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Supplement of

Long-term visibility variation in Athens (1931–2013): a proxy for local and regional atmospheric aerosol loads

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1. **The WMO visibility scale: averaging procedure and uncertainties**

Historical visibility observations involve large uncertainties inherent to subjectivity of different operators over the years, but also the scale used for the different visibility ranges. The WMO scale (Table 2 of the manuscript) which was adopted at NOA, introduces further uncertainties, associated with the different amplitudes of visibility ranges corresponding to each visibility class, approximating a geometric-like distribution.

By plotting visibility ranges (upper/lower and middle distance) against visibility classes in WMO scale, we resulted in exponential relationships, consistent to the geometric-like distribution of visibility ranges. Figures S1/S2 show the exponential relationships between the upper/middle distance of visibility ranges, respectively, and the corresponding visibility classes (Table 2 of the manuscript). The coefficients of determination (R^2) are equal to 0.998 and 0.988 for the upper and middle distances, respectively (Fig. S1, S2).

As already stated, the observation procedure requires that an operator scans the horizon for predetermined objects. In the case of Athens, historical buildings (or other objects in the city), but also certain objects of the surrounding landscape, unaltered over the years, (e.g. objects on the mountains or the islands of the Saronic Gulf, see map of Fig. 1 of the manuscript), were chosen to represent different visibility classes. For instance, the building of the Greek Parliament in Syntagma Square was chosen to represent the WMO visibility class 3 (500 -1000 m, following Table 2), an historical hospital to represent visibility class 4 (1-2 km), the dome of the largest church in Athens (built in 1929) to represent class 5 (2- 4 km) etc. Higher visibility classes were determined from objects on mountains or islands of the Saronic Gulf (e.g. Aegina island, see Fig. 1 of the manuscript).

The most obvious and safe procedure to transform visibility classes into distances would be to consider the middle distance in the ranges of WMO scale. For instance, visibility class 5 would correspond to 3 km. However, by comparing the true distances between NOA site and the predetermined objects, (representative of each visibility class) with the visibility ranges (in km) of WMO scale, we found that the true distances are pretty close to the upper limit of visibility ranges (in km) in WMO scale. For this reason, the exponential relationship corresponding to the upper limit (Fig. S1) was adopted. Nevertheless, a sensitivity test was also performed, to quantify differences between the two choices (upper and middle distance of the visibility ranges) on the time series of the annual visibility.

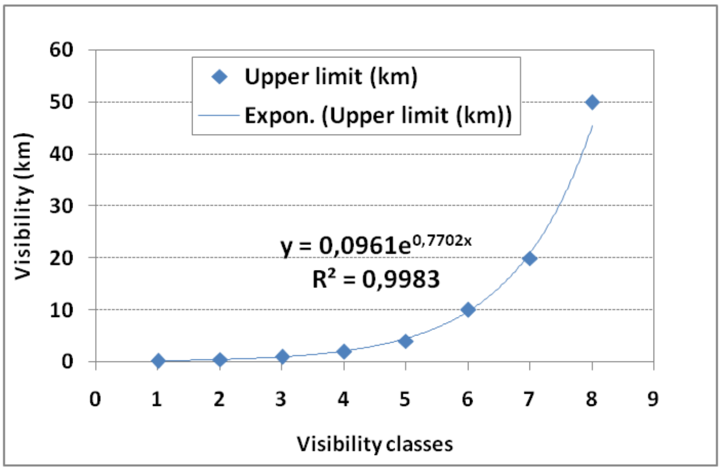

Figure S1: Relationship between the upper distance of visibility ranges (km) and visibility classes in WMO scale (Table 2 of the manuscript).

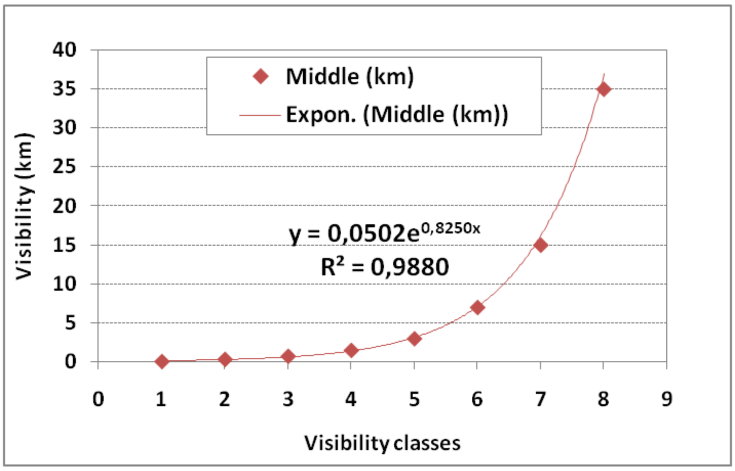

Figure S2: Relationship between the middle distance of visibility ranges (km) and visibility classes in WMO scale (Table 2 of the manuscript).

The following averaging procedure was then followed in the study:

A mean value (arithmetic mean) was estimated for each month, by averaging corresponding daily classes of visibility (1 to 9) for this particular month. This resulted into an 'average visibility index' for each month. The values of this average monthly index ranged from 8.01 (August 1935) to 4.43 (November 2011), with 95th, 50th and 5th percentiles corresponding to 7.53, 6.41 and 5.16 respectively. The exponential relationship reported in Fig. S1 was then applied to transform average monthly values of visibility in km. It is noted that (based on exponential properties) the same results come up, if daily visibility classes are transformed to daily distances (km) using the exponential relationship, and then daily distances are averaged geometrically. Actually, the procedure corresponds to a geometric averaging of daily distances.

A sensitivity test was also performed to evaluate relative differences in visibility time series from the choice of the middle or the upper limit in WMO scale. The time series of the annual visibility were created using the corresponding exponential equations in Figs. S1, S2. Figure S3 illustrates the two time series along with their normalized difference (black line). The normalized difference of annual visibility between the two cases ranges between 20% and 30%, increasing with decreasing visibility. An excellent correlation is observed between the two data sets (R^2=0.99) with middle/upper slope of 0.77 indicating again that our conclusions on visibility trends are valid.

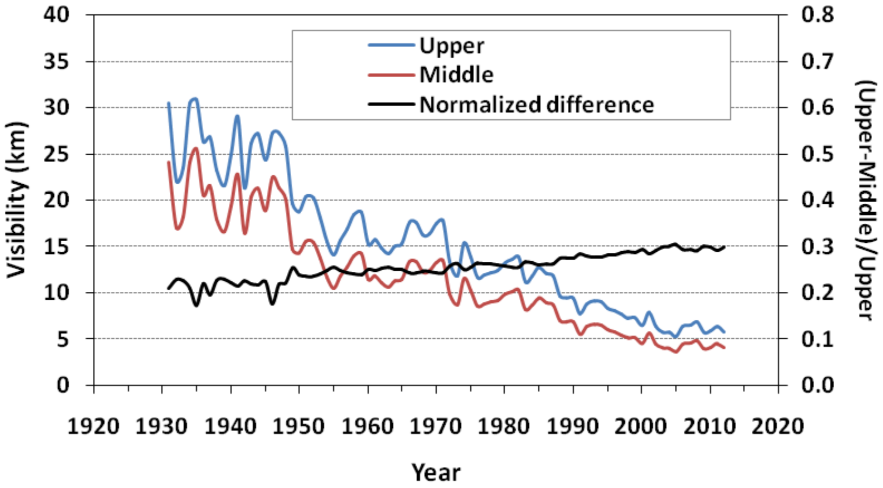

Figure S3: Variation of the annual visibility at NOA from 1931-2013, estimated from the upper distance (blue line) and the middle distance (red line) in the ranges of WMO scale (Table 2 of the manuscript). The black line illustrates the relative difference between the two time series.

2. Visibility and precipitation frequency (PF)

Precipitation (and particularly precipitation frequency, PF) induces scavenging of atmospheric particles, thus affecting visibility levels. Additional time series were created in order to investigate the possible effect of PF on the long-term variation and trends in visibility at NOA. Figure S4 illustrates the time series of two subsets corresponding to ‘rain’ and ‘no-rain’ (precipitation <1 mm d^{-1}) days, along with the annual values (all cases), used in our study. According to climatic values provided in Table 1 of the manuscript, rainy days correspond to only 12% of the total days of a year, thus ‘no-rain’ and ‘all cases’ time series almost coincide. It is concluded that our findings concerning long-term variability and trends in visibility at NOA are not affected by precipitation frequency. Subset of ‘rain days’ correspond almost always to lower visibility compared to ‘no-rain’ and ‘all cases’ (Fig. S4).

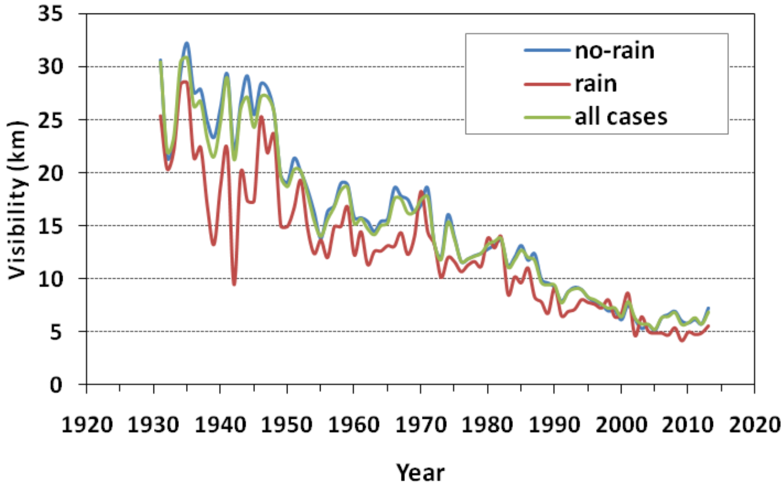

Figure S4: Variation of visibility at NOA from 1931-2013, estimated from subsets of ‘rain’ and ‘no-rain’ days as well as the ‘all cases’ set. A rainy day is defined as a day with total precipitation >1 mm, following WMO.