Authors' response to referees

The authors are grateful to both anonymous referees for their time devoted to this paper and the useful comments and suggestions, aiming at the improvement of the manuscript.

All comments and recommendations of both referees were taken very seriously into consideration for the preparation of a revised version of the manuscript. Additional effort has been put to implement and incorporate suggestions in the manuscript to the best possible degree and prepare a revised version accounting for all comments of referees.

In the following, we present our answers to the referees' comments as well as the changes performed in the manuscript, and a marked-up version of the manuscript (with track changes) in the following order:

A. Comments of referees

A1. Comments of referee #1

A2. Comments of referee #2

B. Authors' answers to the comments of referees

B1. Authors' answers to each comment of referee #1

B2. Authors' answers to each comment of referee #2

C. Changes in the manuscript to account for comments of referees

C1. Changes in the manuscript to account for comments of referee #1

C2. Changes in the manuscript to account for comments of referee #2

D. Marked-up manuscript version

A. Comments of referees

A1. Comments of referee #1

In this article, Founda and coauthors describe the long term trend in visibility in Athens, Greece and compare this trend to meteorological variables, visibility changes at a nonurban site in the area, and satellite-derived aerosol optical depth values. The rapid degradation of visibility after 1950 and slight recovery since 2005 are correlated with meteorological conditions associated with air mass origin, PM10 surface measurements, and aerosol optical depth; these relationships suggest that visibility is a proxy for local and regional atmospheric aerosol levels. This trend and associated analyses provide a novel dataset for understanding long term changes in aerosol concentration near Athens. I'd suggest publication after the following comments have been addressed.

Major comments:

1) While the grouping of 3 periods of visibility trends are appropriate when discussing changes over time, the middle period (1949-2003) is not appropriate when discussing frequencies (Figure 5) and seasonality (Figure 6) because the early part of the period has substantially different visibility conditions from the later period. When not showing a time series, the 1949-2003 period needs to be separated into several periods of more similar visibility conditions.

2) I think that a more comprehensive comparison between emissions changes and visibility trends would help improve the article. Figure 11 needs to have emissions on the y-axis as a magnitude rather than a rate of change, and plotting other types of emissions (NOx, EC, OC, etc) would be interesting to see if available. If the emission data could be segregated by air mass origin, it would be interesting to see if increases/decreases of emissions in certain parts of Europe have affected the visibility in Athens.

3) To add value in the visibility-satellite AOD comparison, I'd suggest examining the much longer-term dataset of AOD values from the Advanced Very High Resolution Radiometer (AVHRR) satellite. Although AVHRR retrieves AOD only over ocean grid cells, selecting the nearest ocean cell to Athens would enable an visibility-AOD comparison since 1981 when visibility values were still degrading.

Minor comment:

1) Many typos and text spacing problems persist in the document and have to be corrected. The first of many are listed by page number; line number (suggested correction): Page 1; Line 18 ("34%"), Page 1; Line 22 ("the 1950s"), Page 2; Line 46 ("containing"), Page 3; Line 82 ("oldest time"), Page 4; Line 90 ("construction"), Page 5; Line 118 ("...the year. The periods..."), Page 5; Line 129 ("Mediterranean"), Page 5; Line 136 ("60%"), Page 6; Line 173 ("Po Valley"), Page 7; Line 180 ("...subsequent reduction in vehicle use..."), Page 7; Line 201 ("with the naked eye."), Page 8; Line 208 ("Davis (1991)."), Page 10; Line 272 ("Overall, visibility did not exceed..."), Page 11; Line 312 ("different approaches, as for instance..."), Page 12; Line 343 ("...resulting in the reduction of visibility."), Page 13; Line 364 ("In other cases..."), Page 14; Line 491 ("increase of construction in the city."). I'd recommend an grammatical editor to correct these and other errors prior to publication in final form.

2) Figure 2a should be referenced in the text before Figure 2b.

A2. Comments of referee #2

General:

The study uses the long-time visibility records along with meteorological variables,emissions and satellite optical depth retrievals over Athens and explores the relationships between these variables over three distinctive sub-periods. The manuscript is clear, well-written with a very good introduction. However, I find the conclusions too long and can be substantially reduced by only pointing to the major outcomes of the study.

Minor comments

Line 237: How far from Athens? Characteristics of the site (emission sources etc)?
Lines 248-254: Better to present the trends in uniform units, per year in this case.

The resolutions of the excel-based figures should be improved.
The relation (zooming) between the two plots in Figure 1 is misleading.
Figure 7: Precipitation Height is misleading, drop the "Height"
Figure 13: Why the different bins have different widths? Does it stand for something?
For instance why 0-0.5 km bin is largest? Please explain.
Figure 15: Can you also add the data for Athens here?

Technical corrections

Line 33: Remove the comma before (WMO, 1992).
Line 38: Replace "at" with "over"
Line 55: : : :. pollutant emissions: : :.
Line 231: Correct as (Kanakidou et al., 2011)
Line 260 and 272: Correct "to 1 km" to "than 1 km"
Lines 261, 266 and 272: Correct "to 500 m" to "than 500 m"
Line 290: ..results IN improvement: : :.
Line 345: Change "as regards" to "regarding"
Line 408: : : :. ARE due to local factors: : :
Line 423: : : :.in accordance WITH: : :
Line 475: INDEPENDENT of the location: : :.

***B. Author's answers to the comments of* referees**

***B1. Author's answers to the comments of* referee #1**

Major Comments

1. Indeed, the grouping of the historical time series was mainly indicated by the different slopes of trends observed in the three sub-periods 1931-1948, 1949-2003 and 2004-2013. It is true that the early part of the much longer sub-period (1949-2003) is characterized by different visibility conditions compared to the latter part. For this reason, the initial grouping was maintained only in trend analysis. In all other cases, namely when studying frequency distribution (Fig. 5), seasonality (Fig.6) but also variation of visibility with wind direction (Fig. 10), the long period 1949-2003, was further divided into two parts, 1949-1975 and 1976-2003. Figures 5, 6 and 10 were reproduced, where the plots concerning the 1949-2003 sub-period, were replaced by plots for the periods 1949-1975 and 1976-2003 (see section C below). The text in the manuscript in sections 3.2, 3.3 and 4.4.2 was revised accordingly, accounting for the new information derived from this additional grouping.

2. Historical data of other types of emissions for Europe such as NOx and OC were also considered and discussed in the manuscript. Plot of rates of changes of SO_2 emissions in Fig. 11 of the manuscript was now replaced with the plot of SO_2 emissions as a magnitude for a more direct comparison with visibility variations. Moreover, a plot of historical NOx emissions for Europe was added in Fig. 11. Details are provided in section C below.

3. We have followed the reviewer's recommendation and used the AVHRR satellite data (available since 1981) in addition to support the current MODIS related analysis concerning AOD and visibility. The additional analysis was incorporated in the manuscript accordingly as described below (see section C).

Minor comments of referee #1

1. Although a grammatical editor had been already used, for some reason it didn't work properly and a number of grammatical and syntax errors remained in the text. Additional effort and a new editor have been used now to cope with this problem. All suggested first corrections by the referee were applied in the text. Additional syntax errors were also found and corrected.

2. This was corrected.

B2. Author's answers to the comments of referee #2

General comments

Section 4 summarizes the findings of the study but also discusses in detail linkage/attribution between the main results of the analysis and possible causes. For this reason, this section is long enough. However, the section was reduced in an effort to focus on the main findings of the study and also avoid duplications.

Minor comments

Line 237: Additional information for the reference station of Finokalia (Crete) was included in the text (see changes in the manuscript, below).

Lines 248-254: This was now corrected in the manuscript.

Some of the excel -based figures were reproduced using a different graphical tool. When not possible, the resolution of excel- based figures was increased.

Indeed, the zooming between the two plots in Fig. 1 is not successful. Fig. 1 was recreated (see below, changes in the manuscript).

Fig. 7: The figure was corrected

Fig. 13: The bin widths are based on the WMO definition on visibility class index. They are not equal as visibility in km and WMO visibility index does not have a linear relationship. The XX' axis is logarithmic.

Fig. 15. PM10 for two stations in Athens from 2004-2014 were added in the figure (see below, changes in the manuscript)

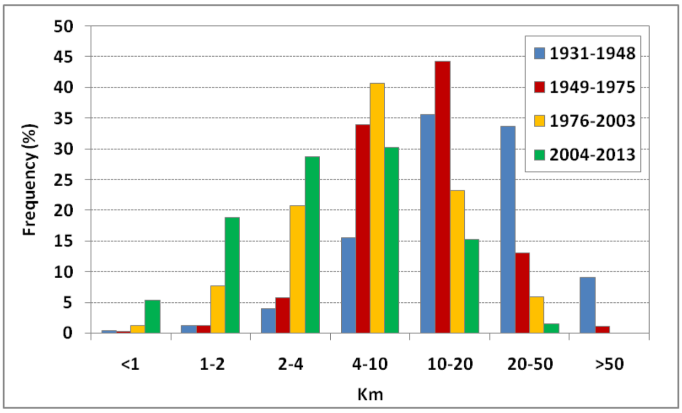
Technical corrections

Although a grammatical editor had already been used, for some reason it didn't work properly and a number of grammatical and syntax errors remained in the text. Additional effort and a new editor have been used now to cope with this problem. All suggested technical corrections were applied in the text.

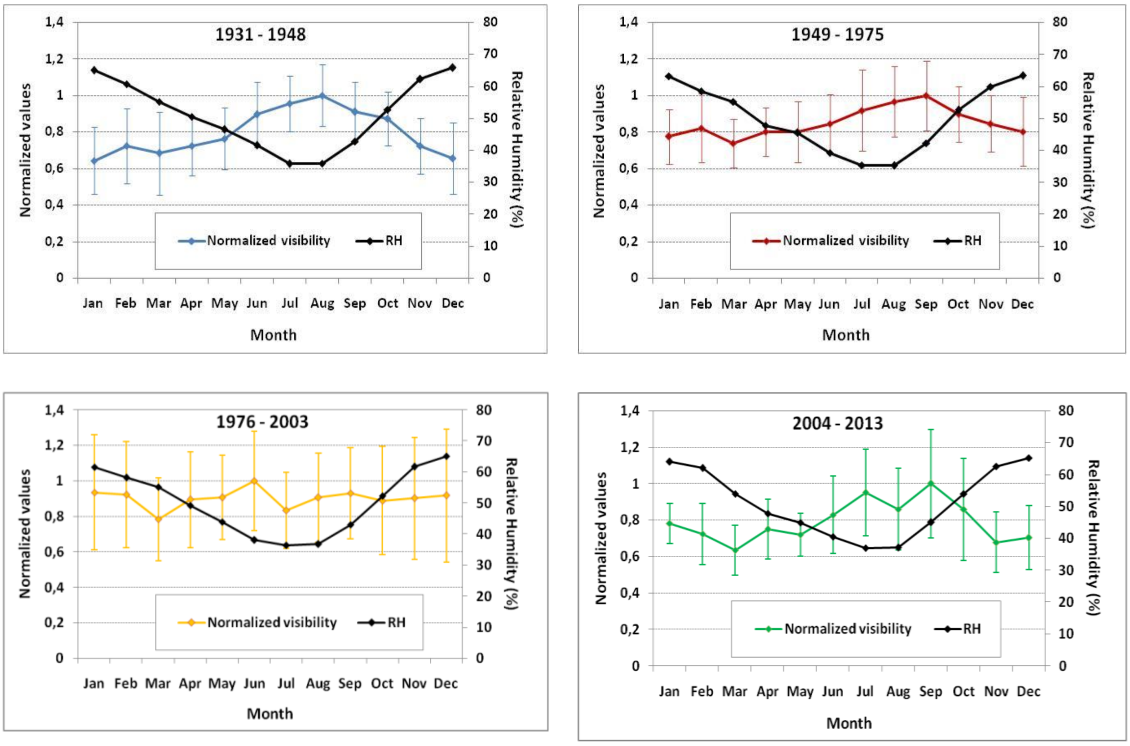
C. Changes in the manuscript to account for the comments of referees

C1. Changes in the manuscript to account for the comments of referee #1

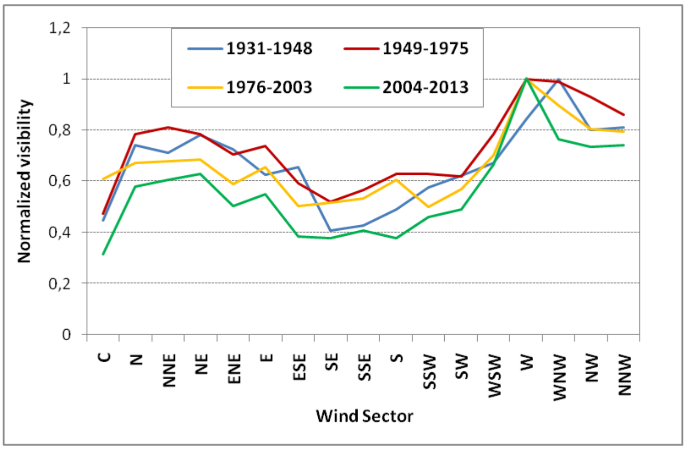
1. In order to comply with major comment 1, Figs 5, 6 and 10 concerning the frequency distribution, seasonality and variation of visibility with wind direction were reproduced. In the new figures, the sub period 1949-2003 was replaced by two additional sub-periods, namely 1949-1975 and 1976-2003. The new Figures 5, 6 and 10 are displayed below



New Fig. 5. Frequency distribution of different visibility ranges (Table 2) in Athens for the sub-periods, 1931-1948, 1949-1975, 1976-2003 and 2004-2013.



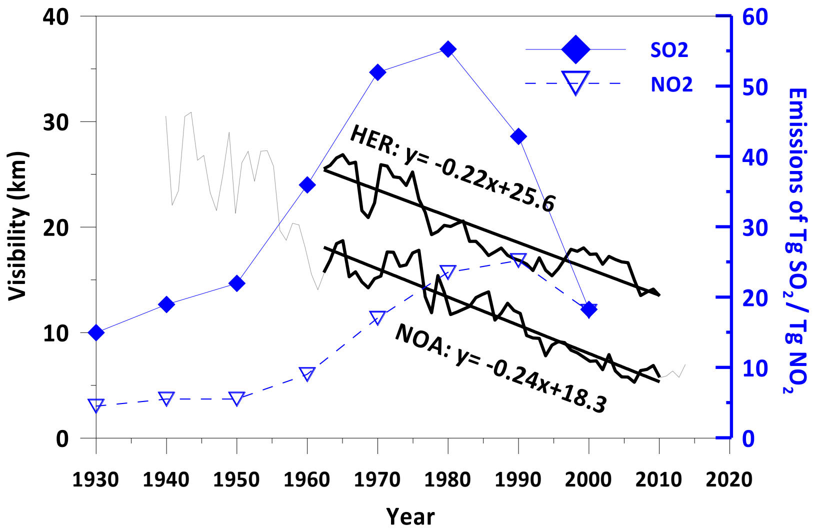
New Fig. 6. Normalized mean monthly values of visibility in Athens for the sub-periods 1931-1948, 1949-1975, 1976-2003 and 2004-2013, along with mean monthly values of relative humidity (RH) for each sub-period. Vertical lines represent standard deviations of monthly visibility means.



New Fig. 10. Variation of visibility with wind direction (sectors) over the sub-periods 1931-1948, 1949-1975, 1976-2003 and 2004-2013. Visibility is normalized by its maximum value at a certain sector for each sub-period. Sector 'C' corresponds to calms (wind speed < 0.3 m s-1). Frequency of each sector approximates closely its climatic value (Fig. 3) in all sub-periods.

The text in the manuscript (sections 3.2, 3.3, 4.4.2 and Conclusions) was revised accordingly, to account for the new information derived from this additional grouping.

2. Historical data of other types of emissions in Europe such as NOx and OC were also considered and discussed in the analysis. Fig. 11 of the manuscript was reproduced. In the new figure, graph of the rates of changes of SO_2 emissions was replaced with graph of historical emissions as a magnitude, for a more direct comparison with visibility variations. Moreover, a plot of historical NOx emissions for Europe was added in Fig. 11. Historical emissions of SO_2 and NOx for Europe were now derived from the studies of Vestreng et al. (2007, Fig. 1) and Vestreng et al. (2009, Fig. 3) respectively since they provide updated emissions data.

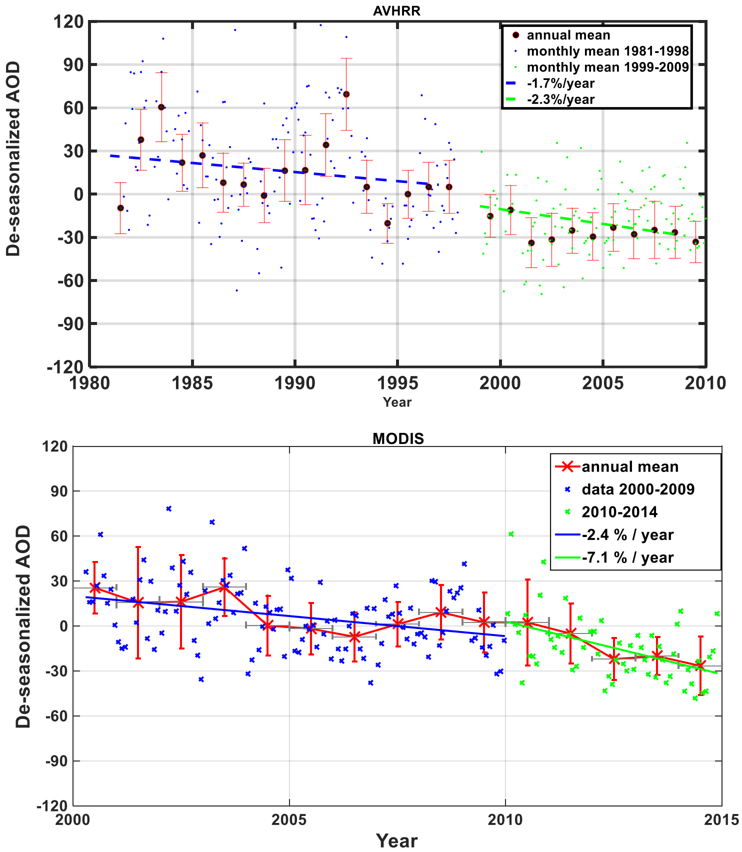


New Fig. 11. Inter-decadal variability of the annual visibility at NOA (urban) and HER (background) stations. Bold black lines represent the common period of observations (1956-2009) at the two stations along with linear trends and slopes. Solid blue line illustrates historical European emissions of SO_2 as reported in Vestreng et al., 2007 and blue dashed line illustrates historical European emissions of NO_2 as reported in Vestreng et al., 2009.

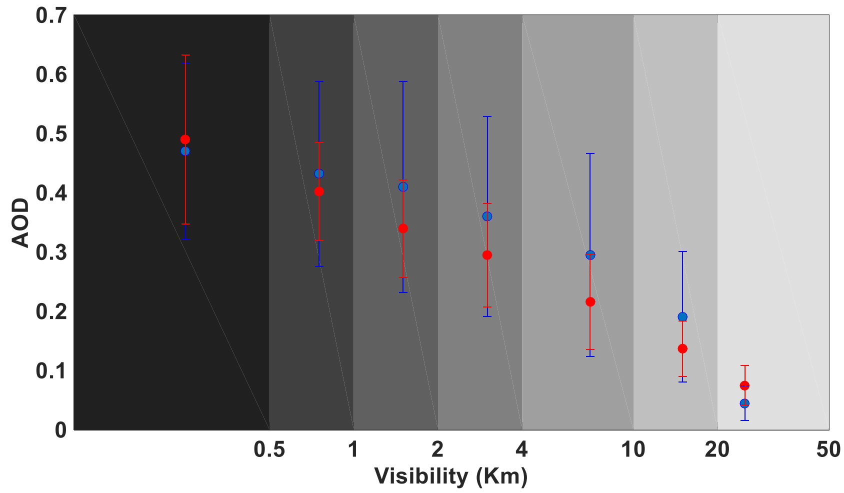
Historical emissions and trends of Organic Carbon as included in the study of Bond et al., (2007, Fig. 6) were also discussed. The segregation of emissions according to air mass origin was also discussed in the text. Information for segregation was based on the same studies (Vestreng et al., 2007, 2008) but also the study of Mylona (1996) and van Aardenne et al., 2001. Emphasis is given on air masses from N, NE directions (North, Eastern Europe), since on an annual basis, air masses from the N and NE sectors dominate in the area of interest (Figs 2, 3 of the manuscript).

3. Section 2.5 has been changed including the reviewer's suggestion to include the AVHRR analysis in addition to the one of MODIS. In this section, we describe the data set used with the respective references and the specific analysis and data set details for the Athens case.

Then section 3.6 has been changed accordingly including the results of the analysis of AVHRR data. In addition, we have included a new figure (Fig. 12a) showing the AOD changes in Athens area from 1981 to 2009 based on AVHRR and we have superimposed the AVHRR related results to Figure 13, describing now the AOD - visibility index relationship from two different data sets.



New Fig. 12. a) Variability of deseasonalized monthly AVHRR-based AOD_{630nm} from 1981 to 2009 (black), along with linear trends for the periods 1981-1997 (blue), 1998-2009 (green). Vertical bars describe the standard deviation of the annual value based on the monthly ones .b) Variability of MODIS-based deseasonalized monthly AOD_{550nm} from 2000 to 2014 (red), along with linear trends for the periods 2000-2009 (blue), 2010-2014 (green). Vertical bars describe the standard deviation of the annual value based on the monthly ones and grey horizontal bars the respective year.



New Fig. 13. MODIS at 550nm (blue) (2000-2014) and AVHRR at 630nm (red) (1991-2009), AOD June-August mean values and standard deviations for each visibility index. Shaded areas represent visibility ranges (km) for each visibility class (Table 2). AOD averages have been represented here in the average distance from each class

Minor comments of referee #1

1. All suggested first corrections by the referee were applied in the text. Additional syntax errors were also found and corrected.

2. Figure 2a is now referenced in the text before Fig. 2b.

C2. Changes in the manuscript to account for the comments of referee #2

General

The length of section 4 was reduced in the manuscript. The discussion focused on the main findings of the study and duplication of information or extended analyses were avoided.

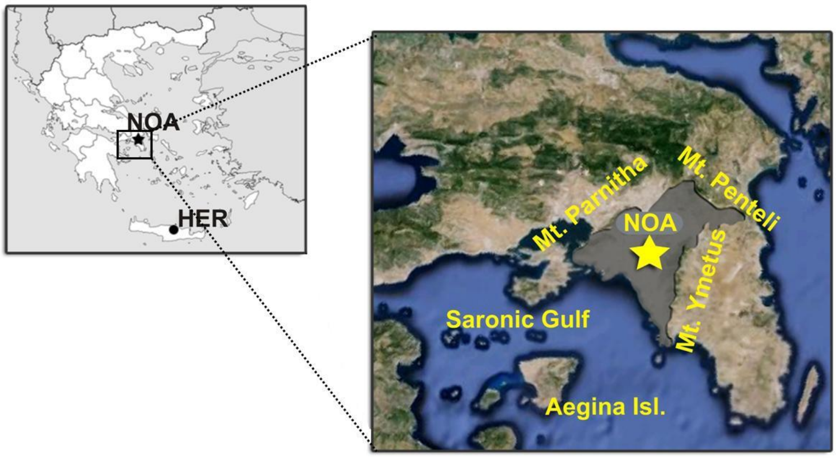
Minor comments

Information for Finokalia station is added in the manuscript: The Finokalia station (35.240° N, 25.600° E) is located on the Northern coast of Crete, Greece, at a distance of approximately 320 Km to the south of Athens. There is no significant human activity within an area of approximately 15km around the station, mainly characterized by a scarce vegetation. The closest large urban area is the city of Heraklion (HER), (see map. of Fig. 1) with 150 000 inhabitants, and located 50 km West from Finokalia. Aerosols at the site are mainly transported from the Southern-Eastern Europe and Northern Africa, and to a lesser extend from central and western Europe (Kouvarakis et al., 2000; Mihalopoulos et al.,1997).

Lines 248-254: The trends of visibility were now expressed as km yr^{-1} in the manuscript .

Some of the excel -based figures were reproduced using a different graphical tool. When not possible, the resolution of excel- based figures was increased.

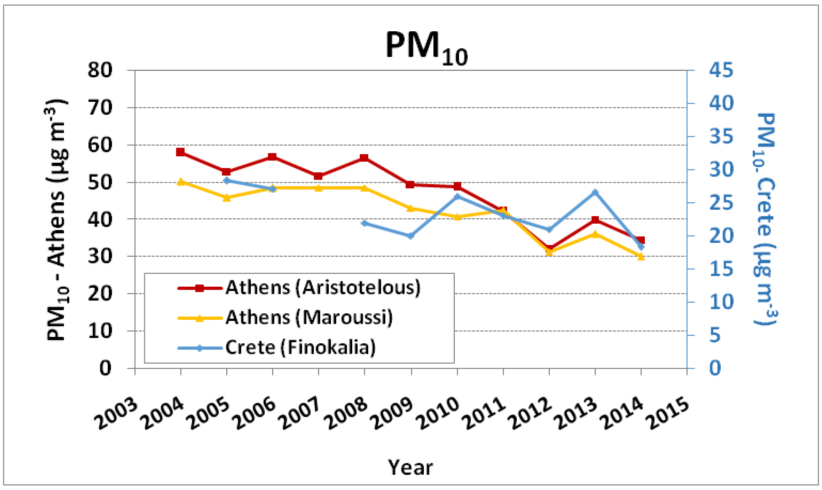
Fig. 1 was reproduced with a proper zooming.



New Fig.1. Map of the study area in Greece, including the Athens urban station (NOA) and a reference, non-urban station (HER) at Heraklion airport, Crete. The gray surface represents the boundary of the Greater Athens Area (GAA).

Fig. 7: The word 'Height' was dropped from the title of y-axis .

Fig. 15: The figure was recreated including annual PM10 values for the two stations of Maroussi and Aristotelous in Athens.



New Fig. 15. Variation of the annual PM10 concentrations at the reference station of Finokalia (Crete) over the period 2005-2014 and at the stations of Maroussi and Aristotelous in Athens (2004-2014).

Technical corrections

Technical corrections suggested by referee #2 were applied in the manuscript (Lines 33, 38, 55, 231, 260, 272, 261, 266, 272, 290, 345, 408, 423, 475)

Additional syntax errors were also found and corrected.

D. Marked-up manuscript version

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

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Abstract. This study explores the inter-decadal variability and trends of surface horizontal visibility at the urban area of Athens from 1931 to 2013, using the historical archives of the National Observatory of Athens (NOA). A prominent deterioration of visibility in the city was detected, with the long~~-~~term linear trend amounting to -2.8 km decade^{-1} ($p < 0.001$), over the entire stud~~yied~~ period. This was not accompanied with any significant trend in relative humidity (RH) or precipitation over the same period. A slight recovery of visibility levels seems to be established in the recent decade (2004-2013). ~~-~~It was found that very good visibility (> 20 km) occurred at a frequency of 34~~-~~% before the 1950s, while this percentage drops to just 2 % during the ~~recent~~ decade 2004-2013. The rapid impairment of the visual air quality in Athens around the 1950s~~1950,~~ points out to the increased levels of air pollution on a local and/or regional scale~~emission sources~~, related to high urbanization rates and/or increased anthropogenic emissions on a global scale at that period.~~. and/or higher rates of anthropogenic emissions increase on a global scale at that period. A marked seasonal cycle was detected in visibility before the1950s, which attenuates afterwards.~~ Visibility was found to be negatively/positively correlated with relative humidity (RH)/wind speed, the correlation being statistically valid at certain periods. Wind regime and mainly

wind direction and corresponding air masses origin were~~was~~ found to highly control visibility levels in Athens. The comparison of~~between~~ visibility variation in Athens and at a reference, non urban site on Crete island,~~,~~ revealed similar negative trends over the common period of observations. This suggests~~observations, suggesting~~ that apart ~~from the contribution of~~ local sources, visibility in Athens is highly determined by aerosol load~~s~~ of regional origin. Satellite derived aerosol optical depth (AOD) retrievals over Athens ~~since 2000,~~ and surface measurements of PM_{10} confirmed the relation of visibility with aerosol load~~s~~.

1 Introduction

Visibility is defined as the greatest distance at which a black object of suitable dimensions (located on the ground) can be seen and recognized, when observed against the horizon sky during daylight, (WMO~~,~~ 1992). Visibility represents one of the dominant features of the climate and landscape of an area. Although it is highly affected by atmospheric circulation and the prevailing meteorological conditions, under clear sky conditions it is mainly determined by~~from~~ the loading of atmospheric aerosols (Davis, 1991; Lee, 1994; van Beelen and van Delden, 2012; Doyle and Dorling, 2002; Singh and Dey, 2012), therefore, visibility can be considered as a strong indicator of air quality over~~at~~ an area. Horizontal visibility has ~~been~~ also been introduced in formulas for the estimation of atmospheric turbidity parameters (e.g. in the Ångström atmospheric turbidity coefficients, Eltbaakh et al., 2012).

Aerosols in the atmosphere contribute to light extinction by scattering and absorbing, thus reducing~~they reduce~~ visibility (Appel et al., 1985; Chan et al., 1999; Elias et al., 2009; Singh and Dey, 2012). The impact of particulate matter on visibility depends on its physical (e.g. particle size distribution) and chemical properties (Dayan and Levy, 2005). In particular, visibility is inversely related to light extinction coefficient, which is determined by~~from~~ scattering and absorption of light by gases and particles, the latter (e.g. sulphate and carbon containing particles) being the main contributor (Malm, 1999; Hand et al., 2002; Baumer et al., 2008; Deng et al., 2011; Wang et al., 2012). Sulphate and carbon containing particles play~~have~~ a major role in light extinction,~~absorption,~~ while the role of relative humidity (RH) on visibility is also important (Larson and Cass, 1989; Malm, 1999), as when RH reaches saturation values, visibility deteriorates due to fog formation and the hygroscopic growth of SO_4^{2-}, NH_4^+ and NO_3^- particles (Tang, 1996; Sing and Dey, 2012).~~-~~ At ~~the~~ local and~~to~~

regional level, wind speed and direction are also very important factors, as they determine the transport and origin of air pollution.

Although the use of visibility as a viable atmospheric variable has been disputed by many researchers due to the numerous biases related to observational procedures (Davis, 1991), visibility statistics have been increasingly used as a surrogate for aerosol load~~s~~ (Zhao et al., 2011), especially since visibility records span quite long-term periods. Today, there is a large number of studies that use visibility observations to investigate the spatial and temporal variation of the optical properties of the atmosphere, mainly in relation to pollutant~~s~~ emissions and aerosol load. These studies~~loads. Studies~~ refer to global, regional and local scales. On a global scale, a decrease of clear sky visibility over land from 1973 to 2007 is reported by Wang et al. (2009). This is interpreted in terms of aerosol concentrations and its impact on incident solar irradiance. A significant decrease of visibility is observed over Asia, South America, Australia and Africa (1973-2007), while over Europe visibility increased after the 1980s, as a result of air pollution mitigation measures. Vautard et al. (2009) found a significant decrease in the frequency of low visibility days in Europe after the 1980s, which is spatially and temporally correlated with SO_2 emissions. Stjern et al. (2011) reported that emission reductions from 1983 to 2008 in the heavily industrialized area of central Europe (the formerly called Black Triangle, BT, ~~(~~named from the triangle of the meeting borders of Germany, Poland, and the Czech Republic) caused an increase ~~of 15 km~~ in the horizontal visibility by 15 km, in contrast to the clean area where visibility increased by only 2.5 km. Doyle and Dorling (2002) observed significant improvement of visibility after the early 1970s at many sites in UK, attributed to anti-pollution measures,~~changes in the use of fuels,~~ while van~~Van~~ Beelen and van Delden (2012) found that the proportion of days with high visibility (>~~-~~19 km) almost doubled since the early 1980s~~,~~ in the Netherlands. These findings for Europe are in line with the so called dimming/brightening periods, referring to observed decreasing/increasing trends of surface solar radiation (SSR), associated with relevant changes in anthropogenic emissions (e.g. Streets et al., 2006; Wild, 2009; Cermak et al., 2010; Folini and Wild, 2011; Nabat et al., 2014).

In contrast to European areas, a tendency towards lower visibility is observed in developing countries (e.g. China, South Korea, South Taiwan, India), where it is still difficult to control air pollution (Ghim et al., 2005; Che et al., 2007; Wan et al., 2011; Singh and Dey, 2012; Wu et al., 2012). Along this line, Wu et al. (2012) found strong correlation between AOD and visibility in China over the period 2000-2009, and an overall decreasing trend in visibility (under sunny conditions) during the last 50 years. Singh and Dey (2012) correlated visibility in Delhi with aerosol composition and reported a rapid decrease of visibility during 1980-2000, and stabilization afterwards.

Urban environments are of particular interest, as air pollution from local sources is superimposed on ~~other~~ regional ones,~~factors,~~ strongly impacting visibility (Davis, 1991; Eidels-Dubovoi, 2002; Tsai et al., 2003, 2007; Dayan and Levy, 2005; Chang et al., 2009; Kim, 2015).

The present study explores the historical observations of visibility in Athens, which is the oldest ~~—~~time series of visibility in Greece and, to our knowledge, one of the oldest, uninterrupted time series of visibility in the eastern Mediterranean. The records are retrieved from the historical climatic archives of the National Observatory of Athens (NOA) and span a period of more than 80 years (1931-2013). In the past, Carapiperis and Karapiperis (1952) reported on the correlation between the visibility and the blue colour of the Attika sky, while Kanellopoulou (1979) analysed visibility in Athens for the period 1931-1977 and reported a pronounced decrease after the 1950s. Since then, there has been no other study to address~~addressing~~ changes in visibility, as well as the factors ~~-~~behind these changes~~,~~ during the last 40 years, when significant changes occurred in Athens in terms of urban expansion, traffic load, 2004 Olympic Games construction~~s~~ and the economic recession (starting in 2008). The inter-decadal variability and long-term trends of visibility in Athens are presented in the study. The role of meteorology and aerosol load~~s~~ (of local and regional origin)~~sources)~~ on the variability and trends of visibility are investigated and discussed, while the relationship between visibility and aerosol load~~ings~~ is investigated, through the analysis of satellite AOD retrievals over Athens~~, since 2000,~~ but also surface measurements of PM_{10} in Athens and Finokalia station (Crete) over shorter periods.

2 Study area and data

2.1 Study area

Athens, the capital of Greece, is~~concentrates~~ the main centre~~largest part~~ of~~ the~~ commercial, financial, societal and cultural activities of the country. The Greater Athens Area (GAA) (Fig. 1) extends beyond the administrative municipal city limits and covers a surface of 433 km^2. The population of GAA is approximately 3.7 million (almost twice the population of 1961) and accounts for more than one third of the Greek population. The growth of the population was coupled with a significant increase in the number of vehicles. Specifically, the number of private cars rose from 2 % of inhabitants in 1964 to 44 % in 2008. The population growth and the increased number of automobiles have~~has~~ caused traffic problems, increased anthropogenic emissions and degradation of air quality in the city. The complex topography, consisting of relatively high mountains around GAA (Fig. 1), induces poor ventilation of the city. Sea/land breezes appear along the ~~axis~~ NE - SW axis and play~~have~~ a dominant~~major~~ role in the accumulation of air pollutants (Kalabokas et al., 1999 a,~~-~~b).

In order to compare our findings for Athens with a reference, remote site, the visibility records from the Heraklion airport (HER) in Crete Island~~,~~ were used (Fig. 1). Heraklion is located about 330 km south of Athens, while its airport is 5 km east of the city with no significant (or systematic) influence by~~from~~ the urban web.

2.2 Climatic features of Athens

Athens has a temperate climate~~,~~ with warm and dry summers and ~~more~~ wet and mild winters, typical for eastern Mediterranean. Table 1 presents monthly and annual normal values along with standard deviations of the daily mean, maximum and minimum air temperature, precipitation amount and precipitation frequency (PF) (defined as the number of days with total precipitation > 1 mm, following WMO), relative humidity and wind speed in Athens, based on the WMO reference period, 1971-2000. July and August are the warmest and driest months of the year. The ~~Actually, the~~ periods from May to September and from October to March represent the dry and wet periods of the year respectively. Precipitation is sparse in summer (June- August), with the total amount averaging 20 mm and precipitation frequency averaging 3 days. Athens receives on average approximately 400 mm of rain per year, corresponding to 43 rainy days (Table 1).

~~-~~During summer, the area is dominated by anticyclonic circulation that enhances air temperature and intensifies urban heat island. Athens has been experiencing a significant warming since the mid 1970's, more pronounced in summer, which is the additive result of regional warming and gradual intensification of the urban heat island (Founda, 2011; Founda et al., 2015). Strong northeasterly winds in ~~late~~ summer, known from antiquity as 'Etesians', induce a relief on air temperature and air pollution levels in the city.

Figure 2a presents the main sectors related to air masses origin in Athens, based on 10-yr climatology of daily air trajectories, while Fig. 2b presents the seasonal variability of air masses origin ~~over Athens~~ according to the sectors defined in Fig. 2a. ~~2a, based on 10-yr climatology of daily air trajectories.~~ The S (south) sector is linked to transport of air masses from arid areas of N Africa, frequently associated with dust events that affect the eastern Mediterranean (Hamonou et al., 1999; Gkik~~k~~as et al., 2015), the N (north) sector accounts for Balkans and the main continental Europe, while the W (west) sector corresponds to SW Europe and the W Mediterranean Basin. Note that air masses transport from the W sector is~~are~~ significantly blocked by the high altitude mountain chain of Pindus (> 2500 m), which~~that~~ expands from North to South along the western Greek mainland. Air masses origin was identified by applying a 4-day back-trajectory analysis, calculated daily at 12:00 UT with the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (version 4.9) (Draxler et al., 2009).

On an annual basis, air masses from the N and NE sectors dominate, contributing by more than 60~~ ~~% and showing profound seasonal variability (maximum in summer). Similar conclusions are drawn from~~were obtained based on~~ surface wind ~~speed and direction~~ measurements, reported in Fig. 3. Winds from N-NE directions prevail in Athens at a frequency of nearly 38 % (Fig. 3). This sector is also associated with the occurrence of high wind speeds, as shown in the same figure. The second most frequent surface winds correspond to S-SW directions (27%). The frequency of occurrence of this sector has maximum~~maximizes~~ during the intermediate seasons (spring and autumn) and is associated with the occurrence of dust events from northern Africa and, in cases of light winds, with sea breezes from the Saronic Gulf (Fig. 1).

2.3 Overview of air pollution in Athens

A short introduction on the factors that diachronically control air pollution levels in Athens is presented here, to facilitate the interpretation of visibility variations in terms of pollutants concentrations.

Air pollution in Athens has been systematically measured since the early 1970s. Road transport, domestic combustion and industrial activity have been the main sources of air pollution in GAA~~,~~ throughout the years. Downward trends of sulfur dioxide, black smoke, carbon monoxide and nitrogen oxides have been reported from the mid 1980s to the late 1990s, attributed to several anti-~~—~~pollution measures adopted by the state (e.g. replacement of the old technology gasoline-powered private cars and the reduction of the sulfur content in diesel oil) (Kalabokas et al., 1999a). Negative trends of NO_2, NO_x and O_3 from the mid 1980s to 2009 are~~is~~ also reported in several urban stations (Mavroidis and Ilia, 2012).

Measurements of particulate matter (PM) had ~~been~~ only occasionally been conducted in Athens before the EU Directive (1999/30/EC) was launched, revealing increased concentrations of PM_{10} (Hoek et al., 1997).

Chaloulakou et al. (2003) reported on PM_{10} and $PM_{2.5}$ at a single road traffic sampling location from 1999-2000 and underlined the contribution of local emission sources, mostly traffic, to~~on~~ the high levels of PM concentration. Grivas et al. (2004) highlighted the significant vehicular contributions to~~in~~ PM_{10} concentrations in Athens during 2001-2004 and quantified the exceedances of the annual limit set by the EU Directive.

Studying the contribution of local sources versus regional and the role of long-range transport over megacities of the eastern Mediterranean, including GAA, Kanakidou et al. (2011) summarized that a significant number of PM exceedances registered in Athens is~~, are~~ associated with regional pollution sources or natural dust transport, clearly highlighting the importance of regional transport processes. Theodosi et al. (2011)~~,~~ compared simultaneous mass and chemical composition measurements of size segregated particulate matter (PM_1, $PM_{2.5}$ and PM_{10}) at two urban and a reference, non-urban background site, concluding that, during the warm season there is no significant (actually < 15 %) difference in PM_1 between the urban and reference sites, while on the other hand, local anthropogenic sources dominate during the cold season. Regarding the coarse fraction, a significant contribution from soil was found in urban locations throughout the year, contributing significantly (up to 33 %) to the local PM_{10} mass.

Regarding columnar aerosol load~~s~~ and using ground-based AOD measurements in Athens, Gerasopoulos et al. (2011) showed that the greatest contribution (40 %) to the annually averaged AOD, comes from regional sources (namely the Istanbul metropolitan area, the extended areas of biomass burning around the north coast of the Black Sea, power plants spread throughout the Balkans and the industrial area in the Po V~~v~~alley). Additional important contributors are dust from Africa (23 %), whereas the rest of Europe contributes another 22 %. Gkik~~k~~as et al. (2015) found good correlation between AOD_{550nm} and surface PM_{10} over the Mediterranean basin during desert dust episodes (2000-2013) and reported higher intensity but lower frequency of such episodes over the central and eastern Mediterranean. Additionally, Hatzianastassiou et al. (2009) found that local anthropogenic emissions in GAA contribute by 15-30 % to the total AOD, as derived from satellite-based AOD measurements.

Vrekoussis et al. (2013) reported on the improvement of air quality in Athens during the period 2008-2013, as a result of the economic recession and the subsequent reduction in vehicle~~cut down on vehicles~~ use and industrial activity. For the same period, Paraskevopoulou et al. (2014) showed that the massive turn of Athens' population to wood burning for residential heating purposes gave rise to smog episodes characterized by high PM spikes during night~~-~~time in winter. A longer-term (2008-2013) analysis of aerosol chemical composition and sources at a suburban site in Athens by Paraskevopoulou et al. (2015) revealed that the area of Athens is now generally dominated by aged, transported aerosols.

2.4 Visibility observations in Athens

The historical climatic record~~s~~ of the National Observatory of Athens (NOA) was~~were~~ used in this study. NOA is located~~established~~ on the Hill of Nymphs (latitude: 37.97 ^{0}N, longitude: 23.71 ^{0}E, altitude: 107 m, above sea level), at the historical center of the city, near Acropolis. The location of the observations on the top of a hill ensures unobstructed view towards all directions. Visibility observations have been conducted uninterruptedly at NOA at least 3 times per day, since the late 1920's. Daily observations of visibility at 14:00 LST (LST = UT + 2hrs), from 1931 to 2013 were used in the study. The time series is complete, with a very short gap of 6 days occurring in December 1944, owed to political convulsion in the country at that period.

Visibility data at other stations (e.g. Heraklion, Crete) were extracted from the network of the Hellenic National Meteorological Service (HNMS) and actually represent visibility observations at the airport station, initiated after ~~mid~~ the mid 1950s. Meteorological data for Athens over the period 1931-2013, was~~were~~ also acquired from the historical archives of NOA. Monthly, seasonal and annual mean values of visibility were derived from the daily observations at 14:00 LST.

An empirical scale of visibility classes, as recommended by the World Meteorological Organization (WMO), has been used for visibility observations at NOA (Table 2). Classes are defined based on the greatest distance at which a predefined object can be seen and recognized with the naked eye. The procedure requires that an operator scans the horizon for predetermined objects. In the case of Athens, some historical buildings in the city, but also certain objects of the surrounding landscape~~,~~ unaltered over the years, (e.g. objects on the mountains or islands of the Saronic Gulf, Fig. 1), were chosen to represent visibility classes and relevant distance ranges. The procedure ~~inevitably~~ introduces inevitably some kind of subjectivity and bias in the measurements, related to individual eyesight of different operators. It is assumed however, that the execution of visibility observations by different operators over the years could have possibly had a compensating effect and an overall reduction of biases. More details about the possible errors and validity of visibility observations have been thoroughly discussed by Davis~~,~~ (1991).

The use of the WMO scale introduces a further uncertainty on visibility observations, associated with the amplitude of visibility ranges corresponding to each visibility class. Information on the use of WMO scale and relative uncertainties, as well as the followed procedure~~ followed~~ for averaging daily visibility observations is provided in Supplementary materials.

2.5 Aerosol data used in the study

Long time series of atmospheric pollution measurements in Athens and the selected reference site would enable drawing ~~direct ~~relationships between visibility and aerosols and would provide evidence for~~on~~ the origin~~character~~ (regional or local) of atmospheric pollution in Athens and its impact on long-term visibility variations. Given that such time series are missing, we used shorter time series of aerosol measurements for a direct comparison between visibility and atmospheric pollution in Athens.

In an effort to explore the relationship between~~of~~ visibility and~~with~~ AOD over Athens, we used the Terra/Modis AOD at 550 nm, available since 2000. NASA's Terra satellite is sun synchronous and near polar-orbiting, with a circular orbit of 705 km above sea level. MODIS is capable of scanning 36 spectral bands across a ~~swath ~~2330 km wide swath. MODIS aerosol products were used in order to analyze the temporal and spatial variability of aerosols over the wide area of interest. In this study, we used daily level-2 collection 5.1 MODIS/Terra AOD at 550 nm. Daily overpass data for the specific area was~~were~~ extracted at a spatial resolution of 50 x 50 km^2. Previous studies have shown that such spat~~ec~~ial resolution product ensures sufficient daily measurements without losing out to the higher spatial resolution and hence provides a better opportunity of correctly viewing the atmospheric aerosol load (Ichoku et al., 2002). The overpass time is 09:35 ± 45 min UT.

In addition, in order to further examine long-term satellite based AOD series in the area, we used the longest satellite time series available from the Advanced Very High Resolution Radiometer (AVHRR). AOD retrievals PATMOS-x AVHRR level-2b channel 1 (630 nm) provide data over global oceans at high spatial resolution (0.1^o X 0.1^o), for one overpass per day. Data used were downloaded from NOAA Climate Data Record (CDR) version 2 of aerosol optical thickness (Zhao and Chan, 2014) and cover the period from August 1981 to December 2009. Version 2 dataset has enhanced cloud screening and retrieves AOD only over non-glint water surface, which has less uncertainties of surface reflectance. AVHRR instrument is not designated for retrieving AOD, thus its product embodies a large variety of uncertainties, including radiance calibration, systematic changes in single scattering albedo and ocean reflectance (Mishchenko et al., 2007). Current dataset radiances have been recalibrated using more accurate MODIS data (Chan et al., 2013). Smirnov et al. (2006) compared 38 days of ship borne measurements with a MICROTOPS-II, on a cruise in Atlantic Ocean to AVHRR AOD retrievals and found an average 0.05 overestimation of satellite data, with correlation coefficient equal to 0.95. We used daily overpass data at the region around Athens (latitude: 37.5º-38.2ºE, longitude: 23.2º-24.4ºN) which included 72 active (ocean) grid-points. The above region was selected based on data availability on each grid with the distance up to 70 km from the visibility observing site.

Surface PM_{10} measurements in Athens were also used to verify the relationship between visibility and particulate pollution from surface measurements. It is well known that desert dust plumes are often transported in altitude over the Mediterranean (e.g. Hamonou et al., 1999: Gkikkas et al., 2015) and a portion of surface PM exceedances in Athens is associated with natural dust transport (Kanakidou et al., (2011). The analysis was based on a short data-set of PM_{10} measurements at two stations in Athens (Aristotelous and Maroussi), covering the period 2008-2012. Aristotelous is an urban street station in the center of the city and Maroussi is a suburban station, at a distance of about 15 km to the North of NOA.

Finally, a data-set of PM_{10} measurements at a reference station in Crete (Finokalia station), covering the period 2005-2014 was used, for the detection of any trends, representative of regional atmospheric pollution trends. The Finokalia station (35.240° N, 25.600° E) is located on the northern coast of Crete (Greece), at a distance of approximately 320 km to the south of Athens. There is no significant human activity within an area of nearly 15 km around the station, mainly characterized by scarce vegetation. The closest large urban area is the city of Heraklion (HER), (see map. of Fig. 1) with 150 000 inhabitants, and located 50 km West from Finokalia. Aerosols at the site are mainly transported from the southern-eastern Europe and northern Africa, and to a lesser extent from central and western Europe (Kouvarakis et al., 2000). Finokalia station is located at a distance of less than 50 km East of Heraklion airport.

3 Results

3.1 Inter-decadal variation and trends of visibility and trends

Figure 4 displays the long-term developmentevolution and variability of the annual visibility in Athens from 1931 to 2013. The population growth in the city of Athens over the same period is also shown, while the figure also displays the long-term variability of the relative humidity in Athens (which is discussed below). It is obvious that the annual visibility in Athens has undergone a very strong and almost continuous decline over the past 80 years, in coincidence with the increase in population. The long-term linear trend over the entire study~~whole studied~~ period was found to be equal to -0.28 km yr^{-1} (or -2.8 km decade^{-1}, ~~(~~p < 0.001). However, this trend is not constant throughout the entire stud~~yied~~ period. The following three~~Three~~ sub-periods, corresponding to different trands, are visually discerned in Fig. 4 (also confirmed by~~with~~ sensitivity tests): (a) 1931-1948, (b) 1949-2003 and (c) 2004-2013. Visibility levels are remarkably higher in the first sub-period, varying around 25 km. A slight negative trend is observed during this period (-0.07~~ (-0.66~~ km yr^{-1}).~~decade^{-1}).~~ In the late 1940s, visibility experienced a striking and abrupt decrease at the time of first population~~ first~~ burst, which was then followed by a progressive deterioration, at least until the early 2000s. In this second sub-period (1949-2003) visibility decreases at a rate of -0.23 km yr^{-1} (or -2.3~~3~~ km decade^{-1}, ~~(~~p < 0.001). A tendency of stabilization or even recovery seems to be established during the more recent decade 2004-2013, when~~with~~ visibility exhibits~~showing~~ a slight increasing trend (+ 0.07 km yr^{-1}). A detailed discussion on the observed trends and their links to~~linkage with~~ air pollution is presented in section 3.5.

3.2 Frequency distribution of visibility ranges

The separation of the time series into three sub-periods was indicated by the fact that they represent periods of changing trends. In the following, the much longer middle sub-period (1949-2003) was further separated into two parts (1949-1975 and 1976-2003) as it corresponds to substantially different visibility conditions. Figure 5 illustrates the frequency of occurrence of different visibility ranges as described in Table 2 for different sub-periods.~~the three sub-periods. In the first sub-period, visibility values lie within the range of 10-20 km at a percentage of 36 % and of 20-50 km at a percentage of 34 %. Very high visibility (> 50 km) accounts for a considerable percentage (~9 %) and poor visibility (< 2 km) corresponds cumulatively to only 2 %. The frequency of visibilities lower to 1 km is very low (0.4 %), while visibility was found to be lower to 500 m only in 9 cases. Cumulatively, visibility exceeding 10 km corresponds to approximately 80 % of the cases during this period.~~

In the first sub-period (1931-1948), visibility values are almost equally distributed between the ranges of 10-20 km and 20-50 km, at frequencies of approximately 35%. Very high visibility (> 50 km) accounts for a considerable portion (~ 9 %) of this sub-period and poor visibility (< 2 km) corresponds cumulatively to only 2%. The frequency of visibility lower than 1 km is very low (0.4 %), while visibility lower than 500 m occurred only in 9 cases. Cumulatively, visibility exceeded 10 km at a frequency of approximately 80% during this period.

A progressive shift of frequency distribution towards lower visibility categoriesvalues is observed in the next sub-periods. In particular, the frequency of very good visibility (20-50 km) drops to 13% and 6% for the periodsduring the second sub-period, namely 1949-1975 and 1976-2003 respectively, while1949-2003. Specifically, the most frequent visibility range is s are 4-10 km (38 %) and 10-20 km (44%) during 1949-1975 and 4-10 km (41%) during 1976-2003.(34 %). The frequency of visibility > 50 km is almost negligible (~ 1% during 1949-1975)(0.6 %) and the frequency of poor visibility (< 2 km) amounts cumulatively to 5.6 % , with 0.9 % and ~ 1% for 1949-1975 and 1976-2003 respectively. Lower than 500 m corresponding to visibility < 1 km. Visibility lower to 500 m was observed only in 12 cases during 1949-1975 and in10 cases during 1976-2003. . Cumulatively, the percentage of days with visibility exceeding 10 km drops to 58 % and 29% for the periods 1949-1975 and 1976-2003 respectively. 45 % during this sub-period.

The frequency distribution changes dramatically during the most recent period (2004-2013). In particular, although visibility range of 4-10 km remains the most frequent (30%), as in the second sub-period 1976-2003, almost similar frequency (~28 %) is also observed in the range of 2-4 km.km, corresponding to a doubling of the percentage of this category. The frequency of poor visibility (< 2 km) rises to approximately 25 %, with a substantial percentage (5.6 %) accounting for visibility lower thanto 1 km and 0.46 % lower thanto 500 m. Overall, Cumulativelly, visibility did not exceed 4 km for half of the days of the year during 2004-2013. The percentage of days with visibility > 10 km is 18%, while frequency of very good visibility (> 20 km) amounts to just 2 %. No case of visibility > 50 km was observed in this last sub-period.

3.3 Seasonal variation of visibility

Since visibility is influenced by the prevailing meteorological conditions (Davis 1991; Sloane 1982), it is expected tothat it will also exhibit a seasonal variability, depending on the intra- annual variability of climatic conditions at the studyexamined area. Mean monthly values of visibility were calculated for theall three sub-periods 1931-1948, 1949-1975, 1976-2003 and 2004-2013.. Figure 6 (a-d) presents the mean monthly values of visibility in Athens over eachthe three sub-period,sub-periods, normalized with the value of the month with the highest visibility. In the same plot, the mean monthly values of the relative humidity (RH), coinciding visibility observations at 14:00 LST over each sub-the period 1931-2013, are also shown. It is noteworthy that RH at NOA does not exhibit any significant trend over the years (as already shown in Fig. 4) and its monthly distribution remains~~is~~ almost unaltered in all sub-periods.~~over the years.~~ As it results~~comes out~~ from Fig. 6 (a-d), visibility exhibits~~shows~~ a~~ distinct~~ seasonal cycle in all ~~three ~~sub-periods, with better visibility occurring in the warm and dry months~~season~~ of the year. Although seasonality is observed in all sub-periods, the pattern is more evident and robust in the first sub-period (Fig. 6a), with much higher visibility values (up to 40%) in the warm and dry ~~months compared to cold and wet~~ months. The pattern of visibility in this period is almost a mirror image of the pattern of RH and reflects the influence of RH on visibility and the anti-correlation between these two variables. The lowest values of RH correspond to July and August (mean value of RH ~35% at 14:00 LST) and this probably results in visibility~~to~~ improvement.~~ of visibility.~~ Moreover, strong northeasterly~~n~~ winds~~ (the so called ‘Etesians’)~~ that prevail in eastern Greece during these months enhance ventilation and induce drier conditions in the city, therefore improving visibility.

The ~~In the other two sub-periods, 1949-2003 and 2004-2013, higher visibility values are also observed during the warm and drier months (Fig. 6), however, the ~~distinct seasonal cycle ~~observed ~~in visibility of the first sub-period ~~has ~~changed. ~~During the second sub-period ~~in the following sub-periods (Fig. 6, b-d). Although the warm and drier months always correspond to higher visibility levels,~~particular,~~ seasonality is noticeably attenuated and visibility differences between the warm and cold period are much lower. ~~is of the order of 10%.~~ This possibly implies a weakening of the influence of meteorological conditions, as a result of (or in combination with) the stronger effect of air pollution on the visual air quality of the city.

The minimum of visibility is constantly observed in March during all sub-periods. Indeed, March is a month of ~~falls in the ~~transitional season~~ of the year~~ and thus bears higher values of RH compared to summer months (mean value of RH at 14.00 LST > 50 % and mean daily value 67 % in March). Additionally, March is a month of~~falls in~~ the growing season, with enhanced pollen and biogenic aerosol emissions which is a known factor for visibility impairment (e.g. Kim, 2007). Increased frequency of dust outbreaks from northern Africa in spring, influence extensively the area of eastern Mediterranean (Hamonou et al., 1999; Gerasopoulos et al., 2005, 2011; Gkik~~k~~as et al., 2015) and thus constitute a major factor for visibility impairment during spring months. Léon et al (1999) reported that ~ 40 % of the days with high aerosol optical depth at 865 nm (AOD_{865nm} > ~~–~~0.18) over Thessaloniki (Greece) were associated with African dust transport events, all observed in the period March – July, while Dayan and Levy (2005) found higher PM_{10} values and lower visibility levels during spring in Tel Aviv, associated with the frequent passage of cyclones that cause natural dust outbreaks.

3.4 Visibility and meteorological conditions

The impact of meteorological conditions on visibility has been investigated by different researchers using different approaches~~-~~, as for instance~~-~~ the classification of synoptic circulation patterns (Sloane, 1982; Davis, 1991; Dayan and Levy, 2005), the application of correction factors on extinction coefficient to account for RH effect (Che et al., 2007), the estimation of correlation coefficients between visibility and meteorological variables (Deng et al., 2011), or simply the comparison of diurnal~~-~~/seasonal cycles and temporal trends of visibility with the relevant cycles and trends of meteorological variables (v~~V~~an Beelen and van Delden, 2012). Sloane (1982) reported that periods with exceptionally maxima or minima of visual air quality were related (apart from sulphate emissions) to~~with~~ favourable synoptic circulation patterns. Studying visibility in Tel Aviv (Israel), Dayan and Levy (2005) reported a strong dependence of visibility levels on~~from~~ meteorological conditions, synoptic weather patterns and air mass origin, with the highest mean values occurring in summer, related to the persistent nature of the summer synoptic weather patterns in the eastern Mediterranean. Deng et al. (2011) found that RH and wind speed were significantly correlated with visibility at an urban area of China, while Ghim et al. (2006) showed a considerable decrease in visibility in South Korea, despite the observed simultaneous decrease of RH~~the relative humidity~~ levels. The relationship and possible impact of different meteorological parameters such as precipitation, RH, wind speed and wind direction on visibility in Athens is discussed below.

3.4.1 Visibility and precipitation

Precipitation is associated with scavenging of atmospheric particles (e.g. Remoudaki et al., 1991a; 1991b), possibly resulting in~~to~~ improvement of visibility. The precipitation frequency in particular, was found to control seasonal variability of the total atmospheric deposition of lead in western Mediterranean (Remoudaki et al., 1991b). Rainy days, on the other hand, are associated with increased relative humidity, resulting in reduction of visibility. A plot illustrating the long-term variability of the annual precipitation amount and precipitation frequency (PF) at NOA from 1931-2013 was created, for the detection of any significant temporal trends ~~which might have an effect on visibility trends~~ (Fig. 7). As it results from the figure,~~According to Fig. 7,~~ no long-term trend is observed in the annual precipitation~~ amount~~ at NOA from 1931-2013, which could have had an effect on long-term trends of visibility. Precipitation frequency, on the other hand, exhibits an overall negative trend over the same period (-1.1 days decade^{-1}) which is~~,~~ not constant throughout the time series. Specifically,~~, though. Actually,~~ PF decreases from the late 1960s to the late 1980s, while it presents an increasing tendency after 1990

(+1.3 days decade^{-1}). The correlation coefficient between annual visibility and PF was found to be positive only during the period from the early 1970s to the late 1980s (+ 0.45, $p < 0.05$). A negative correlation coefficient~~s~~ was found in the post 1990 period (-0.21), not statistically significant.

Subsets of data were also produced for the creation of additional visibility time series, accounting for precipitation influence. Figure 8 presents visibility variability during the wet (October-March) and dry (May-September) period of the year, along with the annual values. Lower values during the rainy and cold period of the year are most probably associated with higher values of relative humidity, resulting in the~~to~~ reduction of visibility. Despite the differences between the time series in Fig. 8, the overall tendency is similar, thus not affecting the validity of our conclusions regarding the~~as regards~~ long-term visibility impairment in Athens. Additional plots, created from subsets of ‘rain’ and ‘no rain’ days are provided in Supplementary materials (Fig. S4).

3.4.2 Correlation between visibility and other meteorological parameters (RH, wind)

Figure 9 presents the running correlation coefficient (15-yrs window) between visibility and relative humidity at NOA, over the period 1931-2013. As expected, the correlation coefficient between visibility and RH is negative, indicating the anti-correlation between these two variables. High RH enhances water uptake by airborne particles, leading to higher light scattering and thus, visibility impairment. Actually, when RH exceeds a threshold level (e.g. > 70%), some inorganic salts, such as ammonium, sulfate and nitrate, undergo sudden phase transitions from solid particles to solution droplets and become ~~disproportionately~~ responsible for visibility impairment, as compared to~~with~~ other particles that do not uptake water~~ molecules~~ (Malm, 1999).

Following~~As it comes out from~~ Fig. 9, the negative correlation between RH and visibility is statistically significant ($p < 0.01$) almost over the entire study~~ied~~ period. However, a progressive weakening of the correlation coefficient with time is observed, indicating a less strong correlation between the two variables over the years. Stronger anti-correlation is found until the early 1970s, followed by lower (still significant) values until the~~till~~ late 1970s. The progressive weakening of the correlation between RH and visibility in Athens, possibly suggests a progressive weakening or mask of RH~~the~~ influence~~ of RH~~ on visibility, compared to the effect of other factors such as atmospheric pollution (although the influence of RH is enhanced by~~in~~ the presence of certain hygroscopic particles). On the contrary, the impact of surface wind speed on visibility seems to be stronger during the late part of the time series~~recent decades~~ (Fig. 9). Higher wind speeds in this case (positive correlation) are related to the dispersion of air pollutants and the more efficient city ventilation. In other~~s~~ cases, wind speed is also used as a proxy for long-range transport, but then a negative correlation would be expected. Lower values of the coefficient in the early part of the time series~~first decades~~ possibly demonstrate that the lack of pollutants at that period detracts from~~diminishes~~ the importance of ventilation. The correlation coefficient increases progressively ~~increases~~ over the years. The rate of increase is higher after the mid 1980s, when correlation becomes statistically significant ($p < 0.01$). Similar values ~~(~ 0.29)~~ of correlation coefficient (~ 0.29) between light extinction coefficient and wind speed are reported by Deng et al. (2011) in China.

Apart from wind speed, visibility was also found to be sensitive to wind direction. A distinct variability of visibility with wind direction is observed in Fig. 10, for all sub-periods. Lower values of visibility are related to southerly winds, as they bring either~~ bring~~ dust from Sahara or warmer and more humid air masses from the sea (see also Figs 1, 2b). Southeasterly winds are, in general, weak winds (see Fig. 3), while southwesterly winds are associated with sea breezes from the Saronic Gulf (Fig. 1). In general, sea breezes and calm wind conditions~~s~~ favor the accumulation of pollutants and~~,~~ the formation of secondary aerosols and photochemical smog in Athens (Colbeck et al., 2002), thus reducing visibility. A number of S/SW events are also associated with strong wind speeds occurring during Sahara dust outbreaks, which enrich Athens atmosphere with dust particles that decrease visibility (Figs 2, 3). As it results~~comes out~~ from Fig. 10, the highest visibility occurs under northwesterly winds and this is robust for all sub-periods.~~over the entire studied period.~~ An explanation for this, is that air masses originated from northwesterly directions are much drier as they have lost water vapor after passing over the high mountainous basin of the Greek mainland (e.g. Pindos mountain), while air pollution is also blocked within the boundary layer by the mountain chain.

3.5 Air pollution and urbanization relations to visibility

In this section, we attempt to interpret the observed inter-decadal variability and trends of visibility in Athens, in terms of air pollution. As already shown in Fig. 4, the pre-1950 period is characterized by considerably higher~~much better~~ visibility levels in Athens. From then on, visibility experienced a rapid decrease, followed by a smoother but continuous decreasing~~negative~~ trend until the early 2000s. The period after 1950 signifies the post World War II epoch but also coincides with the end of a civil war in Greece (1946-1949), which was followed by an important urbanization wave in Athens (Maloutas, 2003). This is in line with the rapid growth of Athens' population, as illustrated in Fig. 4. The greatest rate of population increase is observed between 1950 and 1960, when population in Athens almost doubled. The population growth was associated with a significant increase of constructions in the city. But apartApart from the intense urbanization in Athens, this period is also characterized by the most prominent increase of anthropogenic emissions on a global and European scale (e.g. Mylona, 1996; van Aardenee et al., 2001, Vestreng et al., 2007, 2009).2001), which is discussed below.

Are the changes in visibility in Athens due to local factors or can they be considered representative of a more extensive area? To answer this question, the Athens visibility record was compared with visibility at a reference, non urban station. From the available stations in Greece disposing long-term visibility observations, we chose the station at Heraklion airport (HER) in Crete Island (Fig. 1). Actually, both sites, NOA and HER, are exposed, most of the year, to air masses of similar origin (from north and northeasterly directions) travelling over the Aegean Sea, in contrast to other sites of the country that are strongly affected by the mountainous volumes of the Greek mainland. Visibility observations at HER are available since the mid 1950s. Figure 11 presents the long-term variation of the annual averages of visibility at HER along with the annual visibility at NOA.Although in the second sub-period, 1949-2003, visibility was found to be remarkably lower compared to the first one, a slight recovery of visibility was observed during the recent decade, 2004-2013 (Fig. 4). This improvement could be related to a number of reasons. The years after 2004 Linear trends of the two time series for their common period (1956-2009) are also shown in the figure. The time series were found significantly correlated (correlation coefficient > 0.88, $p < 0.05$).

According to Fig. 11, visibility levels at urban NOA are constantly lower by a few km (~ 7 km) compared to the background station, HER. It is remarkable that, during the first two decades of parallel observations, both curves show significant covariance, easily realized from the peaks in 1959, 1966 and 1970 and the minima in 1963 and 1973, suggesting the impact of large scale phenomena (for instance, volcanic eruptions in 1963) on the modulation of visibility levels. A prominent feature in Fig. 11 is that, the background visibility at the reference site has also been on a downward route since the mid 1950s, in accordance with the observed decreasing trend of visibility in Athens. As already stated, the beginning of the 1950s corresponds to a period with significant increase of emissions in Europe. European emissions of SO_2 in particular, increased almost at a constant rate during the first half of the 20th century, while they experienced a quite abrupt increase in the 1950s (Mylona, 1996; van Aardenne et al., 2001; Vestreng et al., 2007). Figure 11 includes the historical development of SO_2 and NO_2 emissions in Europe since 1930, as reported by Vestreng et al. (2007) and Vestreng et al. (2009) respectively. A slow and constant increase of SO_2 emissions is observed until the 1950s (although the emissions decreased during the World War II), related to the increased energy demand and use of solid fuels. A sharp increase in sulphur emissions takes place afterwards, as a result of ongoing energy demand and availability of liquid fuels (Vestreng et al., 2007), and in the late 1970s sulphur emissions were higher by a factor of neary 2.5, compared to the 1950's levels, exceeding 50 Tg SO_2. After as short stabilization in the 1980s, a sudden reduction in sulphur emissions takes place (most prominent after 1990) which in the 2000s almost correspond to the levels of 1930. Historical development of NO_x emissions in Europe exhibits a similar pattern (Fig. 11), with pronounced increase in emissions from 1950 to 1980, a tendency of stabilization between 1980 and 1990 and a decline thereafter. The plot of NO_x emissions in Fig. 11 refers to all sectors, as included in Vestreng et al. (2009).

Segregation of emissions trends by mass origin would further enlighten their possible effect on visibility variation in Athens. As stated in section 2.2, air masses from the N- NE sectors dominate in Athens, contributing by more than 60% on an annual basis. Following segregation of European SO_2 emissions by country as reported by Mylona (1996) it comes out that emissions by countries of N-NE sector (as defined in Fig. 2a) have the largest contribution in total European emissions. Sulphur dioxide emissions increased by a factor of approximately 2.5 between 1950 and 1980 in these regions, which is analogous to the increase of total European emissions over the same period. According to Mylona (1996), the contribution of emissions from the former USSR (but also Turkey) is very important after 1940. The EMEP part of USSR in particular, contributed to almost one quarter of the total in the 1970s. Sulphur emissions declined after the 1990s in both eastern and western Europe, but with higher rates (by a factor of 1.5) in eastern, as a result of the economic recession after 1990 in these countries (Vestreng et al., 2007; Stjern et al. (2011).

As regards other types of emissions such as organic carbon (OC) or black carbon (BC), historical data reported by Bond et al. (2007) show increase of the order of 50% on a global scale between 1930 and 2000. However, segregation by region indicates that European emissions of OC and BC revealed a slight increase between 1950 and 1970 and decrease thereafter. Decreasing trends are also observed in the former USSR after 1970 (Bond et al., 2007).

A very interesting finding in Fig. 11 is the similar slopes in the negative linear trends of the annual visibility at the background and urban stations over their common period of observations (-2.2 km decade^{-1} and -2.4 km decade^{-1}, respectively). This feature implies that the inter-decadal variability of visibility in the eastern Mediterranean is significantly modulated by large scale processes that control visibility, such as long-range pollution transport. Many studies have identified the eastern Mediterranean as a crossroad of aerosols of different origins, sizes and chemical composition (Lelieveld et al., 2002; Hatzianastassiou et al., 2009; Kanakidou et al., 2011; Gerasopoulos et al., 2011), which inevitably affect optical properties of the atmosphere.

After the early 1990s, the two time series diverge. Background visibility at HER partly recovers, while visibility at NOA keeps declining at the same pace until 2003 (Fig. 11). Recovering of visibility is also found at other Greek areas around the 1990's (Lianou et al., unpublished data) which is in line with visibility improvement in other European areas, related to emissions reduction (Wang et al., 2009; Vautard et al., 2009). This last feature suggests that, during this period, local emissions might have a dominant role in the determination of visibility in Athens.

A slight recovery of visibility is observed during the decade 2004-2013 (Figs. 4, 11). This improvement could be attributed to a number of reasons. The years after 2004, correspond to the post Olympic Games period in Athens. A number of important transport projects were completed prior to the Olympic Games in Athens in 2004. Such projects are for instance the construction of the Attika Ring Road (one of the largest in Europe), the construction of Tramway and the extension of Athens Metro. These projects have contributed to the reduction in~~of~~ the number of vehicles in the city, resulting to less traffic problems and lower air pollution levels. Another possible contributing factor concerns the ~~possible~~ impact of the Greek economic recession (2008-2013) on air quality in Greece, and Athens in particular. Recent studies provide some evidence on~~for~~ this. For instance, Vrekoussis et al. (2013) found strong correlation between different economic metrics and air pollutants after 2007, suggesting that the economic recession has resulted in proportionally reduced levels of air pollutants in the two biggest cities in Greece. This is further supported by other recent research studies that report a significant reduction in energy consumption after 2008, related to the rapid economic degradation (Santamouris et al., 2013).

~~But how far are these changes in visibility in Athens due to local factors or can be considered representative of a more extensive area? To answer this question and also evaluate our findings as regards the urban influence, the Athens visibility record is compared with visibility at a reference, non urban station. From the available stations in Greece disposing long-term visibility observations, we chose the station at Heraklion airport (HER) in Crete Island. Actually, both sites, NOA and HER, are most of the year exposed to air masses of similar origin (from northeasterly directions), travelling over the Aegean Sea, in contrast to other sites of the country that are strongly affected by the mountainous volumes of the Greek mainland. Visibility observations at HER are available since the mid 1950s. Figure 11 presents the long-term variation of the annual visibility at HER along with annual~~

visibility at NOA. Linear trends of the two time series for their common period (1956-2009) are also shown in the figure. The time series were found significantly correlated (correlation coefficient > 0.88, $p < 0.05$).

As it comes out from Fig. 11, visibility levels at urban NOA are constantly lower by a few km (~ 7 km) compared to the background station, HER. It is remarkable that during the first two decades of parallel observations, both curves show significant covariance, easily realized from the peaks in 1959, 1966 and 1970 and the minima in 1963 and 1973, suggesting the impact of large scale phenomena (for instance, volcanic eruptions in 1963) in the modulation of visibility levels. A prominent feature in Fig. 11 is that the background visibility at the reference site has been also on a downward route since the mid 1950s, in accordance to the observed decreasing trend of the visibility in Athens. As already stated, the beginning of the 1950s signifies a period with an outstanding increase of emissions in Europe. European SO_2 emissions in particular, increased almost at a constant rate during the first half of the 20th century, while they experienced a quite abrupt increase in the 1950s and almost doubled their values between 1950 and 1960 (van Aardenne et al., 2001; Mylona, 1996). Figure 11 includes the rates of SO_2 increase per decade in Europe (in Tg S decade^{-1}), as reported by van Aardenne et al. (2001). Constant increasing rates (2 Tg S decade^{-1}) are observed untill 1950, when the rate of increase reached 6 Tg S decade^{-1} between 1950-1970. A decline of the increasing rate is then observed, while in the 1990s European sulfur emissions stabilize. Stabilization of emissions is followed by a continuous decline after 1990. Stjern et al. (2011) reported a prominent decrease of SO_x emissions and sulphate in aerosols in both eastern and western Europe from 1990-2007, but with higher rates of decrease in eastern Europe.

A very important finding in Fig. 11 is the similar slopes in the linear trends of the annual visibility at the background and urban stations, over their common period of observations (-2.2 km decade^{-1} and -2.4 km decade^{-1}, respectively). This feature implies that, apart from the absolutely lower values of visibility in the urban web of Athens, the inter-decadal variability of visibility in the city and the extended area is significantly modulated by large scale processes that control regional visibility, such as long-range pollution transport and/or changes of atmospheric circulation. Many studies have identified the eastern Mediterranean as a crossroad of aerosols of different origins, sizes and chemical composition (Lelieveld et al., 2002; Hatzianastassiou et al., 2009; Kanakidou et al., 2011; Gerasopoulos et al., 2011), which inevitably affect optical properties of the atmosphere. Kanakidou et al. (2011) found that even in the large urban regions of the eastern Mediterranean, particulate matter has a significant contribution by distant anthropogenic pollution sources in the region but also by long-range transport of African dust.

~~After the early 1990s, the time series diverge, with background visibility partly recovering, and visibility in Athens keeping declining at the same pace until 2003 (Fig. 11). Recovering of visibility at other Greek areas around the 1990s is also found by Lianou et al. (unpublished data) which is also in line with the observed visibility improvement in other European areas, related to emissions reduction (Wang et al., 2009; Vautard et al., 2009). This last feature suggests that during this period, local emissions might have a dominant role in the determination of visibility in Athens.~~

3.6 Visibility in Athens and AOD

The rel~~la~~ationship of visibility with AOD over Athens was also explored, using two different satellite based data (AVHRR and MODIS) from 1981-2009 and ~~data since~~ 2000-2014 respectively (see Section 2.5). For the AVHRR~~The~~ AOD at 630 nm, Fig. 12a shows a 1.7% per year decrease from 1981 to 1997 and a 2.4% decrease from 1999 to 2009 (1998 data were not available). It is interesting to point the AOD maxima in 1991 and 1992 that are linked with the Pinatubo eruption period. The AOD time series for the MODIS instrument at 550 nm~~time series~~ showed a significant and similar to AVHRR (2.4%~~(-2.4%~~ per year) decrease from 2000 ~~up~~ to 2010 and a further decrease of ~~(-~~7.4% per year~~)~~ for the period 2010-2014 (Fig.12b).~~period (Fig.12).~~

To investigate the relationship between visibility and AOD changes, the two parameters are plotted together after data binning. Visibility and AOD measurements have been used as follows: Visibility at 12:00 UT was used according to the indices defined in Table 2 and plotted against average AOD from synchronous satellite overpasses of AVHRR and MODIS, separately.~~.~~ The mean AOD and its standard deviation are presented in Fig. 13. The AOD values are related to~~with~~ the visibility data, using as the distance in km the middle point of each visibility bin (range). Only summertime (June-August) MODIS and AVHRR AOD have been used, to keep visibility values unaffected by~~from~~ other atmospheric parameters like low clouds, rain, or relative humidity. It is observed that for average AOD values for Athens (0.25 using the mean June-August AOD at 550nm from our MODIS AOD dataset or 0.23 at 500 nm as reported by Gerasopoulos et al., 2011), visibility varies within the range of 4 km to 10 km. Under~~For~~ cleaner conditions (W-NW-N, 0.12 - 0.17 at 500 nm, Gerasopoulos et al., 2011), visibility can go as high as 20 km, while very low visibility (< 0.5 km) is generally associated with~~to~~ the highest aerosol load~~s~~, with AOD > 0.3 (e.g. in the case of dust events, long-range transport of urban/industrial pollutants and stagnant conditions). It has to be noted that including both satellite datasets in the same figure provides information only on the summertime AOD vs visibility relationship. Average AOD from AVHRR and MODIS are not directly comparable, as they represent different time periods and different wavelengths.

Illustrating the relationship between AOD, which consist in a vertically integrated parameter, and visibility, a horizontally integrated parameter, requires various assumptions. Using satellite based AOD and visibility observations for GAA, when assuming a vertically constant extinction coefficient and a mixing layer that contains all aerosol load we end up describing the theoretical relationship (Koschmieder, 1924): Vis = k / AOD, where k is a function of the mixing layer height.

3.7 Visibility in relation to PM_{10}

An additional analysis was conducted to verify the relationship between visibility and particulate pollution from surface measurements, using a short data~~-~~set of PM_{10} in Athens as described in Section 2.5. Figure 14 presents visibility variation as a function of PM_{10} levels measured at Aristotelous (urban) and Maroussi (suburban) stations. Four different classes of PM_{10} levels were used, as shown in Fig. 14. The frequency of occurrence of each class is also shown in the figure. Despite the different locations and characteristics of the two stations, the observed frequencies are very similar in all classes of PM_{10} levels, with higher frequency corresponding to the class of 30~~-~~-60 μg m^{-3} at both stations. The frequency of $PM_{10} > 90$ μg m^{-3} at Aristotelous is double compared to the respective frequency at Maroussi. Independent~~ly~~ of the location, the same strong relationship is observed between visibility reported at NOA and PM_{10} levels at both stations, revealing a prominent decrease of visibility with increasing PM_{10} levels, in agreement with our conclusions. Average visibility at NOA range~~s~~d between 8 and 9 km under low PM_{10} levels (< 30 μg m^{-3}), but is reduced to less than 3 km under severe episodes of particulate pollution ($PM_{10} > 90$ μg m^{-3}). The correlation coefficient between daily~~ measurements of~~ PM_{10} levels and daily visibility at NOA was found equal to -0.38 ($p < 0.05$) and -0.36 ($p < 0.05$) for Aristotelous and Maroussi sites respectively.

Finally,~~Figure 15 displays~~ the variation of the ~~mean ~~annual averag~~lu~~es of PM_{10} values in Athens (Maroussi and Aristotelous stations) from 2004 to 2014 and at the reference site ~~station ~~of Finokalia (available~~(Crete)~~ over the 10-yr period ~~(~~2005-2014) are displayed in Fig. 15.~~, along with standard deviations.~~ A decreasing tendency of ~~in~~ PM_{10} levels is observed at all sites, indicating changes on both local and regional scale. Decreasing trends are more pronounced in Athens and particularly at Maroussi station (-2.4 μg m^{-3} yr^{-1}). The decreasing trend of PM_{10} levels ~~, which~~ is ~~also~~ consistent with the slight improvement~~recovery~~ of visibility ~~levels~~ in Athens over the same period.

4 Discussion and Conclusions

The present work analyses, for the first time, the historical record of visibility at NOA (~~in~~ Athens)~~ (NOA)~~ from 1931 to 2013 and interprets~~explores~~ its temporal ~~long-term~~ variability and trends~~. An attempt was made to interpret the temporal variations of visibility~~ in terms of relevant changes in~~of~~ atmospheric properties (related to local or regional processes) and/or meteorological conditions. Since this is the longest record of visibility observations in Greece and one of the oldest in the broader area of the eastern Mediterranean, the study provides unique~~analysis provided valuable~~ information on the atmospheric properties of the area in the past, when air pollution records are~~were~~ missing. The study period was divided into sub-periods corresponding to different trends in the time series of visibility, each sub-period being affected by different factors.

-The impact of meteorological conditions~~study period was divided into sub-periods corresponding to different visibility trends in the time series, each sub-period being affected by different factors.~~

~~The role of meteorology~~ on visibility was investigated in different ways. Visibility in Athens was found to follow~~reveal~~ a~~ distinct~~ seasonal cycle, with higher visibility corresponding to the warm and dry months of the year.~~ (namely from May to September) and lower to the colder and wet months.~~ Seasonality is more distinct~~evident~~ in the first sub-period of the time series (1931-1948), while after~~, when visibility in summer is up to 40% larger compared to winter. After~~ the 1950s, the seasonal cycle attenuates. Visibility~~ and the differences in visibility between summer and winter months were found to be much less pronounced (of the order of 10%, Fig. 6). Lower visibility values were observed in March in all sub-periods, resulting from the combination of enhanced pollen and biogenic aerosols emissions, but also to increased dust outbreaks from northern Africa and relatively higher RH levels.~~

~~As expected, visibility~~ was found to be negatively correlated with RH, the~~but~~ correlation being ~~is~~ stronger in the early part of the time series and attenuating~~first sub-period and attenuates~~ over the years. On the contrary, a positive correlation between visibility and wind speed was found,~~detected which is~~ statistically significant ~~(p <~~

~~0.01) only~~ during the late part of the time series, suggesting the increasing role of ~~recent decades. Actually, stronger~~ winds on the~~seem to improve visibility as they induce a~~ cleanup of the atmosphere from air pollutants.

~~-~~Visibility was also found to be ~~very~~ sensitive to wind direction, reflecting the influence of air masses origin on visibility.~~–~~ Lower visibility levels are constantly observed under southerly winds, corresponding ~~(Fig. 10). Such winds correspond~~ to sea breeze circulation, but also to dust outbreaks. ~~associated with increased humidity levels but also to accumulation of air pollutants in the city and formation of secondary air pollutants. In addition, some S/SW events are associated with strong wind speeds (Fig. 3) occurring during Sahara dust outbreaks. These events enrich Athens with airborne particles, thus decreasing visibility.~~

The study demonstrated that visibility in Athens has undergone a prominent impairment since the early 1930s. The overall trend of the annual visibility averages was found equal~~amounts~~ to -2.8 km decade^{-1}.~~–~~ The impressively higher levels of visibility in Athens before the 1950s (also characterized by strong seasonality) reflect the transparency of the atmosphere at that period, coherent with the~~inherent to~~ poorer aerosol load~~s~~ from anthropogenic emissions (urban and/or regional). The dramatic decrease of the visual air quality in the 1950s coincides with a number of events (end of wars, rapid urbanization and rapid increase of anthropogenic~~, increased~~ emissions on local and regional scale) and points to the prominent role of aerosol load~~s~~ in the atmosphere of Athens. Air pollution has gradually incurred a severe visual pollution in the city, with visibility lower to 4 km corresponding to ~~observed during~~ more than half of the year during the ~~recent~~ decade~~,~~ 2004-2013.~~The significant decrease of visibility in Athens was not accompanied with analogous significant trends in RH or precipitation (Figs 4, 7).~~

The comparison of the annual averages of visibility in Athens and~~with visibility~~ at a reference, non urban site (HER) in Crete, revealed ~~some very interesting features. First, visibility in Athens was found to be constantly lower compared to HER, possibly suggesting the impact of local anthropogenic emissions in the urban web. However, both time series revealed~~ similar and statistically significant negative trends at both sites, suggesting ~~over their common period of observations (after the mid 1950s), pointing to~~ the major contribution of long and regional range transport of natural and anthropogenic pollution sources in the GAA.~~. urban area. Visibility deterioration after the mid 1950s is also reported in most European areas, followed by stabilization and/or improvement around the 1980s or later (Vautard et al., 2009; van Beelen and van Delden, 2012; Stjern et al., 2011).~~ An improvement of visibility at HER around the 1990s was not associated with synchronous~~analogous~~ improvement of visibility in Athens, where visibility deterioration continued until the early 2000s. Although ~~(Fig.~~

11). At that period, negative trends of main gaseous air pollutants are reported in Athens at that period (Kalabokas et al., 1999a),1999a). However, the direct effect of such pollutants on light extinction is negligible compared to suspended particles and particularly to fine particles (< 1μm).

As already stated in Section 2.3, the contribution of both local and distant emission sources in PM concentrations in Athens is suggested by a number of studies (e.g. Kanakidou et al., 2011; Gerasopoulos et al., 2011). Mainly local emission sources (e.g. traffic) have been found to contribute to PM_{10} concentration (Chaloulakou et al., 2003; Grivas et al., 2004), while local anthropogenic sources seem to control PM_1 concentration only during the cold months of the year (Theodosi et al., 2011). Using satellite-based AOD measurements, Hatzianastassiou et al. (2009) found that local anthropogenic emissions in GAA contribute up to 30% to the total AOD.

A strong anticorrelation was found between visibility at NOA and PM_{10} levels in Athens, measured at two different stations (urban and suburban) in Athens over the period 2008-2012 (Fig. 14). The relationship between AOD and visibility in Athens was also examined in the study, using MODIS and AVHRR satellite data (Figs 12, 13). Illustrating the relationship between AOD, which consist in a vertically integrated parameter, and visibility, a horizontally integrated parameter, requires various assumptions. Using satellite based AOD and visibility observations for GAA, when assuming a vertically constant extinction coefficient and a mixing layer that contains all aerosol load we end up describing the theoretical relationship (Koschmieder, 1924): Vis = k / AOD, where k is a function of the mixing layer height.13), and confirmed their negative correlation.

The analysis showed a recent stabilization (or even slight improvement) of visibility in Athens, consistent with the observed decreasing trends of PM_{10} in the city from 2004 to 2014 (Fig. 15). This could possibly be related to reduced local anthropogenic emissions as a result of important transport infrastructures, but also of the economic recession in Greece. Although this last argument is already supported by some recent research studies, the impact of economic recession on local emissions seems to be more complicated and drawing conclusions remains tentative. Besides, in the same period, regional atmospheric pollution presents a decreasing tendency (Fig. 15), which is also consistent with the recent recovery of visibility in Athens.

The 82-years long time series of visibility in Athens unfolded for first time information on the atmospheric conditions overin the area, for periods when atmospheric pollution measurements are missing. Although the analysis is subject to several limitations and assumptions, associatedmainly related to the methods of visibility observations, the results are robust and statistically significant, as the outstanding degradation of the visual air quality in the city over the years.

The observed stabilization (or even slight improvement) of visibility in Athens in the very recent years could possibly be related to reduced local anthropogenic emissions as a result of important transport infrastructures (executed in view of Olympic Games) but also of the economic crisis in Greece. Although this last argument is already supported by some recent research studies (e.g. Vrekoussis et al., 2013; Santamouris et al., 2013), the impact of the economic crisis on local emissions seems to be more complicated and drawing out conclusions remains tentative. Besides, in the same period regional atmospheric pollution presents a decreasing tendency, as reflected in the negative trend of PM_{10} levels measured at the background station of Finokalia in Crete (Fig. 15) which is also consistent with the recent recovery of visibility in Athens.

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References

Appel, B.R., Tokiwa, Y., Hsu, J., Kothny, E., and Hahn, E.: –Visibility as related to atmospheric aerosol constituents, Atmos. Environ., 19, 1525-1534, doi:10.1016/0004-6981(85)90290-2, 1985.

Bäumer, D., Vogel, B., Versick, S., Rinke, R., Möhler, O., and Schnaiter, M.: -Relationship of visibility, aerosol optical thickness and aerosol size distribution in an ageing air mass over South-West Germany, Atmos. Environ., 42, 989-998, doi:10.1016/j.atmosenv.2007.10.017, 2008.

Bond, T.C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S., Roden, C., Streets, D.G., and Trautmann, N.M.: Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850-2000, Glob. Biochem. Cycles, 21, GB2018, doi:10.1029/2006GB002840, 2007.

Carapiperis, L.N., and Karapiperis, P.P.: On the ocean colour of the sky in Athens, Academy of Athens, 27, 213, 1952.

Cermak, J., Wild, M., Knutti, R., Mishchenko M.I., and Heidinger, A.K.: Consistency of global satellite-derived aerosol and cloud data sets with recent brightening observations, Geophys. Res. Lett., 37, L21704, doi:10.1029/2010GL044632, 2010.

Chaloulakou, A., Kassomenos, P., Spyrellis, N., Demokritou, P., and Koutrakis, P.: Measurements of PM_{10} and $PM_{2.5}$ particle concentrations in Athens, Greece, Atmos. Environ., 37, 649 – 660, doi:-10.1016/S1352-2310(02)00898-1, 2003.

Chan, Y.C., Simpson, R.W., Mctainsh, G.H., Vowles, P.D., Cohen, D.D., and Bailey, G.M.: Source apportionment of visibility degradation problems in Brisbane (Australia) using the multiple linear regression techniques, Atmos. Environ., 33, 3237–3250, doi:10.1016/S1352-2310(99)00091-6, 1999.

Chan, P.K., Zhao, X.P., and Heidinger, A.K.: Long-Term Aerosol Climate Data Record Derived from Operational AVHRR Satellite Observations, Dataset Papers in Geosciences, vol. 2013, Article ID 140791, 5 pages, 2013. doi:10.7167/2013/140791, 2013.

Chang, D., Song, Y., and Liu, B.: Visibility trends in six megacities in China 1973–2007, Atmos. Res., 94, 161–167, doi:10.1016/j.atmosres.2009.05.006, 2009.

Che, H.–Z., Zhang, X.–Y., Li, Y., Zou, Z.–J., and Qu, J.–J.: Horizontal visibility trends in China 1981-2005, Geophys. Res. Lett., 34, L24706, doi:10.1029/2007GL031450, 2007.

Colbeck, I., Chung, M.C., and Eleftheriadis, K.: Formation and transport of atmospheric aerosol over Athens, Greece, Water Air Soil Pollut., 223-235, doi:10.1023/A:1021335401558, 2002.

Davis, R.–E.: A synoptic climatological analysis of winter visibility trends in the mideastern United States, Atmos. Environ., 25b, 165-175, doi:10.1016/0957-1272(91)90052-G ,1991.

Dayan, U., and Levy, I.: The Influence of Meteorological Conditions and Atmospheric Circulation Types on PM10 and Visibility in Tel Aviv, J. Appl. Meteorol., 44, 606-619, doi: /10.1175/JAM2232.1, 2005.

Deng, J.J., Wang, T.-J., Jiang, Z.Q., Xie, M., Zhang, R.-J., Huang, X.-X., and Zhu, J.-L.: Characterization of visibility and its affecting factors over Nanjing, China, Atmos. Res., 101, 681–691, doi:10.1016/j.atmosres.2011.04.016, 2011.

Doyle, M., and Dorling, S.: Visibility trends in the UK 1950-1997, Atmos. Environ., 36, 3161-3172, doi:10.1016/S1352-2310(02)00248-0, 2002.

Draxler, R., Stunder, B., Rolph, G., Stein, A., and Taylor, A.: Hybrid Single–Particle Lagrangian Integrated Trajectories (HY–SPLIT): Version 4.9 - User's Guide and Model Description, http://www.arl.noaa.gov/documents/reports/hysplit user guide.pdf, 2009.

Eidels-Dubovoi, S.: Aerosol impacts on visible light extinction in the atmosphere of Mexico City, Sci. Total Environ., 287, 213–220, doi:10.1016/S0048-9697(01)00983-4, 2002.

Elias, T., Haeffelin, M., Drobinski, P., Gomes, L., Rangognio, J., Bergot, T., Chazette, P., Raut, J.C., and Colomb, M.: Particulate contribution to extinction of visible radiation: pollution, haze, and fog, Atmos. Res., 92, 443–454, doi:10.1016/j.atmosres.2009.01.006, 2009.

Eltbaakh Y.-A., Ruslan, M.-H., Alghoul, M.-A., Othman, M.-Y., and Sopian, K.: Issues concerning atmospheric turbidity indices, Renw. Sustain. Energy Rev., 16, 6285-6294, doi: 10.1016/j.rser.2012.05.034, 2012.

Folini, D., and Wild, M.: Aerosol emissions and dimming/brightening in Europe: Sensitivity studies with ECHAM5-HAM, J. Geophys. Res., 116, D21, doi:10.1029/2011JD016227, 2011.

Founda, D.: Evolution of the air temperature in Athens and evidence of climatic change: A review, Advances in Building Energy Research, 5, 7- 41, doi:10.1080/17512549.2011.582338, 2011.

Founda, D., Pierros, F., Petrakis, M., and Zerefos, C.: Inter-decadal variations and trends of the Urban Heat Island in Athens (Greece) and its response to heat waves, Atmos. Res.,161, 1-13. doi:10.1016/j.atmosres.2015.03.016, 2015.

Gerasopoulos, E., Kouvarakis, G., Vrekoussis, M., Kanakidou, M., and Mihalopoulos, N.: Ozone variability in the marine boundary layer of the Eastern Mediterranean based on 7-year observations, J. Geophys. Res., 110, D15309, doi:10.1029/2005JD005991, 2005.

Gerasopoulos, E., Amiridis, V., Kazadzis, S., Kokkalis, P., Eleftheratos, K., Andreae, M.-O., Andreae, T.-W., El-Askary, H., and Zerefos, C.-S.: Three-year ground based measurements of aerosol optical depth over the Eastern Mediterranean: The urban environment of Athens, Atmos. Chem. Phys., 11, 2145-2159, doi:10.5194/acp-11-2145-2011, 2011.

Ghim, Y.S., Moon, K., Lee, S., and Kim, Y.-P.: Visibility trends in Korea during the past two decades, J. Air Waste Manage Assoc., 55, 73-82, doi:10.1080/10473289.2005.10464599, 2005.

Gkikas, A., Basart, S., Hatzianastassiou, N., Marinou, E., Amiridis, V., Kazadzis, S., Pey, J., Querol, X., Jorba, O., Gassó, S., and Baldasano, J. M.: Mediterranean desert dust outbreaks and their vertical structure based on remote sensing data, Atmos. Chem. Phys., 15, 27675-27748, doi:10.5194/acpd-15-27675-2015, 2015.

Grivas, G., Chaloulakou, A., Samara, C., and Spyrellis, N.: Spatial and temporal variation of PM10 mass concentrations within the Greater Area of Athens, Greece, Water Air Soil Pollut., 158, 357-71, doi:10.1023/B:WATE.0000044859.84066.09, 2004.

Hamonou, E., Chazette, P., Balis, D., Dulac, F., Schneider, X., Galani, E., Ancellet, G., and Papayannis, A.: Characterization of the vertical structure of Saharan dust export to the Mediterranean basin, J. Geophys. Res, 104, 22257-22270, doi:10.1029/1999JD900257, 1999.

Hand, J.L., Kreidenweis, S.M., Sherman, D.-E., Collett, Jr J.L., Hering, S.V., Day, D.E, and Malm, W.C.: Aerosol size distributions and visibility estimates during the Big Bend Regional Aerosol and Visibility Observational (BRAVO) study, Atmos. Environ., 36, 5043-5055, doi:10.1016/S1352-2310(02)00568-X, 2002.

Hatzianastassiou, N., Gkikas, A., Mihalopoulos, N., Torres, O., and Katsoulis, B.-D.: Natural versus anthropogenic aerosols in the eastern Mediterranean basin derived from multiyear TOMS and MODIS satellite data, J. Geophys. Res., 114, D24202, doi:10.1029/2009JD011982, 2009.

Hoek, G., Forsberg, B., Borowska, M., Hlawiczka, S., Vaskovi, E, Welinder, H., et al.: Wintertime PM-10 and black smoke concentrations across Europe: results from the Peace study, Atmos. Environ., 31, 3609-3622, doi:10.1016/S1352-2310(97)00158-1, 1997.

Ichoku, C., Chu, D.A., Mattoo, S. et al.: A spatio-temporal approach for global validation and analysis of MODIS aerosol products, Geophys. Res. Lett., 29, 1- 4, doi:10.1029/2001GL013206, 2002.

Kalabokas, P.-D., Viras, L.G., and Repapis, C.C.: Analysis of 11-year record (1987-1997) of air pollution measurements in Athens, Greece, Part I: primary air pollutants, Global Nest, 1, 157-167, 1999a.

Kalabokas, P.D., Viras, L.G., Repapis, C.C., and Bartzis, J.G.: Analysis of 11-year record (1987-1997) of air pollution measurements in Athens, Greece, Part II: photochemical air pollutants, Global Nest, 1, 169-176, 1999b.

Kanakidou, M., Mihalopoulos, N., Kindap, T., Im, U. et al.: Megacities as hot spots of air pollution in the East Mediterranean, Atmos. Environ., 45, 1223-1235, doi:10.1016/j.atmosenv.2010.11.048, 2011.

Kanellopoulou, E.: Study of the visibility of Athens. PhD Thesis (in Greek), 1979.

Kim, K.–W.: Physico-chemical characteristics of visibility impairment by airborne pollen in an urban area, Atmos. Environ., 41, 3565–357, doi:10.1016/j.atmosenv.2006.12.054, 2007.

Kim, K.W.: Optical Properties of Size-Resolved Aerosol Chemistry and visibility Variation Observed in the Urban Site of Seoul, Korea, Aerosol Air Qual. Res., 15, 271–283, doi: 10.4209/aaqr.2013.11.0347, 2015.

Koschmieder, H.: Theorie der horizontalen sichtweite, Beitr. Phys. Frei. Atmos., 12, 171–181, 1924.

Kouvarakis G., Tsigaridis, K., Kanakidou, M., and Mihalopoulos, N.: Temporal variations of surface regional background ozone over Crete Island in southeast Mediterranean, J. Geophys. Res., 105, 4399-4407, doi:10.1029/1999JD900984, 2000.

Larson, S.M., and Cass, G.R.: Characteristics of summer midday low-visibility events in the Los Angeles area, Environ. Sci. Technol., 23, 281–289, doi: 10.1021/es00180a003, 1989.

Lee, D.–O.: Regional variations in long-term visibility trends in the UK (1962–1990), Geog., 79, 108–121, http://www.jstor.org/stable/40572408, 1994.

Léon, J.-F., Chazette, P., and Dulac, F.: Retrieval and monitoring of aerosol optical thickness over an urban area by spaceborne and ground-based remote sensing, Appl. Opt., 38, 6918-6926, doi:10.1364/AO.38.006918, 1999

Lelieveld, J., Berresheim, H., Borrmann, S., Crutzen, P.–J., et al.: Global Air Pollution Crossroads over the Mediterranean, Science, 298, 794-799, doi: 10.1126/science.1075457, 2002.

Malm, W.-C.: Introduction to Visibility, Air Resources Division, National Park Service, Cooperative Institute for Research in the Atmosphere (CIRA), NPS Visibility Program, Colorado State University, Fort Collins, CO, May, 1999.

Maloutas, T.: The self promoting housing solution in post war Athens, Discussion Paper Series 9(6) 95-110, Available online at: http://www.prd.uth.gr/research/DP/2003/uth-prd-dp-2003-6_en.pdf, 2003.

Mavroidis, I., and Ilia, M.: Trends of NOx~~X~~, NO2 and O3 concentrations, at three different types of air quality monitoring stations in Athens, Greece, Atmos. Environ., 63,135–147, doi:10.1016/j.atmosenv.2012.09.030, 2012.

Mishchenko, M.I., Geogdzhayev, I.V., Rossow, W.B., Cairns, B., Carlson, B.E., Lacis, A.A., Liu, L., and Travis, L.D.: Long-term satellite record reveals likely recent aerosol trend, Science, 315, no. 5818, p. 1543, doi:10.1126/science.1136709, 2007.

Mylona, S.: Sulfur dioxide emissions in Europe 1880-1991 and their effect on sulphur concentrations and depositions, Tellus, 48, 662-689, doi/10.1034/j.1600-0889.1996.t01-2-00005.x, 1996.

Nabat, P., Somot, S., Mallet, M., Sanchez-Lorenzo, A., and Wild, M.: Contribution of anthropogenic sulfate aerosols to the changing Euro-Mediterranean climate since 1980, Geophys. Res. Lett., 41, 5605-5611, doi:10.1002/2014GL060798, 2014.

Paraskevopoulou, D., Liakakou, E., Gerasopoulos, E., Theodosi, C., and Mihalopoulos, N.: Long-term characterization of organic and elemental carbon in the $PM_{2.5}$ fraction: the case of Athens, Greece, Atmos. Chem. Phys., 14, 13313–13325, doi:10.5194/acp-14-13313-2014, 2014.

Paraskevopoulou, D., Liakakou, E., Gerasopoulos, E., and Mihalopoulos, N.: Sources of atmospheric aerosols from long-term measurements (5 years) of chemical composition in Athens, Greece, Sci. Total Environ., 527–528, 165–178, doi:10.1016/j.scitotenv.2015.04.022, 2015.

Remoudaki, E., Gergametti, G., and Losno, R.: On the dynamic of the atmospheric input of copper and manganese into the western Mediterranean Sea, Atmos. Environ., 25A, 733-744, doi:10.1016/0960-1686(91)90072-F, 1991a.

Remoudaki, E., Gergametti, G., and Buat-Ménard, P.: Temporal variability of atmospheric lead concentrations and fluxes over the northwestern Mediterranean Sea, J. Geophys. Res., 96, 1043-1055, doi:10.1029/90JD00111, 1991b.

Santamouris, M., Paravantis, J.A., Founda, D., Kolokotsa, D., Michalakakou, P., Papadopoulos, A.–M., Kontoulis, N., Tzavali, A., Stigka, E.-K., Ioannidis, Z., Mehilli, A., Matthiessen, A., and Servou, E.: Financial Crisis and Energy Consumption: A household Survey in Greece, Energy Build., 65, 477-487, doi:10.1016/j.enbuild.2013.06.024, 2013.

Singh, A., and Dey, S.: Influence of aerosol composition on visibility in megacity Delhi, Atmos. Environ., 62, 367- 373, doi:10.1016/j.atmosenv.2012.08.048, 2012.

Sloane, C.S.: Visibility trends - II. Mideastern United States 1948-1978, Atmos. Environ., 16, 2309-2321, doi:10.1016/0004-6981(82)90117-2, 1982.

Smirnov, A., Holben, B. N., Sakerin, S.M., Kabanov, D. M., Slutsker, I., Chin, M., Diehl, T.L., Remer, L.A., Kahn, R.A., Ignatov, A., Liu, L., Mishchenko, M., Eck, T.F., Kucsera, T.L., Giles, D.M., and Kopelevich, O.V.: Ship-based aerosol optical depth measurements in the Atlantic Ocean, comparison with satellite retrievals and GOCART model, Geophys. Res. Lett., 33, L14817, doi:10.1029/2006GL026051, 2006.

Stjern, C.-W., Stohl, A., and Kristjánsson, J.-E.: Have aerosols affected trends in visibility and precipitation in Europe? J. Geophys. Res., 116, D02212, doi:10.1029/2010JD014603, 2011.

Streets, D.–G., Wu, Y., and Chin, M.: Two-decadal aerosol trends as a likely explanation of the global dimming/brightening transition, Geophys. Res. Lett., 33, L15806, doi:10.1029/2006GL026471, 2006.

Tang, I.N.: Chemical and size effects of hygroscopic aerosols on light scattering coefficients, J. Geophys. Res., 101, 19245–19250, doi: 10.1029/96JD03003, 1996.

Theodosi, C., Grivas, G., Zarmpas, P., Chaloulakou, A., and Mihalopoulos, N.: Mass and chemical composition of size- segregated aerosols (PM_1, $PM_{2.5}$, PM_{10}) over Athens, Greece: local versus regional sources, Atmos. Chem. Phys., 11, 11895– 11911, doi:10.5194/acp-11-11895-2011, 2011.

Tsai, Y.-I, Lin, Y.H., and Lee, S.-Z.: Visibility variation with air qualities in the metropolitan area of southern Taiwan, Water Air Soil Pollut., 144, 19-40, doi:10.1023/A:1022901808656, 2003.

Tsai, Y.I., Kuo, S.C., Lee, W.J., Chen, C.L., and Chen, P.T.: Long-term visibility trends in one highly urbanized, one highly industrialized, and two rural areas of Taiwan, Sci. Total Environ., 382, 324–341, doi:10.1016/j.scitotenv.2007.04.048, 2007.

van Aardenne, J.-A., Dentener, F.-J., Olivier, J.-G.-J., Klein Goldewijk, C.-G. M., and Lelieveld, J.: A 1°×1° resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990, Glob. Biochem. Cycles, 15, 909-928, doi: 10.1029/2000GB001265, 2001.

van Beelen, A.J., and van Delden, A.J.: Cleaner air brings better views, more sunshine and warmer summer days in the Netherlands, Weather, 67, 21-25, doi: 10.1002/wea.854, 2012.

Vautard, R., Yiou, P., and Oldenborgh, G.: Decline of fog, mist and haze in Europe over the past 30 years, Nat. Geosci., 2, 115-119, doi:10.1038/NGEO414, doi:10.1038/ngeo414, 2009.

Vestreng, V., Ntziachristos, L., Semb, A., Reis, S., Isaksen, I.S.A., and Tarrasón, L.: Evolution of NOx emissions in Europe with focus on road transport control measures, Atmos. Chem. Phys., 9, 1503-1520, doi: 10.5194/acp-9-1503-2009, 2009.

Vestreng, V., Myhre, G., Fagerli, H., Reis, S., and Tarrasón, L.: Twenty five years of continuous sulphur dioxide emission reduction in Europe, Atmos. Chem. Phys., 7, 3663-3681, doi:10.5194/acp-7-3663-2007, 2007.

Vrekoussis, M., Richter, A., Hilboll, A., Burrows, J.-P., Gerasopoulos, E., Lelieveld, J., Barrie, L., Zerefos, C., and Mihalopoulos, N.: Economic crisis detected from space: Air quality observations over Athens, Greece, Geophys. Res. Lett., 40, 458-463, doi:10.1002/grl.50118, 2013.

Wan, J.M., Lin, M., Chan, C.Y., Zhang, Z.S., Engling, G., Wang, X.M., Chan, I.N., and Li, S.Y.: Change of air quality and its impact on atmospheric visibility in central-western Pearl River Delta, Environ. Monit. Assess., 172, 339-351, doi: 10.1007/s10661-010-1338-2, 2011.

Wang, K., Dickinson, R.E., and Liang, S.: Clear sky visibility has decreased over land globally from 1973 to 2007, Science, 323, 1468-1470, doi:10.1126/science.1167549, 2009.

Wang, K.-C., Dickinson, R.-E., Su, L., and Trenberth, K.-E.: Contrasting trends of mass and optical properties of aerosols over the Northern Hemisphere from 1992 to 2011, Atmos. Chem. Phys., 12, 9387–9398, doi:10.5194/acp-12-9387-2012, 2012.

Wild, M., 2009: Global dimming and brightening: A review, J. Geophys. Res., 114, doi: 10.1029/2008JD011470, 2009.

Wu, J., Fu, C., Zhang, L., and Tang, J.: Trends of visibility on sunny days in China in the recent 50 years, Atmos. Environ., 5, 339-346, doi:10.1016/j.atmosenv.2012.03.037, 2012.

World Meteorological Organization: The WMO Automatic Digital Barometer inter comparison (J. P. van der Meulen), Instrument and Observing Methods Report No.46, WMO/TD-No.474, Geneva, 1992.

Zhao, P., Zhang, X., Xu, X. and Zhao, X.: Long-Term Visibility Trends and Characteristics in the Region of Beijing, Tianjin, and Hebei, China, Atmos. Res., 101, 711–718, doi:10.1016/j.atmosres.2011.04.019, 2011.

Zhao X., Chan, P., and NOAA CDR Program: NOAA Climate Data Record (CDR) of AVHRR Daily and Monthly Aerosol Optical Thickness over Global Oceans, Version 2.0. AOT1, NOAA National Centers for Environmental Information. doi:10.7289/V5SB43PD, 2014.

FIGURES 1, 3, 5, 6, 7, 10, 11, 12, 13 15 WERE ALSO REVISED OR RECREATED