated by the highly intercorrelated nature of the variations in the data. Many variables of interest tend to rise and fall more or less in tandem which presents problems for statistical analysis and interpretation. Principal component analysis can provide valuable insight into the underlying chemical and physical processes of the atmosphere. The

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*Present address: California Air Resources Board, Technical Support Division, P. O. Box 2815, Sacramento, CA. 95812 primary purpose of this work has been to apply a method to screen and analyze large air quality data sets and to identify the major pollutant sources which contribute to visibility degradation.

Trijonis (1982) showed that two significant areas of heavy man-made visibility impact in California are the Los Angeles basin and the San Joaquin Valley. The extremely low visibilities in the central and eastern parts of the Los Angeles basin are likely caused by the high concentration of SO_x, NO_x, hydrocarbon, and particulate emissions in the air basin. The problem is exacerbated by relatively low wind speeds, strong inversion layers, and intense sunlight. The low visibility in the San Joaquin Valley may be related to the high level of SO_x emissions in that area, the relatively long residence times of air parcels in the San Joaquin Valley, the transport of secondary aerosol precursors from the San Francisco Bay area, and particulate matter from agricultural burning and dust sources. In contrast, visibility degradation in far-northern California (an area of lesser impact) is mainly due to natural factors such as fog and relative humidity.

Principal component analysis is a powerful tool for relating a large set of elemental concentrations to their possible sources without prior assumptions as to the number or nature of the sources. Relations between visibility and air pollution by particles have been studied with factor analysis and principal components analysis (Linak and Peterson, 1981; Pitchford *et al.*, 1981; Flocchini *et al.*, 1981). Ashbaugh *et al.* (1984) applied principal component analysis to a data set of 40 air samplers in the western United States to obtain spatial patterns of sulfur concentrations. After rotation of the initial eigenvectors, two large regions were identified which accounted for 33.1% of the variance in the data. Three other smaller regions were identified which also had significant variance. A statistical approach, Specific Rotation Factor Analysis (SRFA), has been developed by Koutrakis and Spengler (1987) to identify and apportion sources of ambient air pollutants. This method diverges from other multivariate techniques in that eigenvectors are obtained by analyzing the covariance matrix. Empirical Orthogonal Functions (EOFs) have been applied (Henry and Gebhart, 1988) to airborne sulfur in the western united states.

Finally, it should be mentioned that few applications of principal component analysis to air quality data have been reported as yet. There appear to be few, if any published accounts of a principal component analysis for a size segregated particulate data set similar to that analyzed in this work, which covers a variety of topographic and meteorological conditions throughout diverse regions in California. Data and site descriptions are given in Motallebi *et al.* (1990).

Statistical procedures

Principal component analysis is a statistical procedure which explores the data-reduction possibilities by constructing a set of new variables on the basis of the interrelations exhibited in the data set. Its objective is to transform a given set of m variables into a new set of composite variables or principal components that are orthogonal to each other.

The principal component model can be expressed as:

$$Z_i = \sum A_{ip}.F_p \qquad\qquad (p = 1,\ 2, ..., m)$$

where each one of the observed Z variables is described in terms of P new components, F_1, F_2, ..., each one of which is in turn defined as a linear combination of the original variables . The A_{ip} terms are called factor loadings and they represent the weight of each variable in each one of the components.

The key to the calculation of the principal components is the correlation matrix R (m,m) which is diagonalized. The first component associated with the largest eigenvalue is the best linear combination which explains, as far as possible, the overall variance of the system. The second component is the second linear combination, orthogonal to the first , which explains, as best as possible, the residual variance of the system, and so on. Generally the importance of a component may be evaluated by examining the proportion of the overall variance it explains (Harman, 1976).

Climatology of the regions

The following section is a brief description of climatology of the regions. Figure 1 shows a map of the locations. The lower Sacramento valley, where Sacramento is located, enjoys a mild climate and abundance of sunshine most of the year. Prevailing winds at Sacramento are southerly every month except November, when they are northerly. This is due to the north-south direction of the valley and the deflecting effect of the towering Sierra Ranges on the prevailing oceanic winds which move through the Carquinez Straits at the junction of the Sacramento and San Joaquin Rivers. The meteorology and climate of the Sacramento valley are favorable for the development of air pollution. Light winds and atmospheric stability provide frequent opportunities for pollutants to accumulate in the atmosphere. The general circulation, characterized by summertime up-valley

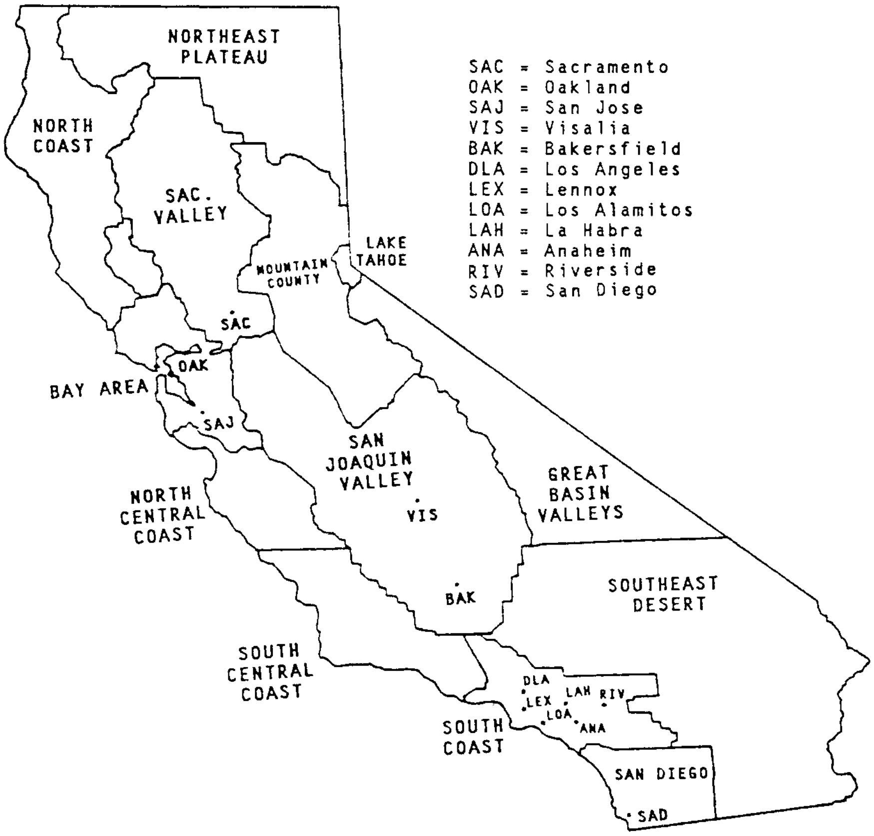

Fig. 1. Map of the California indicating the sampling sites for particulate a gaseous pollutants.

and wintertime down-valley winds, permits the transport of pollution over long distances along the axis of the valley. Photochemical smog in the summer and early fall is enhanced by the almost unbroken succession of warm, sunny days during these seasons. Carbon monoxide, oxides of nitrogen, total suspended particles, and suspended particulate lead concentrations in the late fall and winter are highest when there is little interchange of air between the valley and the coast. This type of the weather is associated with the tule fog regime when temperature inversions at ground level persist over the entire valley for several weeks and air movement is virtually absent.

Bakersfield, situated in the extreme southern end of the San Joaquin Valley, is partially surrounded by a horseshoe-shaped rim of mountains with an open side to the northwest and the crest at an average distance of 40 miles. Generally, pollutants transported into the San Joaquin Valley from the California Delta, San Francisco Bay and Livermore regions were found to primarily impact the western side of the northern half of the San Joaquin Valley (Reible *et al.*, 1981). The principal exit region for pollutants from the valley is the Tehachapi mountain ridge which forms the southeast border of the valley. The upslope flow along the north-south edges of the valley is relatively inefficient in removing pollutants because of its diurnal nature and the broad width of the valley. Consistent with the influx of air at the mouth of the valley, the transport within the valley was generally southward. The nocturnal jet enhances this southward movement and provides a mechanism for distributing pollutants throughout the valley.

San Jose is located at the center of the Santa Clara Valley. The valley, which slopes gently toward San Francisco Bay on the north, is approximately 60 miles long and 15 to 20 miles wide. On the east, the Mt. Hamilton Range rises to an elevation of 4,000 feet, protecting the valley from the hot winds of the interior, on the west, the Santa Cruz Range, with an elevation of 2,500 feet, shelters the valley from ocean winds and fog. Prevailing winds are from the northwest in a combination land-and-sea breeze. Wind speeds are generally light with the average velocity ranging from 5.4 mph in November to 7.4 mph in June. The same climate that made the valley a center of agricultural production has contributed to the development of an industrial and research complex that includes many of the nation's corporate giants.

Predominating influences on the climate of the Los Angeles site are the Pacific Ocean to the west, the southern California coastal mountain ranges which line the inland side of the coastal plain, and the large scale weather patterns which allow pacific storm paths to extend as far south as the Los Angeles area only during late fall, winter, and early spring. Visibility at the Los Angeles site is frequently restricted by haze, fog, or smoke. Low visibilities are favored by a layer of moist marine air with warm dry air above. Lowest visibilities usually occur with weak winds, but at times a moderate afternoon sea breeze will bring a fog bank ashore and over the airport. Light fog occurs at some times nearly every month, but heavy fog is observed least during the summer and can be expected on about one night or early morning in four during the winter.

The climate of Los Alamitos site is considerably influenced by local topography. In fact, the topography plays a greater role in the climate conditions at this station than the more general movements of pressure systems which dominate other sections of the country. The Pacific ocean, few miles south and west, has a moderating effect on temperatures.

Two general types of climate are experienced in Riverside County. In the western portion of the county, the marine influence of the Pacific Ocean is felt while in the eastern part the marine influence is essentially cut off by the San Jacinto and Little San Bernardino Mountains. Airflow over the county is from the northwest quadrant much of the time. Winds from the southeast quadrant are also common over the central portion of the county. Riverside County's most serious air pollution is oxidant followed by particulate matter. While there are sources of oxidant emitted within the county, the major source is the metropolitan area of Los Angeles County.

The results of the principal component analysis

The data from six air quality stations in California were subjected to a principal component analysis involving elemental, gas, and meteorological variables. The initial components were rotated using the orthogonal varimax method to obtain the final results. It should be mentioned that we performed a direct quartimin rotation of the same elements. The quartimin pattern is quite similar to that obtained from the orthogonal case with the exception of an interchange of factors. This appears to be due to the ordering of the factors according to the variance explained, which is different in each case. The intercomponent correlations, which are obtained from a direct quartimin rotation, are well below 0.3. As mentioned by Ashbaugh *et al.* (1984), "since there is little intercorrelation between principal components, there is little to gain by using an oblique rotation". Meanwhile, by constraining the rotation to orthogonality, one can easily determine the variance explained by each component, whereas it requires additional calculation with the oblique rotation.

Figures 2 through 11 illustrate the loading on the first six components for the different pollution monitoring stations. Dotted lines in these graphs show a factor loading of 0.5. The variables are listed according to decreasing loadings on the principal components and only the highest loadings (> 0.300) are listed. Variables were labelled with numbers 2 and 3, indicate intermediate size

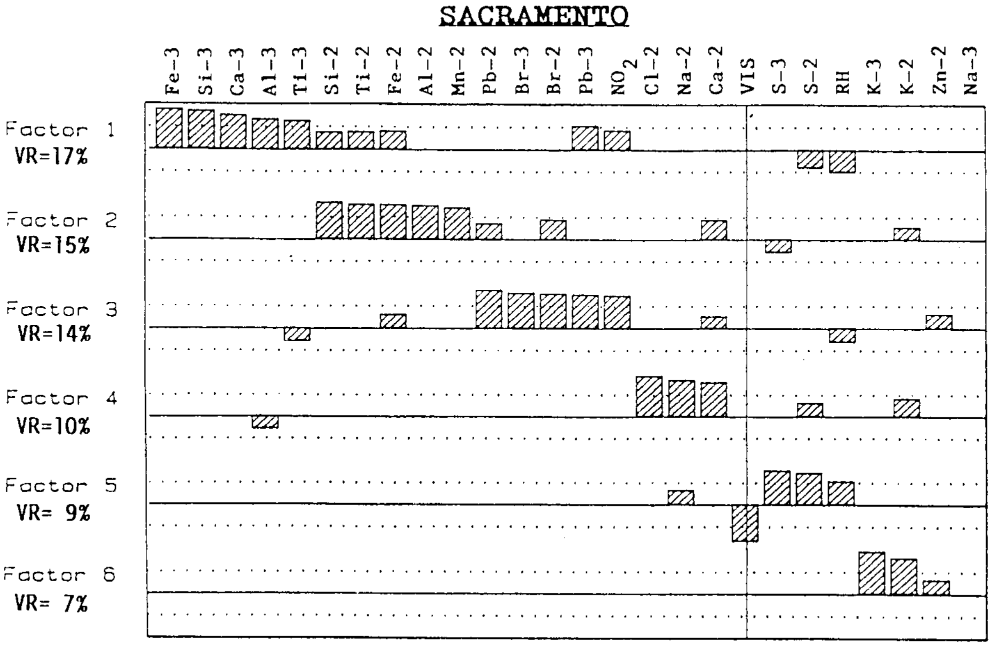

Fig. 2. Sorted rotated factor loadings, summer 1977, 2. indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

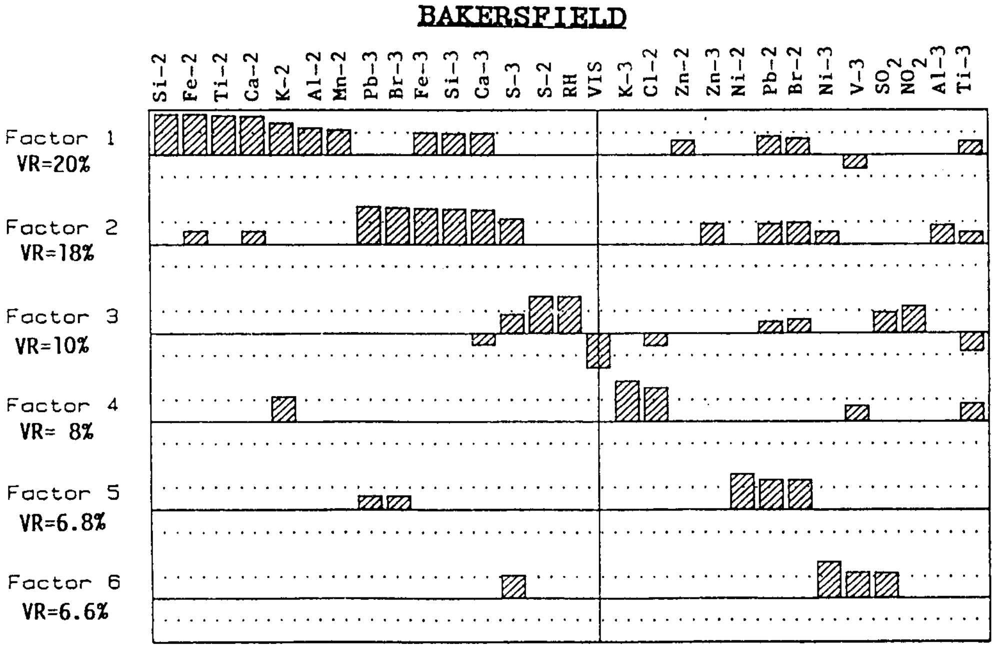

Fig. 3. Sorted rotated factor loadings, summer 1977, 2. indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

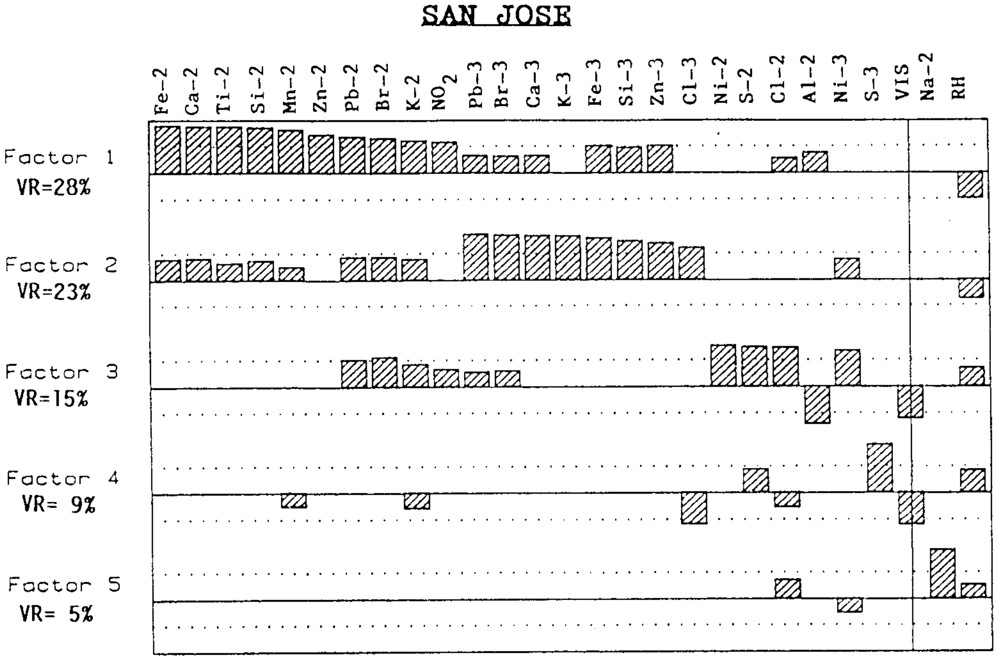

Fig. 4. Sorted rotated factor loadings, winter 1977, 2. indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

particles (.65 μm to 3.6 μm) and fine particles (0.1 μm to 0.65 μm), respectively. The abbreviation VR in Figures 2 through 11 indicates the percent of variance explained by the component. The higher the loading of a variable the more that variable contributes to the variation accounted for by the principal component. The inclusion of the five or six factors of the following figures is based on the scheme developed by Overland and Preisendorfer (1982). Ashbaugh *et al.* (1984) used

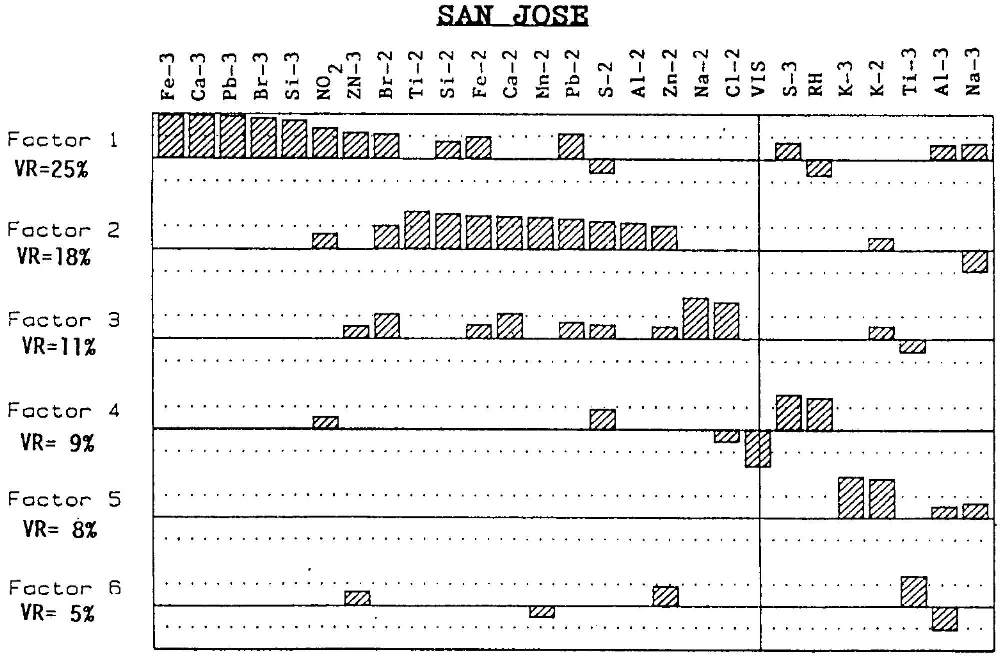

Fig. 5. Sorted rotated factor loadings, summer 1977, 2. indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

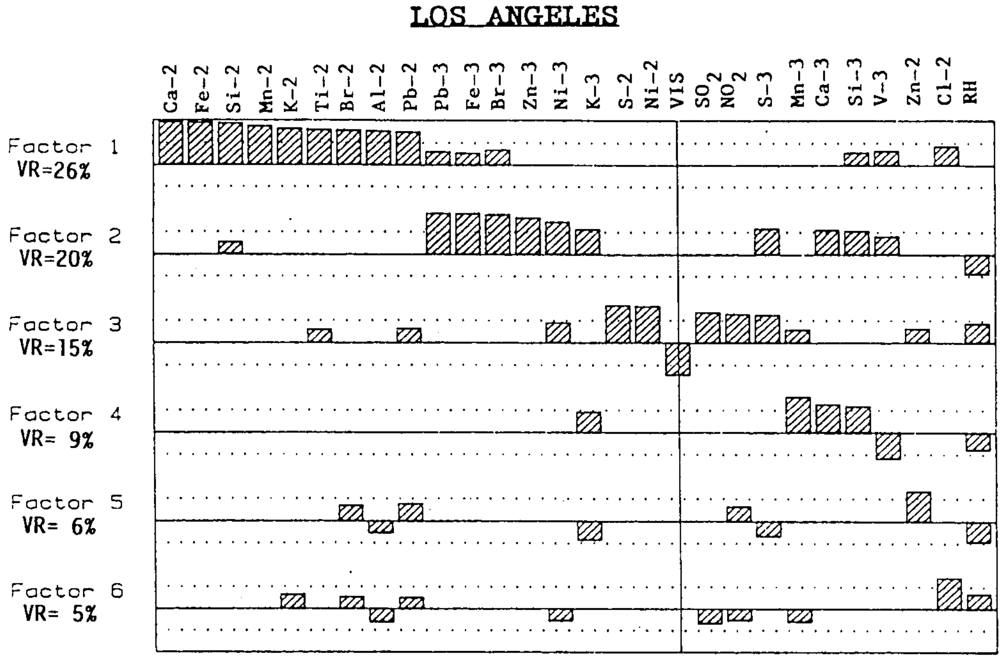

Fig. 6. Sorted rotated factor loadings, winter 1977, 2. indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

this test for an objective cutoff for the number of principal components to keep in an analysis. Meanwhile, several combinations of variables were used in the principal component analytical procedure. Only the final solution is presented here, as there were no qualitative differences in factor resolution among the different trials.

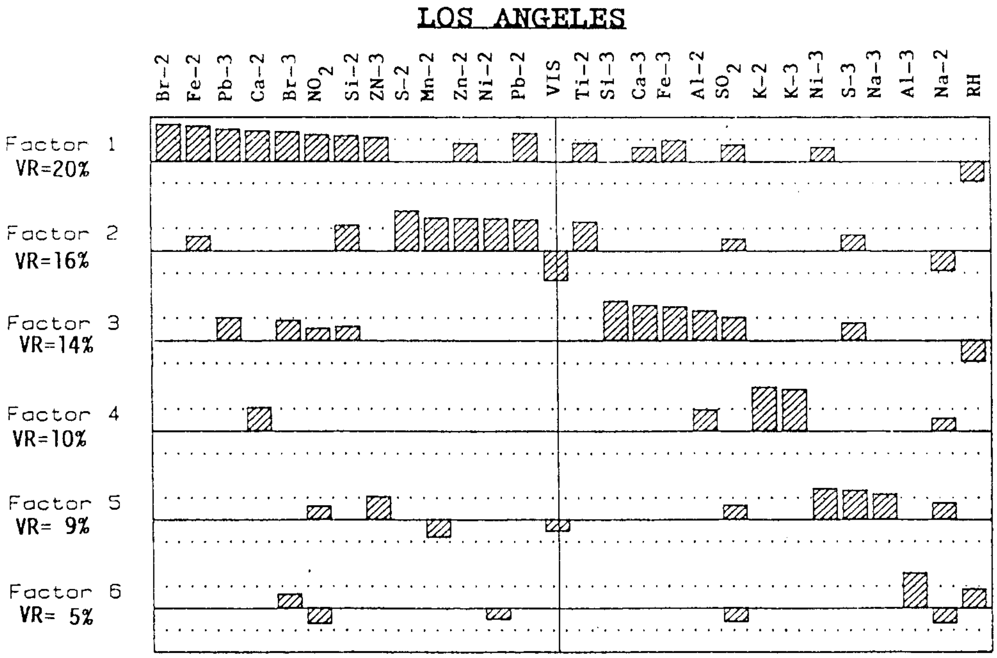

Fig. 7. Sorted rotated factor loadings, summer 1977, 2. indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

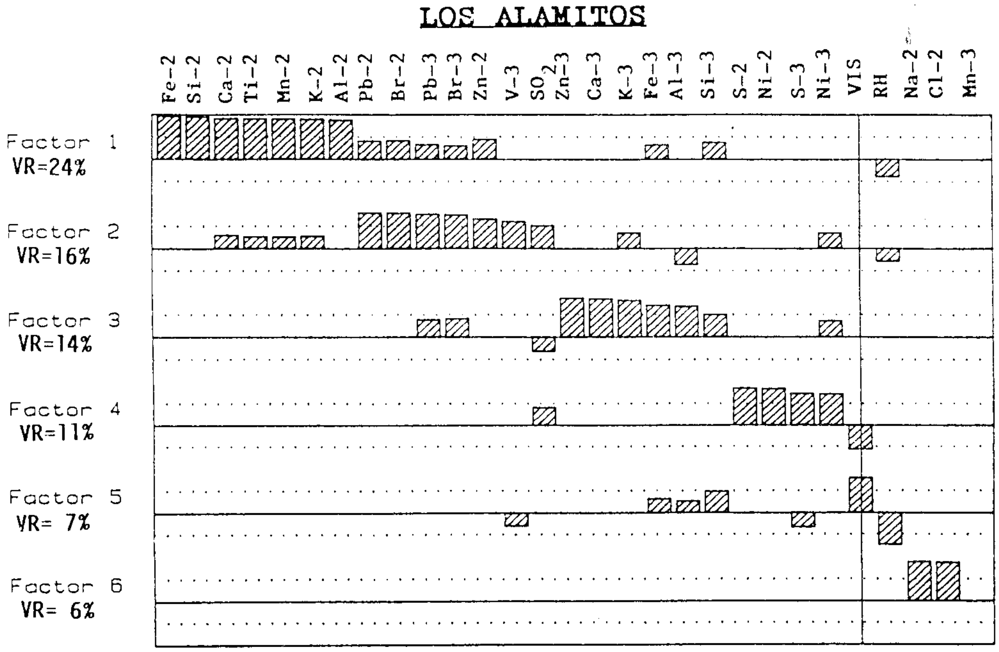

Fig. 8. Sorted rotated factor loadings, winter 1977, 2. indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

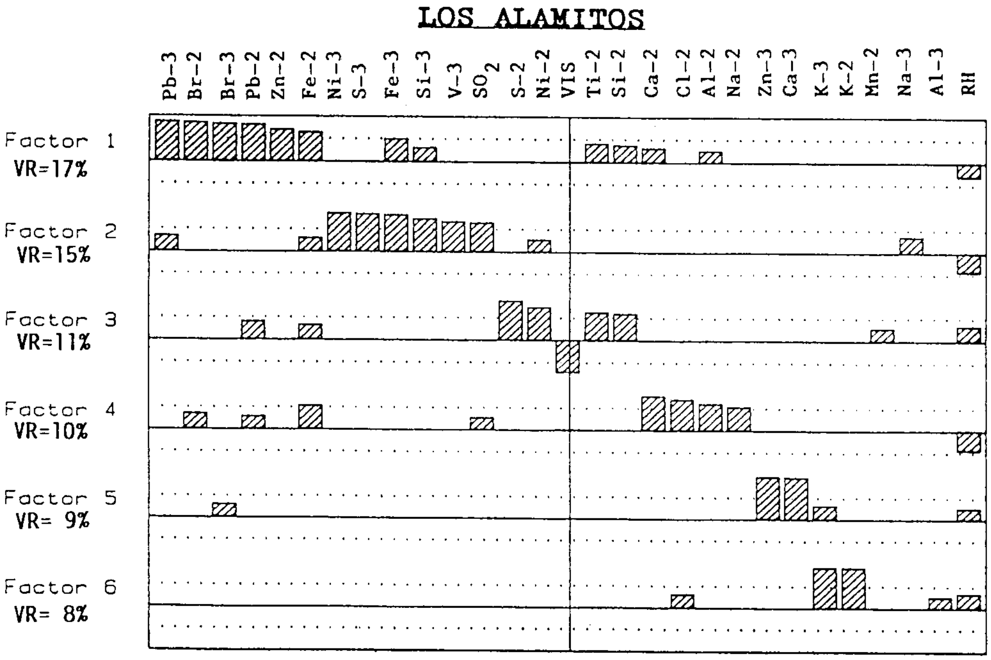

Fig. 9. Sorted rotated factor loadings, summer 1977, 2 indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

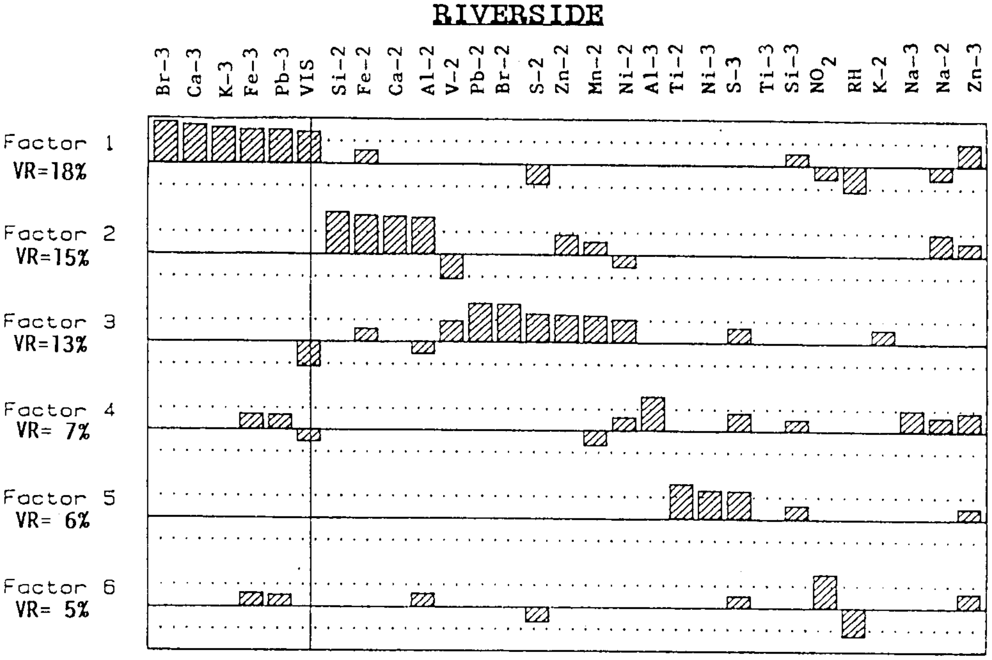

Fig. 10. Sorted rotated factor loadings, summer 1975, 2 indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

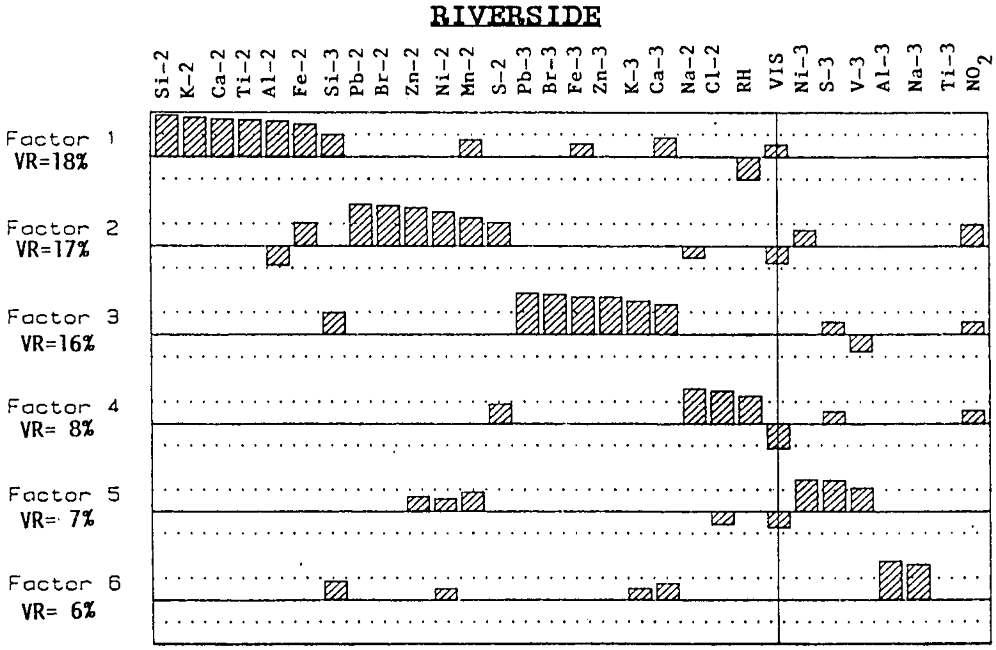

Fig. 11. Sorted rotated factor loadings, winter 1976, 2 indicates intermediate size particles 0.65-3.6 μm, 3. indicates fine particles 0.1-0.65 μm.

Tables 1 and 2 represent summer and winter-time percentage of variance for the probable source type. The results show that in the factor analysis for any of the locations, the soil factor, which is characterized by silicon (Si), calcium (Ca), iron (Fe), aluminum (Al), manganese (Mg), and titanium (Ti), usually appears first. Nickel (Ni) shows variable loadings on this factor at the different locations. The variability of nickel loading on this factor is consistent with the previous suggestion that atmospheric nickel concentrations results from both a soil and nonsoil source. At some locations zinc (Zn) also shows some loading on the soil factor. The loadings of sodium (Na) and potassium (K) are reduced at several locations. The reduced quantities of particles from the soil source have allowed variance from more minor source to be visible.

Table 1 shows that the major sources at the Sacramento site are fine and coarse soil-derived particles, automotive emission, oceanic aerosol, marine aerosol with sulfur, and agricultural burning. Lead (Pb), bromine (Br), and nitrogen dioxide ($N0_2$) are associated with automotive emission. Lead is acknowledged to be an unambiguous particulate tracer for automotive sources in the absence of lead industries. Several studies have indicated a well-aged auto aerosol reaches a Br/Pb equilibrium ratio (mass ratio) of about 0.25 (Lyons *et al.*, 1979). Oceanic aerosol is characterized principally by sodium and chlorine with a typical elemental ratio Na/Cl of 1.13 (Lyons *et al.*, 1979). Transport of suspended particles across the coast range is evident from the presence of these elements in the predicted ratio in the valley. The marine aerosol with sulfur can be responsible for visibility degradation at this site. Visibility reduction is most strongly related to relative humidity and sulfur aerosol. Increase in relative humidity can account for the high sulfur concentration (SO_2 to SO_2^{-2} conversion) at this site.

The agricultural burning is usually characterized by potassium at intermediate and fine size ranges. Agricultural burns generally emit over 90 percent submicron particles. The Sacramento

Table 1. Summer-time percentage of variance for the significant factors

Factor	Sacramento (1977)	Bakersfield (1977)	San Jose (1977)	Los Angeles (1977)	Los Alamitos (1977)	Riverside (1975)
Fine Soil Derived	17%	****	****	14%	****	****
Coarse Soil Derived	15%	20%	18%	****	11%	15%
Automotive	14%	18%	25%	20%	17%	18%
Marine Aerosol	10%	****	11%	****	10%	****
Marine Aerosol &Sulfur	9%	10%	9%	16%	11%	13%
Agricultural Burnig	7%	8%	8%	10%	8%	****
Fuel Oil	****	7%	****	9%	15%	6%

****** indicates no factor**

Table 2. Winter-time percentage of variance for the significant factors

Factor	San Jose (1977)	Los Angeles (1977)	Los Alamitos (1977)	Riverside (1976)
Fine Soil Derived	****	9%	14%	****
Coarse Soil Derived	28%	26%	24%	18%
Automotive	23%	20%	16%	17%
Marine Aerosol	5%	5%	6%	5%
Marine Aerosol &Sulfur	15%	****	****	8%
Fuel Oil	****	15%	11%	7%

****** indicates no factor**

Valley in California is periodically inundated with aerosols produced by open-field agricultural burning. These episodes are primarily products of the burning of waste rice straw, and reach peak concentrations in October and November when the need for burning coincides with generally stable atmospheric conditions. During these periods, state and federal total suspended particulate standards are often exceeded, and visibility is drastically reduced. The impact of agricultural burning was determined by decreased visibility (from both visual and nephelometer readings). However, these visibility measurements represent optical extinction by all particles, rather than that produced exclusively by the agricultural burning. Thus, a low visibility reading at Sacramento may be representative of urban pollution rather than agricultural emissions. Similarly, low visibilities at more rural sites may be products of transport from Sacramento or Bay Area sources. It should be mentioned that the unique potassium signature of the rice smoke, however, permits an extensive and accurate estimation of the extent of the agricultural plumes influence at Sacramento.

At Bakersfield, the major sources are coarse soil-derived particles, automotive emission, marine aerosol with sulfur, agricultural burning, and fuel oil. The inclusion of bromine (Br) in soil-derived particles may indicate the lead contamination results from auto exhaust (Zimdahl and Skogerboe, 1977). This factor could also indicate that the fine soil and the auto exhaust sources are in the same directional transport wind components. At the Bakersfield both intermediate and fine sulfur aerosols and relative humidity are strongly related to reduce visibility. The sulfur aerosols may be due to the conversion of SO_2 to sulfate in the presence of oxidant, as indicated by the high loading from SO_2. The negative loadings on calcium (Ca) and titanium (Ti) are probably indicative of the role wind (wind blown soil) plays on improving visibility. Finally, fuel oil is generally characterized by sulfur with small amounts of vanadium and nickel, which appear unique to this source. In the above cases, the particulate samplers are located at urban air monitoring stations that also supply gaseous pollutant data.

For the San Jose site, the coarse soil-derived particles and automotive emission collectively account for about 42% of the variance. Oceanic aerosols account for only 5% of the variance among the data in winter. At the San Jose site, visibility reduction is most strongly related to sulfur and nickel (a catalyst used in the petroleum cracking process) during the winter of 1977. The strong negative loading on aluminum is probably indicative of a directional transport wind components which cleans the air. Humidity with a fairly high positive loading is also related to visibility reduction. Generally, this factor has higher variance in winter than summer. The negative loading from chlorine suggests that ocean breezes clean the air at the San Jose site during summer 1977. Potassium has been suggested as a tracer for agricultural burning. It has been postulated that acid production also accounts for a significant percentage of the total potassium at this site (Flocchini *et al.*, 1981). This factor accounts for 8% of the variance.

For the Los Angeles site the major sources are fine soil-derived particles, automotive emission, marine aerosol with sulfur, agricultural burning, and fuel oil. During winter the major sources are coarse soil-derived particles, automotive emission, marine aerosol, and fuel oil. The role of particle size should be mentioned at this time. Automotive emission has same variance in summer as in winter. Automotive lead remains firmly in the accumulation mode in summer and winter. Sulfur shows a considerable seasonal variation. The winter sulfur size values appear similar to lead, clearly accumulation mode and quite fine. Summer values, however, grow in size up to the very upper edge of the accumulation mode. During this period, the amount of sulfate more than doubles while the trace elements associated with oil combustion, vanadium and nickel, remain at their winter size profile. Thus, the primary combustion sulfates have a constant accumulation mode size profile, and the new summer sulfate mode is a secondary aerosol. The summer sulfate aerosol appears hygroscopic, growing in size and accelerating gas to particulate conversion via the aqueous state. Generally, there is a sharp rise in scattering effectiveness with increasing particle size until particle's radius is approximately equal to the wavelenght of the incident light beam. The importance of the fine soil aerosol at the Los Angeles site is probably due to wind blown soil aerosols from the nearby valley areas in summer. Agricultural burning also has some variance among the data in summer.

At Los Alamitos and Riverside, Table 1 indicates that the major factors, accounting for about 64% of the variance, contain coarse soil-derived particles, automotive, fuel oil, oceanic aerosol, and marine aerosol with sulfur at the Los Alamitos site. In winter, the major sources are fine and coarse soil-derived particles, automotive emission, and fuel oil. Sulfur aerosols have the strongest association with reduced visibility among all pollutants at this site. A cluster of power plants and petrochemical operations located near the Los Alamitos site may be the source for these aerosols. At Riverside, the major factors are fine soil-derived particles, automotive emission, and marine aerosol with sulfur in summer and winter. Automotive emission appears to be locally generated aerosol. Soil-derived particles can be an aged aerosol transported from Los Angeles, and hence contributes to the visibility reduction. At this site both sodium and chlorine, relative humidity, and sulfur are strongly related to reduced visibility in winter. Thus, low visibilities are favored by a layer of moist marine air.

Finally, the resulting correlations among species associated with a specific type of source served to identify general impacting source categories. However, no quantitative assessment of source impact is possible. For example, a good correlation between Pb and Br at a particular site would suggest an impact from vehicle exhaust, but it would not indicate how much of an impact that source category had. One aspect of these correlations should be noted: a low correlation between species suggests that the species may not be related to each other. However, a low correlation does not preclude the possibility that there is a relationship between species. It is possible that the true relationship is hidden by sampling errors, the range of variation among the variables, or other interferences.

Conclusions

The data from six air quality stations in California were subjected to a principal component analysis involving elemental, gas, and meteorological variables. The application of principal component analysis to the data set produces statistically independent linear combinations of the original variables. For the great majority of individual principal components analyses performed on these data, the factor explaining the most variance contained such elements as silicon, calcium, iron, and aluminum. The variance that was common to these species was due to their source, the soil. The majority of the principal factors were tentatively assigned to pollution from automobiles, fuel burning, and agricultural burning on the basis of their chemical composition. The result of the principal component analysis also indicated that a marine aerosol was present at every location, and sulfur aerosols were strongly related to reduced visibilities at each of the sites studied. A caution must be expressed, however, in that since the sulfur component association with visibility degradation appears to be dominated by secondary processes, one must anticipate that nitrates and secondary organics may be also causably connected with visibility degradation. High correlations may exist between fine sulfate and nitrate (Matsuda *et al.,* 1986) but no such correlation existed for organic. Thus, one may be able to use secondary sulfates as a surrogate for hard-to-measure secondary nitrates.

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