ENVIRONMENTAL DYNAMICS AT ATHENS: FROM RURAL TO URBAN REGIONS

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During the period of 1901–1940 regular measurements were made of the surface ozone concentration (SOC) at Athens, using De James photosensitive paper, which provided a unique set of observational data for SOC in South–Eastern Europe. Recent studies have shown, however, that the color sensitivity of the colorimetric paper is greatly dependent on the air humidity, exposure time, and concentration of minor atmospheric gaseous constituents which exhibit oxidation and recovery properties. Because of this a correction procedure has been developed and applied to the early observational data. The resulting set of adjusted data has been subjected to statistical processing in an effort to analyze time variations of SOC. A most important result of the analysis is the fact that SOC was found to increase from 20 ppb *in 1901 to approximately* 28 ppb *in 1920 and then to decrease down to* 15 ppb *in 1940. In addition, distinct 12–month, 6–month, and 4–month periodicities have been noted in SOC behavior. When the annual cycle of SOC observed at Athens from 1901 to 1940 and 1987–1990 was compared to similar results obtained at other locations, it became apparent that SOC was higher at Hohenpeissenberg (Germany) from 1971 to 1988 than that at Athens throughout the observation period under review. Conversely, the annual cycle at Arosa (Switzerland) as evidenced by the data for 1954–1958 differs but slightly from that observed at Athens from 1901 to 1940. The phases of the annual cycle of SOC at Montsouris (France) from 1977 to 1910 coincide with those observed at Athens whereas the surface ozone values are, on the average,* 4 ppb *less. The analysis of the surface ozone variation with wind conditions shows that* O_3 *concentration is predominantly determined by the variation in the photochemical ozone production.*

1. INTRODUCTION

An analysis of the results obtained from observations of SOC over the current century revealed a distinct trend towards an increase in SOC even in rural regions of Western Europe. Within the past 100 years SOC roughly doubled.[1,2] Naturally, this fact has aroused a heightened interest in further investigation of SOC trends aimed at estimation of SOC dynamics in different regions. Upon examination of the SOC data obtained at Athens in the early twentieth century (1901–1940) Varotsos and Cartalis[3] have discovered that the daily ozone mixture ratio was about 20 ppb and its maxima (minima) in the annual cycle were observed in April–June (October–December). Considering the extreme fragmentariness of the SOC data early in this century it is to be stated that the large sets of data for SOC over Athens are unique.

A comprehensive analysis of SOC variations in Europe in the latter part of the twentieth century was made by Bozhkov (see Ref. 4) using the evidence derived from several stations. The analysis showed that the maximum diurnal partial pressure of O_3 varied within $1.7–2.3\cdot10^{-3}$ Pa, i.e., it was approximately half as much as that observed nowadays with a distinct annual cycle exhibiting a maximum in April–May and a minimum in October–December. Bozhkov[1] also discovered that for the past two decades there was an average trend towards a ~1% increase in SOC over the course of a year, with the rate of the increase being faster (slower) in November–January (May–October). Low et al.[5] examined large sets of data for SOC at Hohenpeissenberg (1971–1988) and Arkona (1956–1988) and confirmed the existence of the 1% positive trend while discovering considerable short–term fluctuations.

A substantial increase in SOC may be caused by the increasing anthropogenic pollution of the troposphere as evidenced, for example, by a considerable body of observations and computer simulations reported in the technical literature which suggest that the content of nitrogen oxides NO_x is correlated with that of hydrocarbon compounds both in urban and in rural regions.[2–12] Following Liu et al.[13] who examined the ozone production conditions in the urban atmosphere and the effect of the urban sources of O_3 on its regional and global distributions, the SOC fields in rural regions are quite predictable if the sources of nitrogen oxides are known. The contribution of the non–methane hydrocarbon compounds to the surface ozone production was also estimated and found to be extremely high. Hence the observational data for SOC can be used in estimating the content of the precursor gases responsible for the ozone production.

The recognition of oppositely directed trends in tropospheric and stratospheric ozone concentrations[4,14,15]

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brings to the fore the pressing problem of validation of the observational data arrays available. As regards the SOC measurements performed in the late nineteenth – early twentieth centuries, the procedure used for that purpose relied on the colorimetric paper and was far from perfect. Subsequent investigations[14,16] revealed that the color sensitivity of the paper to the ozone content was dependent on the exposure time and relative humidity. However, the development of the appropriate correction procedure has provided a solid basis for correlation and intercomparison between the results obtained from the present–day observations and those dated to the early twentieth century.

Originally the tropospheric ozone production concept assumed that its primary source was stratosphere[17] and that the tropospheric ozone came from exchange processes between the stratosphere and troposphere at the tropopause level, followed by turbulent mixing resulting in the downward transport of O_3 as low as the earth's surface and its subsequent breakup therein. Research into the phenomenon of photochemical smog first carried out in Los Angeles and then elsewhere suggested another O_3 source in the troposphere. Photochemical smog was long thought to be an exclusively local phenomenon, affecting the surroundings only through the long–range transport. Crutzen[19] was the first to prove that smog reactions accompanied by oxidation of methane and other natural hydrocarbon compounds could be likewise responsible for the photochemical ozone production in the unpolluted atmosphere. While the possibility of stratospheric origin of O_3 in the free troposphere has been borne out by the present–day observations, for example, by experiments conducted within the framework of EUROTRACK, it is amply clear that the ozone production in the boundary layer of the atmosphere (especially near the earth's surface) is largely governed by local photochemical reactions.[19,20]

The main objective of this paper is to perform statistical analysis of unique long–term SOC observations made at Athens from 1901 to 1940 with an eye to revealing certain regularities and periodicities inherent in variations, long–term trends, and short–term fluctuations in SOC. The integrated analysis of the evidence for SOC and wind field is aimed at the elucidation of the photochemical character of the surface ozone production given heavy escape of industrial pollution components identified as precursors to O_3. An important practical aspect of the problem is the fact that oil rather than coal is now used as a power source on an ever increasing scale, which was not the case in the early twentieth century.

2. OBSERVATIONAL DATA

SOC observations were performed at the National Laboratory of Athens from 1901 to 1940, using De James colorimetric paper. Paper strips protected against solar radiation and precipitation were exposed to the ambient air over different periods of time to serve as SOC indicators. A chromatic scale employed 21 gradations of color varying with the ozone concentration. The major difficulty encountered in the interpretation of the ozone data was due to the need to account for color variations depending on the exposure time and relative humidity which were subsequently found to be quite appreciable.[3,14,16]

Figure 1 illustrates the relationship between the Schönbein color index and SOC at a relative humidity of 80%. According to the data reported by Kley et al.[14] and shown in Fig. 1 to a color index of 6 there corresponds SOC of 10 ppb, whereas Fig. 2 (see Linvill et al.,[16] solid line) gives SOC equal to 50 ppb. The dramatic overestimation of SOC is due to the fact that, in the construction of the relevant correlation plot, the color variations of the colorimetric paper with the relative humidity alone were accounted for while the effect of the exposure time was disregarded. However, the comparison between the two figures indicates that it is a factor of great importance.

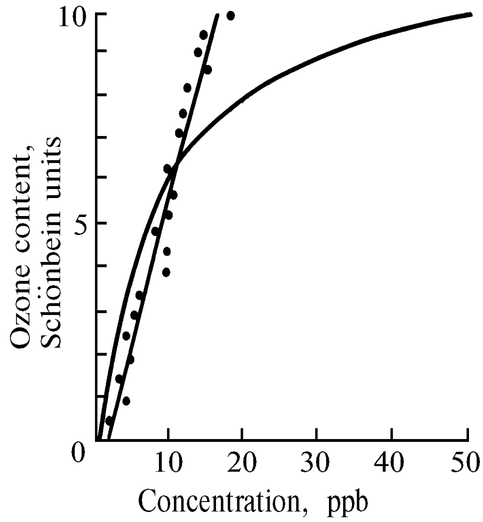

*FIG. 1. Relationship between the Schönbein index and SOC (*ppb*) at a relative humidity of* 80% *(straight line running through the points). Solid line corresponds to UV spectrophotometric measurements reported by Kley et al.*[14]

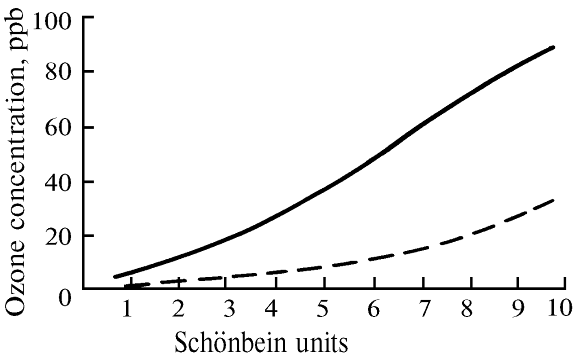

*FIG. 2. New estimates of the relationship between Schönbein index and SOC (*ppb*) at a relative humidity of* 80% *(dotted curve).*

Assuming that there is a match between Schönbein and Salleron scales (both of these scales have 21 color gradations) a comparison was made between the De James and Schönbein scales, using a unique set of SOC observations at Montsouris[21] to show the validity of the new correlation plot taking into account the dependence of SOC on the relative humidity at a fixed exposure time of 12 hours. In doing so, the relationship was specified between the De James scale and the evidence from direct measurements taken by Varotsos and Cartalis[3] (see the dotted curve in Fig. 2).

A similar calibration made it possible to obtain a revised set of observational data for SOC at Athens from 1901 to 1940. Full details favoring this evidence are given in Ref. 3. Figure 3 illustrates the importance of the data correction under consideration. It is obvious from the curves that the maximum (minimum) improved ozone values were observed in April–June (October–December) while the uncorrected values exhibit a reverse annual cycle.

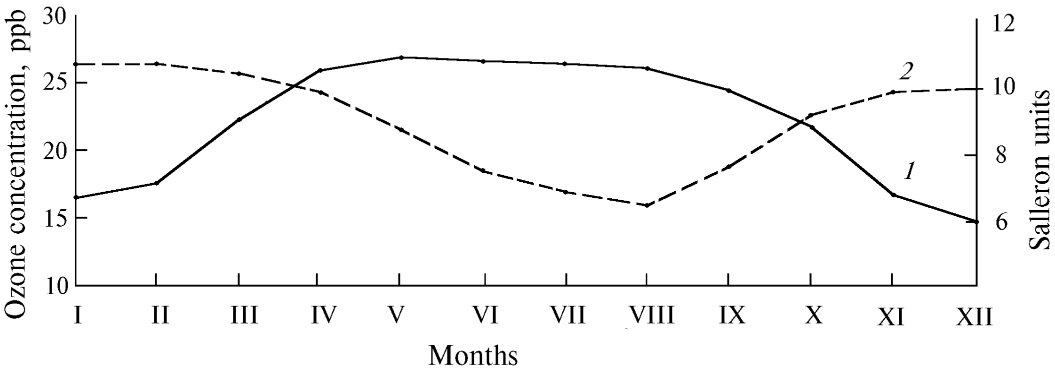

FIG. 3. Average annual cycle of SOC at Athens observed from 1901 to 1940. Curves 1 and 2 illustrate adjusted and original (uncorrected) data, respectively.

3. OBSERVATIONAL DATA PROCESSING PROCEDURE

3.1. Trend analysis. Daily observational data for SOC were processed to explore regular trends in the SOC annual cycle at Athens and compare those results with the evidence from other locations. To estimate the background SOC the entire series of long–term ozone observations was examined. A comparison with the data for other stations gave an insight into special features inherent in the spatiotemporal variations of the SOC field (see Ref. 3).

3.2. Harmonic analysis. The stage that followed involved further individual inspection of the daytime and nighttime observational data. Statistical processing of the daytime data for 1901–1940 provided values for the amplitude, phase, and relative contributions of the first three Fourier harmonics to the overall variability. The intention of the harmonic analysis was to reveal the most important periodicity components, compare their contributions to the overall variability, and verify the correction procedure for the original data set for SOC. In doing so, use was made of only improved values for SOC. The data related to the early twentieth century formed the basis for estimation of the background SOC, which furnished a means for a more plausible assessment of the role of photochemical processes in the surface ozone production nowadays.

4. RESULTS AND DISCUSSION

4.1. SOC variations. Figures 4*a* and *b* depict regular trends in monthly average values of SOC at Athens from 1901 to 1920 and from 1921 to 1940. During the first period of observations there was a clear linear trend towards an increase in SOC followed by some decrease in the second period. This evidence is supplemented with Fig. 5, illustrating average annual values of SOC in those time intervals. The average SOC tended to grow from approximately 20 ppb in the early twentieth century to 28 ppb twenty years later, while from 1921 to 1940 there occurred a drop in SOC down to roughly 15 ppb in 1940.

It is of interest to compare the annual cycle of SOC observed at Athens from 1901 to 1940 and from 1987 to 1990 with large sets of data obtained from other locations: Montsouris (1877–1907), Hohenpeissenberg (1971–1988), and Arosa (1954–1988). From 1987 to 1990 nearly continuous records (every 30 min) of SOC over Athens were kept at three stations of the National Atmospheric Pollution Monitoring Service. The stations located in close proximity to the National Observatory of Athens measured SOC by means of ultraviolet photometers with a detection threshold of 0.1 ppb. All three monitoring stations were situated in the central part of Athens and had to endure the effect of photochemical processes operating in the polluted urban atmosphere.

An intercomparison of data for the annual cycle of SOC observed at Athens (Fig. 6) from 1901 to 1940 and from 1987 to 1990, on the one hand, and recent measurements made in different areas in Western Europe (Montsouris, Hohenpeissenberg, and Arosa), on the other, bears witness to a more vigorous ozone production under polluted atmospheric conditions.

An average increase of the order of 20 ppb was observed at variations between 10 and 30 ppb. The surprising thing is that SOC at Hohenpeissenberg (1971–1988) was found to be higher than at Athens (1987–1990), where the atmosphere is heavily polluted. This can be attributed to the fact that the observatory of Hohenpeissenberg is located a short distance from a big industrial city like Munich, where surface ozone precursors are "generated" and transported towards Hohenpeissenberg.[22]

Figure 6 also shows a number of other interesting peculiarities. Among other things, SOC featured higher values from May to September of 1987–1990 than during the period of 1901–1940, while the reverse trend occurred in the rest of the months. SOC recorded at Arosa from 1954 to 1958 approaches that registered at Athens from 1901 to 1940 even though it is somewhat lower than at Athens from June to December. The time where SOC reaches its maximum varies within approximately two months, ranging from May (Athens, 1901–1940; Montsouris and Arosa) to July (Athens, 1987–1990, Hohenpeissenberg).

A fascinating peculiarity of SOC at Athens is a comparatively small difference between SOC levels observed from 1901 to 1940 and 1987–1990, which is associated with the use of average diurnal values for SOC. If only morning observations of SOC are considered, the difference rises sharply, with SOC from 1987 to 1990 being twice as much as that found from 1901 to 1940 (Fig. 7). The difference in the magnitude of SOC accounts for the present–day enhancement of photochemical processes responsible for the ozone production when ozone precursors like nitrogen oxides and hydrocarbon compounds are discharged into the atmosphere.

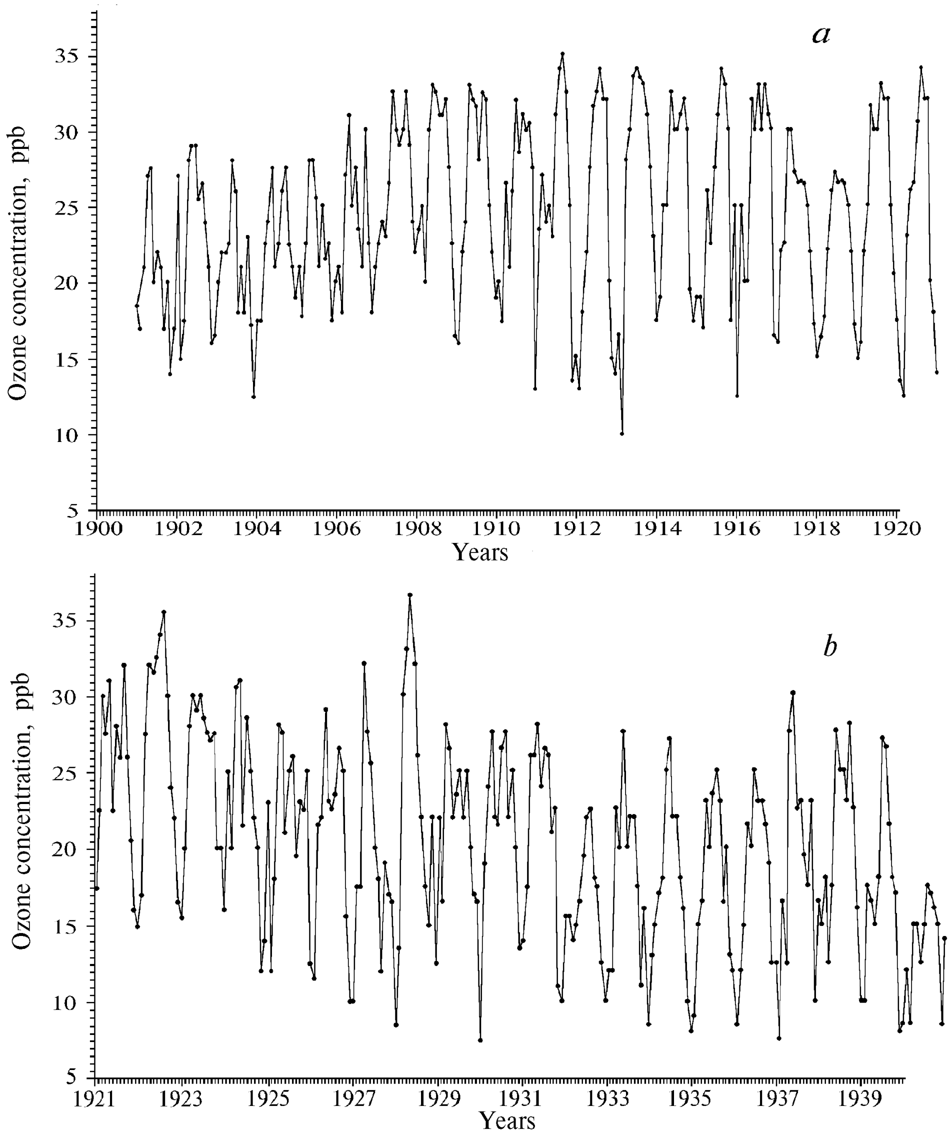

FIG. 4. Adjusted monthly average values for O_3 *mixture ratios at Athens from 1901 to 1920 (a) and from 1921 to 1940 (b).*

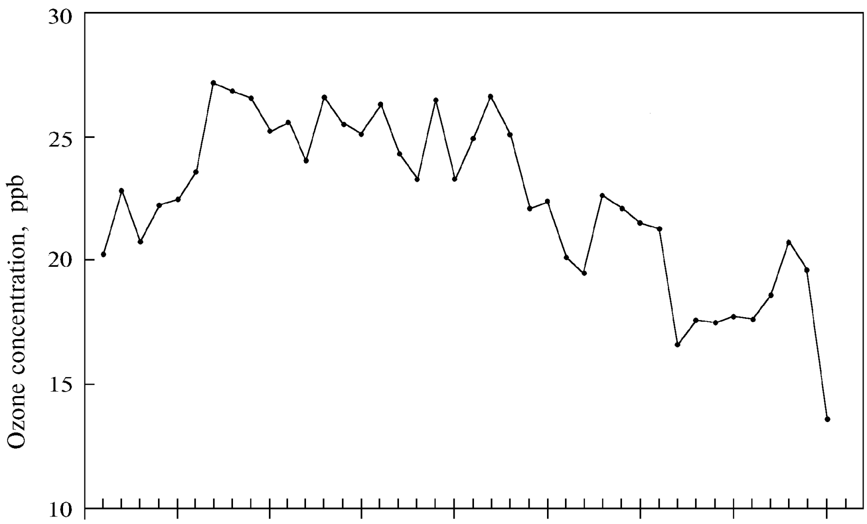

FIG. 5. Adjusted annual average values for O_3 *mixture ratios from 1901 to 1940.*

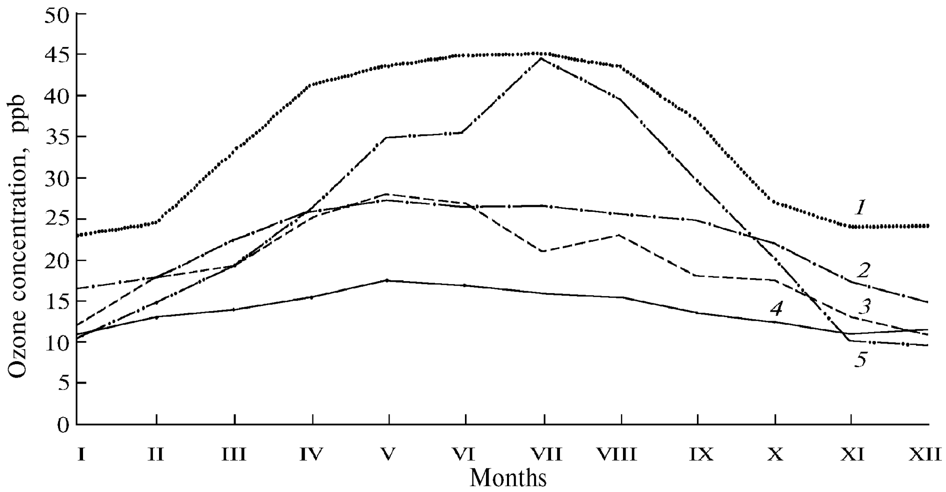

FIG. 6. Intercomparison between the annual cycles of O_3 *mixture ratios at different localities as averaged over different periods of time: Curve 1 – Hohenpeissenberg (1971–1988); curve 2 – Athens (1901–1940); curve 3 – Arosa (1954–1958); curve 4 – Montsouris (1877–1907); and, curve 5 – Athens (1987–1990).*

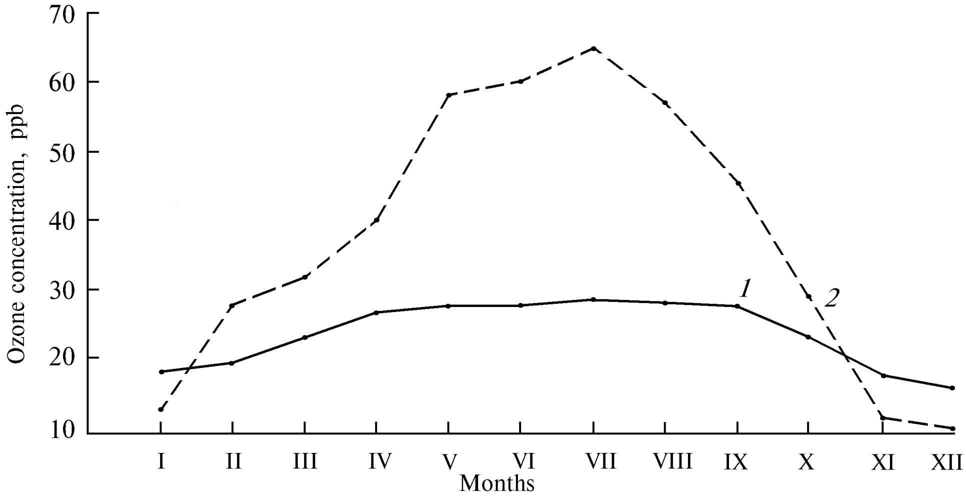

FIG. 7. Annual cycles of adjusted mixture ratios of O_3 *observed in early hours of the day at Athens from 1901 to 1940 (1) and from 1987 to 1990 (2).*

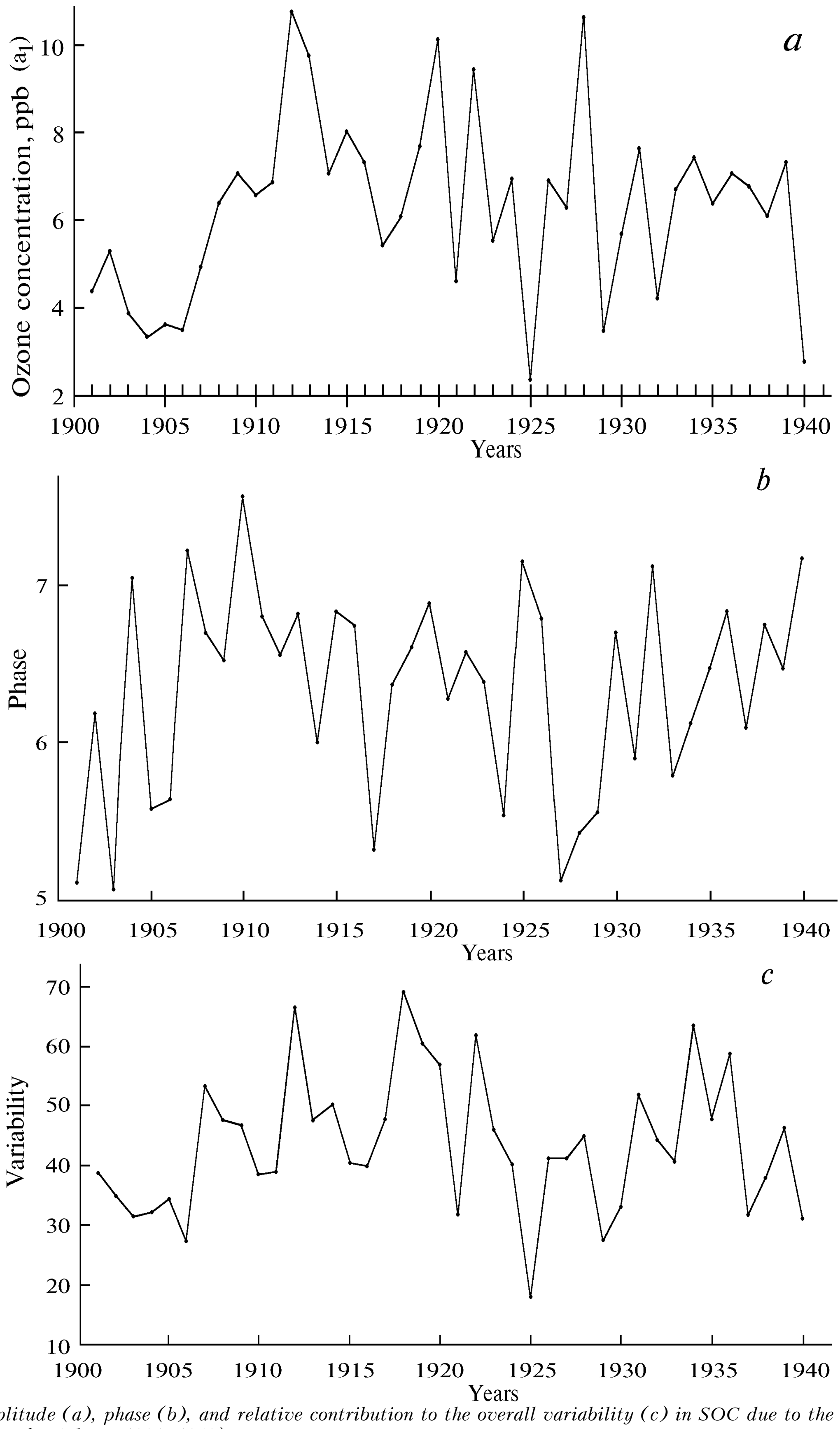

FIG. 8. Amplitude (a), phase (b), and relative contribution to the overall variability (c) in SOC due to the annual cycle (adjusted data for Athens, 1901–1940).

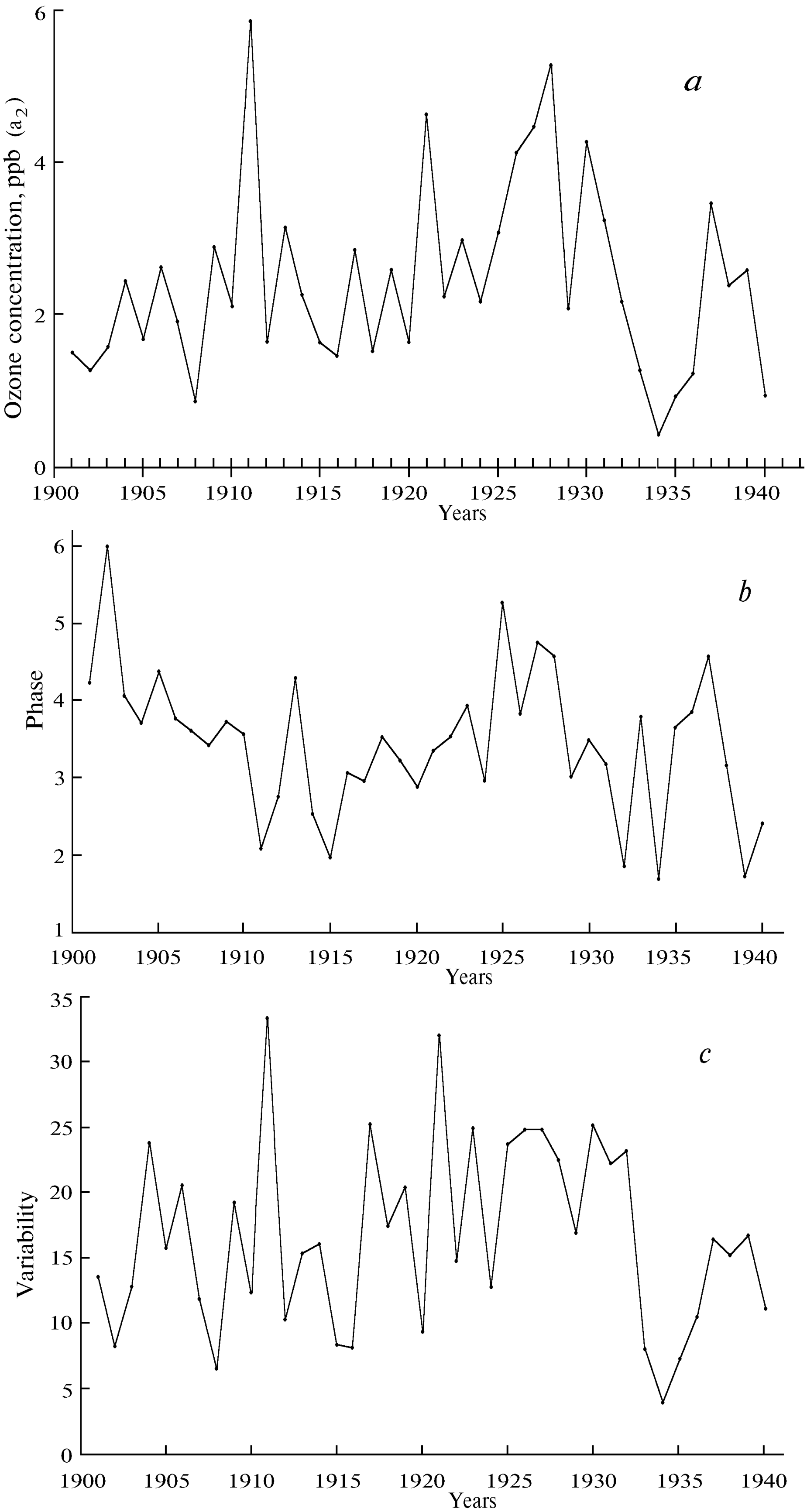

FIG. 9. Amplitude (a), phase (b), and relative contribution to the overall variability (c) in SOC due to the 6–month cycle (adjusted data for Athens, 1901–1940).

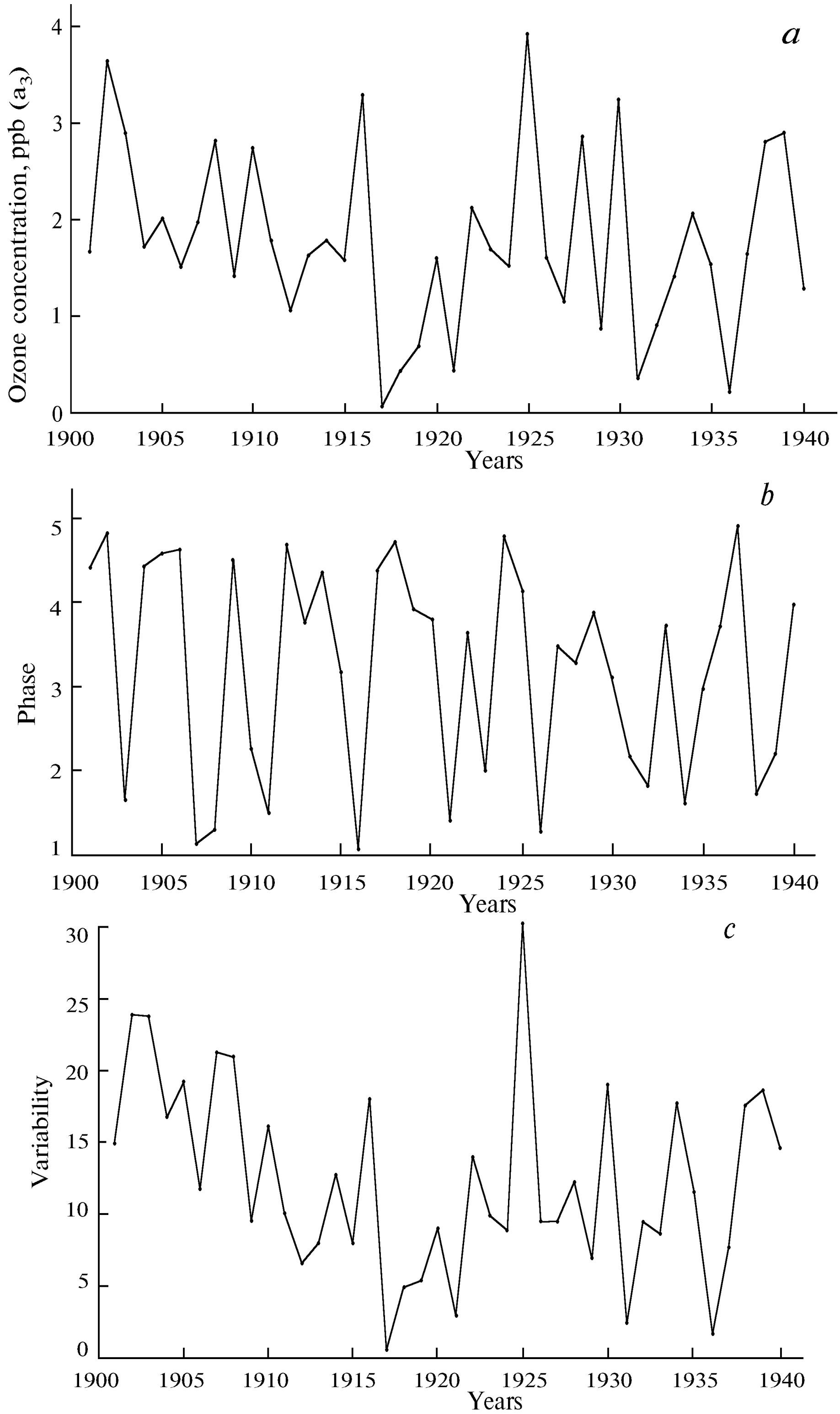

FIG. 10. Amplitude (a), phase (b), and relative contribution to the overall variability (c) in SOC due to the 4–month periodicity (adjusted data for Athens, 1901–1940).

A later data processing stage included statistical analysis of the morning SOC values for the purpose of calculating the amplitude, phase, and relative contribution of the first three harmonics in the overall variability. The results obtained are given in Figs. 8 through 10. The plots explicitly indicate a predominant role for the annual cycle from the viewpoint of the amplitude and relative contribution to the overall variability, which is reasonable given the observational data available. A more interesting result is the fact that, for some subperiods (1901–1903, 1907–1908, 1924–1928, 1933–1940) the four–month wave (third harmonic) appeared to be higher in magnitude and its contribution to the overall variability greater than that of the six–month cycle. The time where SOC peaks (referred to as phase) varies between May and August (with the highest recurrence period being in July) for the first harmonic (Fig. 8*b*) and between March and April for the six–month cycle (Fig. 9*b*). In the case of the third harmonic, drastic phase changes during the first five months of the year make it difficult to identify the time where SOC is at a maximum (Fig. 10*b*).

A similar harmonic analysis was performed for monitoring the data obtained at three stations in Athens from 1987 to 1990. To this end, only the highest values for SOC were selected for examination. As before (for a few subperiods from 1901 to 1940) the amplitude and relative contribution to the overall variability due to the third harmonic appeared to be comparable with the corresponding values for the six–month cycle. A maximum in the annual cycle of SOC was observed in July and with the six–month cycle it occurred one month earlier than from 1901 to 1940. The third harmonic is characterized by phase variability during January–April.

Since the size of the data set for SOC observations at Athens has been limited in recent years, no analysis of trends for this period has been made. By the same reasoning, care should be exercised in the estimation of trends on the evidence derived from 1987 to 1990.

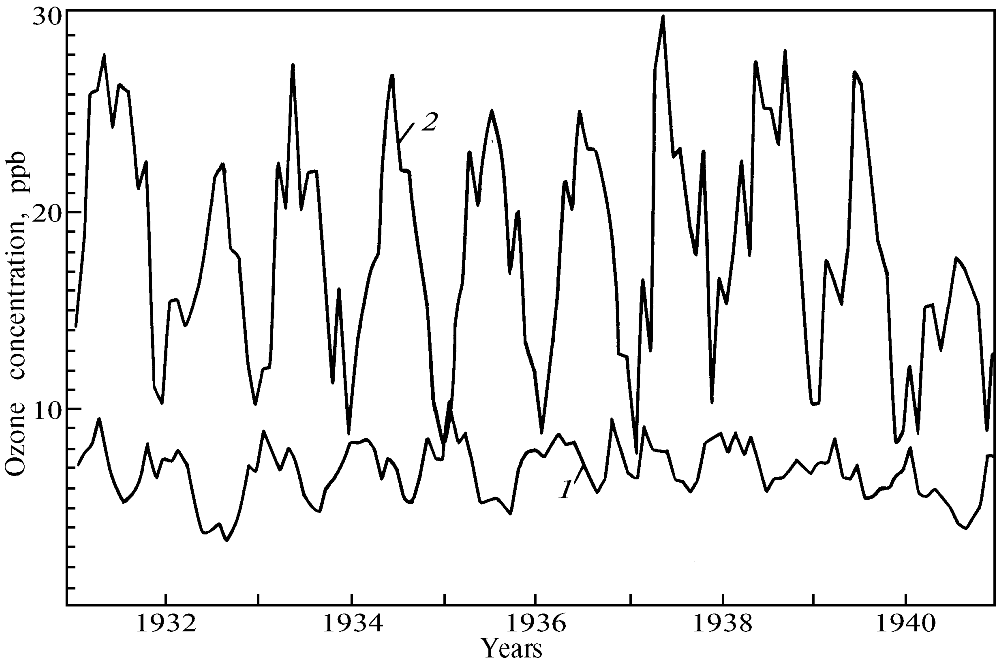

FIG. 11. Monthly average values for SOC at Athens (1901–1940).

4.2. Wind effect on SOC variations. Figure 11 shows monthly average values for SOC at Athens from 1931 to 1940 obtained from original (curve *1*) and adjusted (curve *2*) data.[3] The corrections were introduced for the effects of the relative humidity and exposure time. The main conclusion to be reached from these data is that SOC exhibits a distinct annual cycle with a maximum in summer and a minimum in winter which accounts for the anthropogenic impact on SOC in summer. The peak values for SOC are approximately three times higher than the minimum values which is due to heavy discharges of anthropogenic ozone precursors, like nitrogen oxides or hydrocarbon compounds, coupled with intense insolation. These combined effects stimulate the vigorous photochemical processes which govern ozone production.[23,24] Early in the twentieth century there was scarcely any annual cycle in SOC under unpolluted background atmospheric conditions.[2]

In the early 1920s Athens was a big city with a population of 700 000 people where industries developed at a fast pace.[25] For the most part, industrial enterprises were located to the west of Piraeus harbor (Fig. 12). Undeniably the industrial development, along with wide use of vehicles and heating systems, resulted in increased ejection of pollutants identified as ozone precursors which, in turn, was liable to give rise to a pronounced annual cycle in SOC typical of a medium–size modern city. This is supported by the evidence for SOC derived in the vicinity of the National Observatory of Athens, located on a hill (107 m above the sea level) in close proximity to the center of the city. Figure 13 presents wind rose in that area (variation in the recurrence rate of the wind direction). The data take no account of wind velocities lower than 2 m/sec. It is obvious from the picture that three wind directions were found to prevail as determined by statistics on the meteorological conditions and effect of the local topography. A distinguishing feature of the synoptic situation characteristic of the area in question is that Etesian winds blowing from the north/north–east tend to persist from mid–June until late August. It is precisely this phenomenon which governs the principal distinctive property of wind rose (Fig. 13).

Of frequent occurrence at Athens (in ~30 percent of instances, predominantly in spring and autumn) is the breeze situation where the synoptic condition is characterized by a scant wind combined with an intense insolation.[26] The prevailing breeze direction is south–east. Figure 14 shows three "ozone roses" (wind directions associated with the most vigorous ozone production), namely, for 1931–1940 (curve *1*), 1931–1932 (curve *2*), and 1939–1940 (curve *3*). The observational data obtained throughout the period under review display a marked lack of homogeneity. For example, peak values for SOC (nearly 22 ppb) were detected in the case of the west wind, decreasing down to 17–18 ppb with Etesian winds and breeze circulation with the north/north–east and north–east or south–west and south/south–west winds prevailing. Very low values for SOC were recorded under the south–west winds.

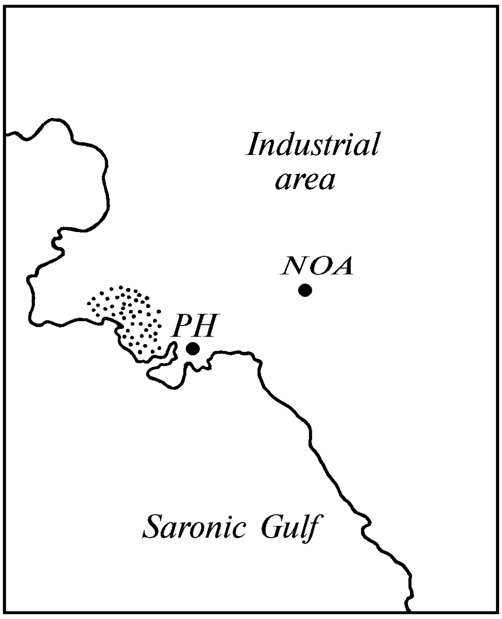

FIG. 12. Map of Athens and vicinities: NOA is the National Observatory of Athens and PH is Piraeous harbor.

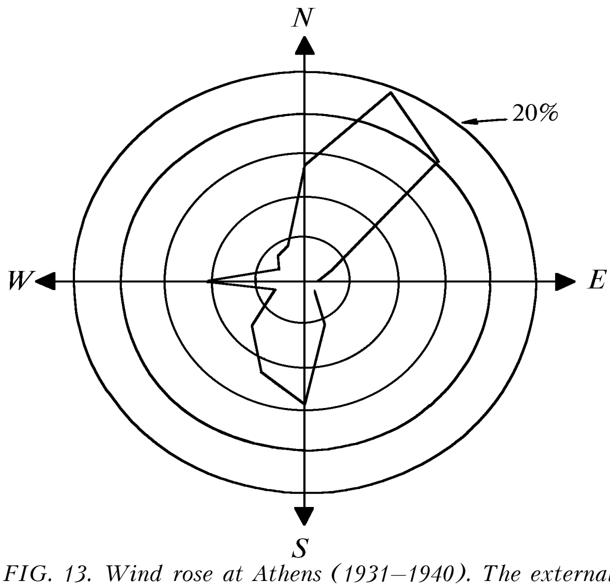

FIG. 13. Wind rose at Athens (1931–1940). The external circle shows a wind recurrence of 20%.

When analyzing the data for "ozone roses" in terms of the photochemical ozone production conditions, it is essential to bring to mind the existence of an industrial area to the west of Piraeus harbor. If the west winds blow, the National Observatory of Athens appears to be in the way of the prevailing pollution transport from the industrial area. In these circumstances it is vital to assess the contribution of anthropogenic photochemical processes to the ozone production, using the observational data for SOC (among which are background values for SOC corresponding to the north/north–east and north–east winds). An important point is that the two wind directions generally prevail. SOC is found to increase under the west winds in spite of the oppositely directed action of the sulfur dioxide discharge in the industrial area.[27]

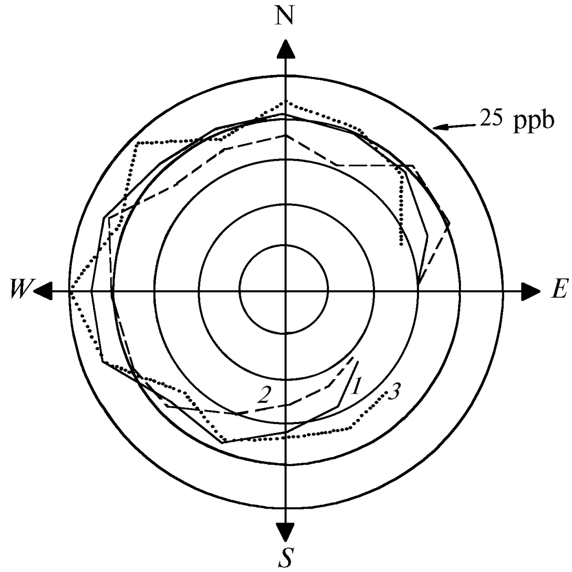

FIG. 14. Ozone roses at Athens: curve 1 – 1931–1940; curve 2 – 1939–1940; curve 3 – 1931–1932. The external circle shows a mixture ratio of 25 ppb.

As discussed earlier for the south–west and south/south–west winds blowing under breeze conditions, the SOC level is as low as in the case of Etesian winds. This fact is of great importance because, as shown in a number of papers, sea breezes act to enhance photochemical processes responsible for the ozone production in big cities located in the coastal regions.[27,28] In these conditions ozone precursors are transported by the breeze over the sea, where ozone is produced in the early hours of the day and then recycled back, which favors an increase in SOC as opposed to the case of no breeze. It remains to be seen why a similar situation does not occur at Athens, where the "signal" accompanying the enhanced photochemical ozone production is lacking. This may be due to the fact that the amount of ozone precursors discharged into the atmosphere was moderate, which is why the intensity of photochemical reactions was inadequate for the ozone production. Furthermore, it is not easy to understand the reasons for the extremely low level of SOC (7–10 ppb) under the south/south–east, south–east, and south winds. It is not inconceivable that, in this instance, the results were not statistically substantiated for lack of observational data.

Peculiar features of the "ozone roses" from 1931 to 1932 (dotted line) and from 1939 to 1940 (dashed line in Fig. 14) proved to be similar, even though in the former period the SOC level was persistently higher than in the latter. This effect is most conspicuous in the case of those wind directions (south/south–east, south, north, north/north–east, and west) which are associated with a high SOC level. This phenomenon was caused by the development of Athens in the 1920s when both power consumption and car use increased. SOC is normally lower in the centers of big cities than in the suburbs (the representative data for SOC at Athens have been cited earlier in this paper). Such SOC behavior is conditioned by the effect of nitrogen oxide ejections into the atmosphere (for the most part as exhaust gases), leading to a very fast ozone breakup.

5. SUMMARY

SOC in the vicinity of the National Observatory of Athens was monitored from 1931 to 1940. The results obtained from interpretation of the observational data indicate that variations in SOC are attributable to anthropogenic enhancement of photochemical processes which govern ozone production when variations in the wind direction are also considered. The data array in question constitutes a wealth of information on the evolution of the background SOC during the period under review. These data acquire great importance in view of the fact that SOC observations with the use of colorimetric paper were discontinued in the majority of areas during World War II, and it was some time later when work on the project was resumed using more sophisticated procedures based on electrochemical and spectroscopic measurements.

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