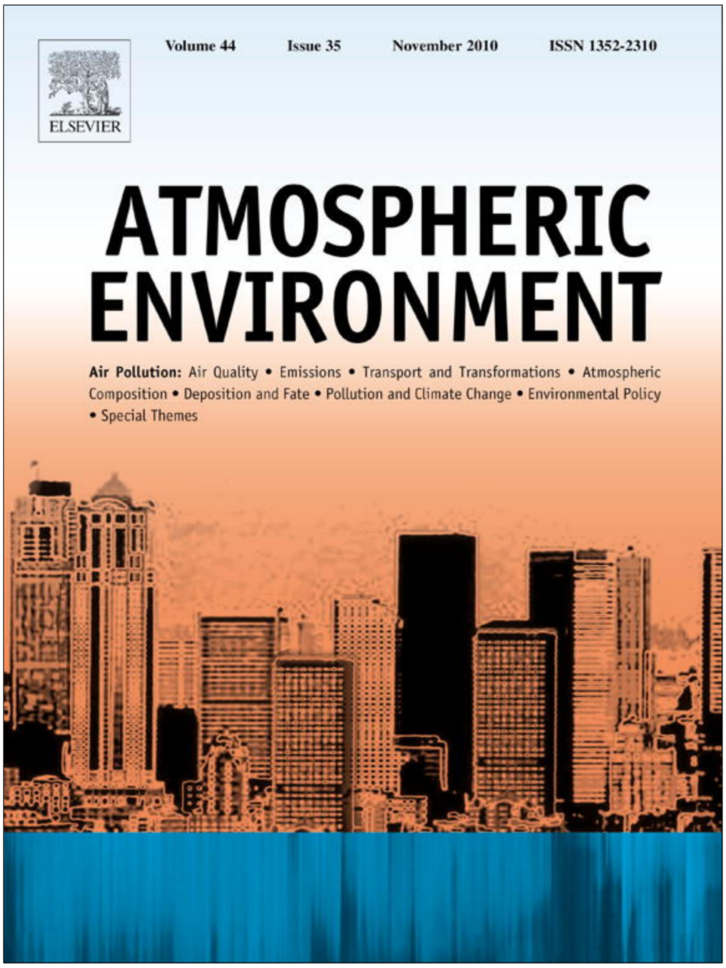
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Short communication

Effect of fireworks on ambient air quality in Malta

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ABSTRACT

Religious festivals (*festas*) in the densely populated Maltese archipelago (Central Mediterranean) are ubiquitous during summer when 86 of them are celebrated between June and October, each involving the burning of fireworks both in ground and aerial displays over a period of 3 days or longer per festival. We assessed the effect of fireworks on the air quality by comparing PM_{10} and its content of Al, Ba, Cu, Sr and Sb which materials are used in pyrotechnic compositions. PM_{10} was collected mainly from two sites, one in Malta (an urban background site) and the other in Gozo (a rural site) during July–August 2005 when 59 feasts were celebrated and September–October 2005 when only 11 feasts occurred. For both Malta and Gozo, PM_{10} and metal concentration levels measured as weekly means were significantly higher during July–August compared to September–October and there exist strong correlations between PM_{10} and total metal content. Additionally, for Malta dust, Al, Ba, Cu and Sr correlated strongly with each other and also with total concentration of all five metals. The same parameters measured in April 2006 in Malta were at levels similar to those found in the previous October. Ba and Sb in dust from the urban background site in Malta during July–August were at comparable or higher concentration than recently reported values in PM_{10} from a heavily-trafficked London road and this suggests that these metals are locally not dominated by sources from roadside materials such as break liner wear but more likely by particulate waste from fireworks. Our findings point to the fact that *festa* firework displays contribute significantly and for a prolonged period every year to airborne dust in Malta where PM_{10} is an intractable air quality concern. The presence in this dust of elevated levels of Ba and especially Sb, a possible carcinogen, is of concern to health.

1. Introduction

The importance to cultural life of fireworks in Malta is probably unequalled anywhere else. Summer on this central Mediterranean archipelago of two main islands (Malta and Gozo) is the *festa* season when no less than 86 religious festivals dedicated to some Catholic saint or other are held between June and September in the various towns and villages. The archipelago is geographically very small (316 km^2) and densely populated (*ca.* 405 000) and 200 t of chemicals related to the manufacture of pyrotechnic material were imported in 2007 (NSO, 2007). Ground firework displays draw large crowds that congregate in the town square and aerial fireworks which dominate the Maltese *festa* are let off at intervals for several days preceding the principal festival day on the Sunday and culminating in intensity and colourful display during the evening of the Saturday and *festa* Sunday. Smoke from fireworks, laden with metallic salts derived from the presence in pyrotechnic compositions of colour-generating Cu, Ba, Sr and other salts as well as Sb_2S_3 used as tinder is produced in abundance and is harmful if inhaled. A number of publications have appeared describing the environmental (Kulshrestha et al., 2004; Wang et al., 2007; Vecchi et al., 2008; Steinhauser et al., 2008; Barman et al., 2008) and health impact (Smith and Vu, 1975; Hirai et al., 2000; Murty, 2000) of firework smoke. It is not unreasonable to expect that the burning of fireworks in Malta throughout the long *festa* season could be exerting a significant impact on the local air quality and also possibly the health of the population.

The objective of this work was to establish whether the concentration and composition of PM_{10} in ambient air in Malta is effected measurably by fireworks emissions. Sampling was performed continuously during the period July to October 2005 and again on 28th April 2006 and the amount of PM_{10} and content of five trace elements postulated to be specific to fireworks were determined with the main objective of comparing the abundance and properties of dust collected in the period July–August when a total of 59 feasts are celebrated with that of September–October when only 11 feasts take place. The measurement of April 2006, made in a period during which no religious festivals are celebrated,

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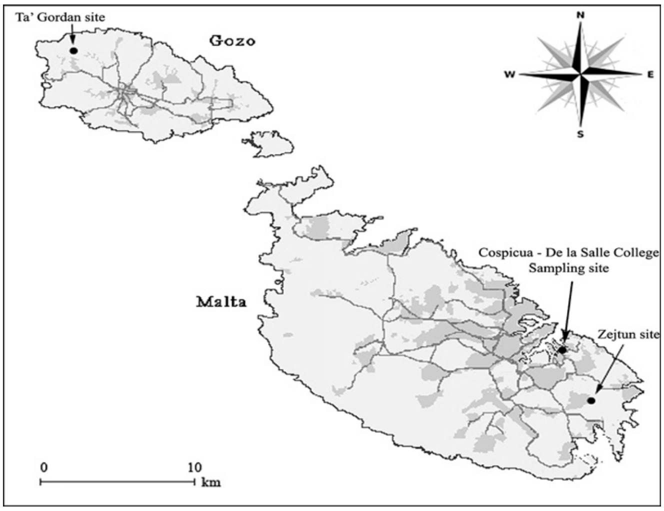

Fig. 1. Map of Malta and Gozo showing the sites of collection of PM_{10} dust.

served as further background control. This investigation differs from prior work on the subject in that it targeted for measurement a summer-long observable effect on air quality rather than a short term impact immediately following a single pyrotechnic display.

2. Experimental

For the period July to October 2005, weekly samples of PM_{10} were collected from two sites, one upwind (as 'background') and the other ('target' site) downwind from the mainland areas. Since the prevailing winds blow from the NW, the target site was at the south eastern town of Zejtun (Malta) while the background site was at Ta' Gordan Lighthouse in the north western part of the island of Gozo (Fig. 1). At Zejtun, the PM_{10} sampler was located on the roof of a two storey residential unit at 6 m above street level in an area of the town regarded as a 'background urban site' by the Malta Environment and Planning Authority. The lighthouse at Ta' Gordan is located at 200 m above sea level in a rural agricultural area about 1 km from the northern coast of Gozo. One week samples were collected nearly-simultaneously for successive periods during July–October 2005 from Zejtun and Ta' Gordan. One 24 h sample was collected on 28th April 2006 from the roof of De La Salle College in Cospicua, Malta, a town that is also located in the south-east of the island close to Zejtun (Fig. 1). All samples were collected using Leckel small volume air samplers equipped with a PM_{10} inlet. The sampler maintains a constant flow rate of 2.3 $m^3 h^{-1}$ and the inlet conforms to EN12341, a European standard for PM_{10} monitoring. Teflon filters (Whatman), diameter 46.2 mm, were conditioned for 48 h at 52% relative humidity and 24 ± 1 °C prior to weighing with a Sartorius balance (Model BL120S) having a precision of ± 0.1 mg. Loaded filters were stored at <10 °C pending analysis and field blanks (one in every 10 samples) were taken as controls against contamination.

The loaded filters were dissolved in a mixture of concentrated nitric (2.5 mL) and hydrochloric (0.5 mL) acids (Fluka, *puriss* grade) in a Teflon cup inside a Teflon bomb (Parr Scientific) heated by microwave oven (Boamann) at 525 W and using a digestion cycle that ensures that the bomb's external wall temperature remains below 40 °C. The contents, after cooling, were made up to 25 mL with distilled water and stored in polypropylene bottles pending analysis by graphite furnace atomic absorption spectrophotometry (Shimadzu Model AA6800). Calibration standards were prepared by dilution from concentrated standards.

The recovery efficiency for each analyte was assessed by adding 100 μL of a 50% aqueous isopropanol (Aldrich, Analar grade) solution containing each of the five analyte cations at 20 ppm onto a Teflon filter in a Teflon cup which was allowed to stand for 1 h to evaporate the solvent. The filter was then worked up as for the field samples to produce 50 mL of solution. A 4-point standard addition calibration method was adopted and each sample was analysed twice for concordancy using an injection volume of 20 μL for each run. Palladium(II) nitrate (Fluka, *puriss* grade) was used as a matrix modifier in the analysis of antimony. All calibration plots had R^2 values >0.99. The detection limits (estimated as $3\sigma/S$ where S is the sensitivity or gradient of the calibration curve and σ the standard deviation of the absorbance values of the blank controls) and recovery efficiencies for each analyte were as shown in Table 1.

Table 1
Detection limits and recovery efficiencies of analytes.

Metal	Cu	Sr	Ba	Al	Sb
Detection limit (ppb)	0.030	0.12	0.18	0.078	0.12
Recovery efficiency (%)	102.5	98.4	93.9	100.5	98.7

3. Results and discussion

Except for three occasions due to problems with the sampling, each dust sample taken during July–October was collected over a 7 day period and hence analytical information generated on it is referred to as a 'weekly mean'. Weekly mean concentrations of PM_{10} collected from Zejtun Malta are shown in Table 2a which also shows the occurrence or otherwise of precipitation. Table 2b shows corresponding data for Ta' Gordan Gozo. The weekly means and their calculated monthly average PM_{10} values for July–October 2005 are plotted in Fig. 2: the monthly average value for August for dust collected in Gozo incorporates the value for the week starting 28th July.

It is clear that, during July–October 2005, PM_{10} loads in Malta air were always higher than corresponding values in Gozo. Moreover, the Pearson correlation factor for PM_{10} at the two sites is 0.969 ($p = 0.000$) which suggests that the sites are effected by the same

Table 2a
Mean weekly mass concentration and chemical composition of PM_{10} for Zejtun, Malta.

Malta – Zejtun sampling site														
Sampling start date		20/7/05	27/7/05	3/8/05	8/8/05	17/8/05	27/8/05	30/8/05	6/9/05	13/9/05	27/9/05	7/10/05	15/10/05	20/10/05
Mean Weekly PM_{10} ($\mu g\ m^{-3}$)		42.9	49.6	58.0	40.1	37.1	28.5	41.6	40.3	39.3	25.1	27.1	33.3	42.7
Mean weekly concentration ($ng\ m^{-3}$)	Cu	10.9	11.6	9.0	3.2	9.2	5.6	6.5	1.7	11.6	4.7	5.3	4.7	2.4
	Sr	8.3	12.8	14.7	5.8	14.2	4.7	4.7	6.6	16.5	3.2	3.7	3.0	6.7
	Ba	18.3	33.2	18.4	28.4	46.6	10.2	4.9	4.5	17.2	4.6	5.5	3.8	3.9
	Al	348.0	413.7	357.5	75.5	298.0	106.1	105.6	79.5	389.8	56.8	101.2	41.8	190.9
	Sb	20.9	19.3	3.1	0.9	2.7	2.4	1.7	0.9	2.7	1.3	1.0	1.1	0.3
Precipitation[a]		–	–	–	–	–	+	–	–	–	+	–	–	–

[a] + = rainfall of >2 mm; – = no rain or rainfall <2 mm.

Table 2b
Mean weekly mass concentration and chemical composition of PM_{10} for Ta' Gordan, Gozo.

Gozo –Ta' Gordan sampling site											
Sampling start date		28/7/05	01/8/05	09/8/05	25/8/05	31/8/05	13/9/05	20/9/05	27/9/05	07/10/05	14/10/05
Mean Weekly PM_{10} ($\mu g\,m^{-3}$)		35.2	39.6	21.2	14.4	24.2	17.6	10.2	11.5	13.3	17.8
Mean weekly concentration($ng\,m^{-3}$)	Cu	4.2	2.8	1.7	1.1	2.9	1.2	1.2	1.2	0.8	1.4
	Sr	4.7	6.3	2.7	0.9	2.2	2.4	1.3	1.5	1.5	1.8
	Ba	5.0	5.4	1.7	4.3	3.5	6.5	0.9	1.0	1.3	1.2
	Al	253.2	511.4	89.7	31.4	89.9	119.5	12.1	33.8	38.2	53.8
	Sb	0.3	0.5	0.2	0.7	1.9	0.4	0.3	0.2	0.1	0.4
Precipitation		–	–	–	+	–	–	+	+	–	–

source/s although to different extent. At both sites PM_{10} concentrations decrease from July to October although not linearly with the number of *festa* occurrences which were as follows: July 32; August 27; September 10 and October 1. Tables 2a and 2b also contain data on the chemical composition of the dust in the form of mean weekly concentrations of the metals for Malta and Gozo respectively and this data are plotted in Fig. 3a and b which also show the average value for each month. Except for Sb in Gozo air, higher levels of the metals were recorded for all samples during July and August: the average for Zejtun dust for July–August contained Cu and Sr that were 1.6 times higher than the average value for September–October. Ba concentration in July-August (26 $ng\,m^{-3}$) decreased by a factor of 4 in September–October and Sb by a factor of 6. Metal concentrations in the dust sample collected in April 2006 (Table 2c) were significantly lower than those measured during the summer *festa* period of July–August. Even though these values refer to 24 h means and are therefore not directly comparable with the weekly means measured during the previous study period, they do indicate that, during the non-festival months, the metals in PM_{10} probably remain at concentrations similar to those measured in October. It is not unreasonable to presume that these values increase again during the summer in response to the next festival fireworks season of the following year.

The Pearson correlation coefficient was used to establish whether changes in the mean weekly concentration of the five metals were correlated with each other and with PM_{10}. For this purpose, only the data collected during July–October 2005 were employed. The group Sb, Ba, Cu and Sr hypothesised to be sourced mainly from fireworks were designated as 'exotic metals' while all five including Al were designated as 'metals'. Each element was separately correlated with the sum of the 'metals' and that of the 'exotic metals' respectively. Fig. 4 illustrates the correlation results for Malta graphically: a high degree of correlation (at $p < 0.01$) is characterized by a steep gradient of the trend line.

For Malta dust, Al, Ba, Cu and Sr correlated strongly with each other and also with total 'exotic metals' and 'metals'. Sb also correlated but not as strongly with Cu, Al and 'metals' and 'exotic metals' but not at all with Sr or Ba. These results suggest that the metals originate from the same source which is most likely fireworks burning. It is to be noted that antimony (as Sb_2S_3) is mainly used in flash crackers as a tinder and the lack of correlation of this metalloid with Sr and Ba (employed in other compositions) is not unexpected. PM_{10} is correlated with Al, Sr and total 'metals' and this finding suggests that fireworks do affect the ambient air quality in Malta. During the entire period of measurement, there were no firework displays at Zejtun because the village feast had already occurred in June: thus ambient air in this town was affected negatively for at least two months by fireworks in towns that are 2 to 10 km away. If air at Zejtun was so influenced, we conclude that air quality over most of the territory of Malta is similarly influenced for the duration of this period and possibly to a higher degree depending on the proximity of the town to the displays.

For Gozo dust (Fig. 5) Al correlated even more strongly with PM_{10} and this metal was also strongly correlated with Sr, 'exotic metals' and 'metals' and moderately with Cu but not with Ba and Sb. Cu and Sr were also strongly correlated with PM_{10} and with 'exotic metals' and 'metals' but again not with Sb and Ba. Ba only correlated with the 'exotic metals'. In Gozo both 'metals' and 'exotic metals' were very strongly correlated with PM_{10} and this suggests that in the relatively clean air at Ta' Gordan lighthouse, background particulate matter is possibly influenced more significantly by fireworks than in Malta where competing sources of these metals confound and hence reduce somewhat the degree of correlation.

There are other sources, besides fireworks, that contribute to different degrees towards the presence, in the atmospheric environment, of the five metals subject of this study. Thus, Al, here always found present in much higher abundance, is probably contributed mainly by geological materials (De Tomasi et al., 2003).

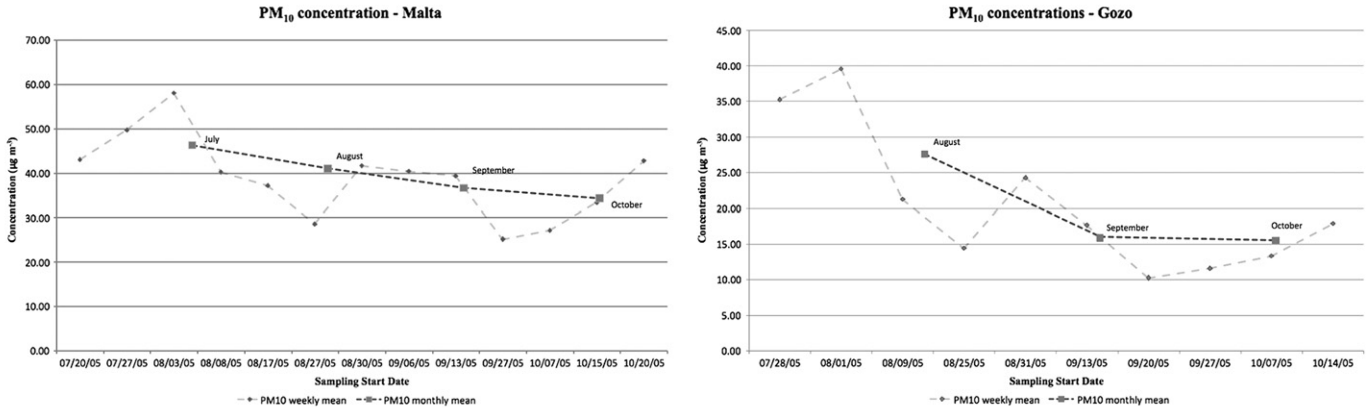

Fig. 2. Weekly mean concentrations of PM_{10} for Malta and Gozo and the associated monthly averages.

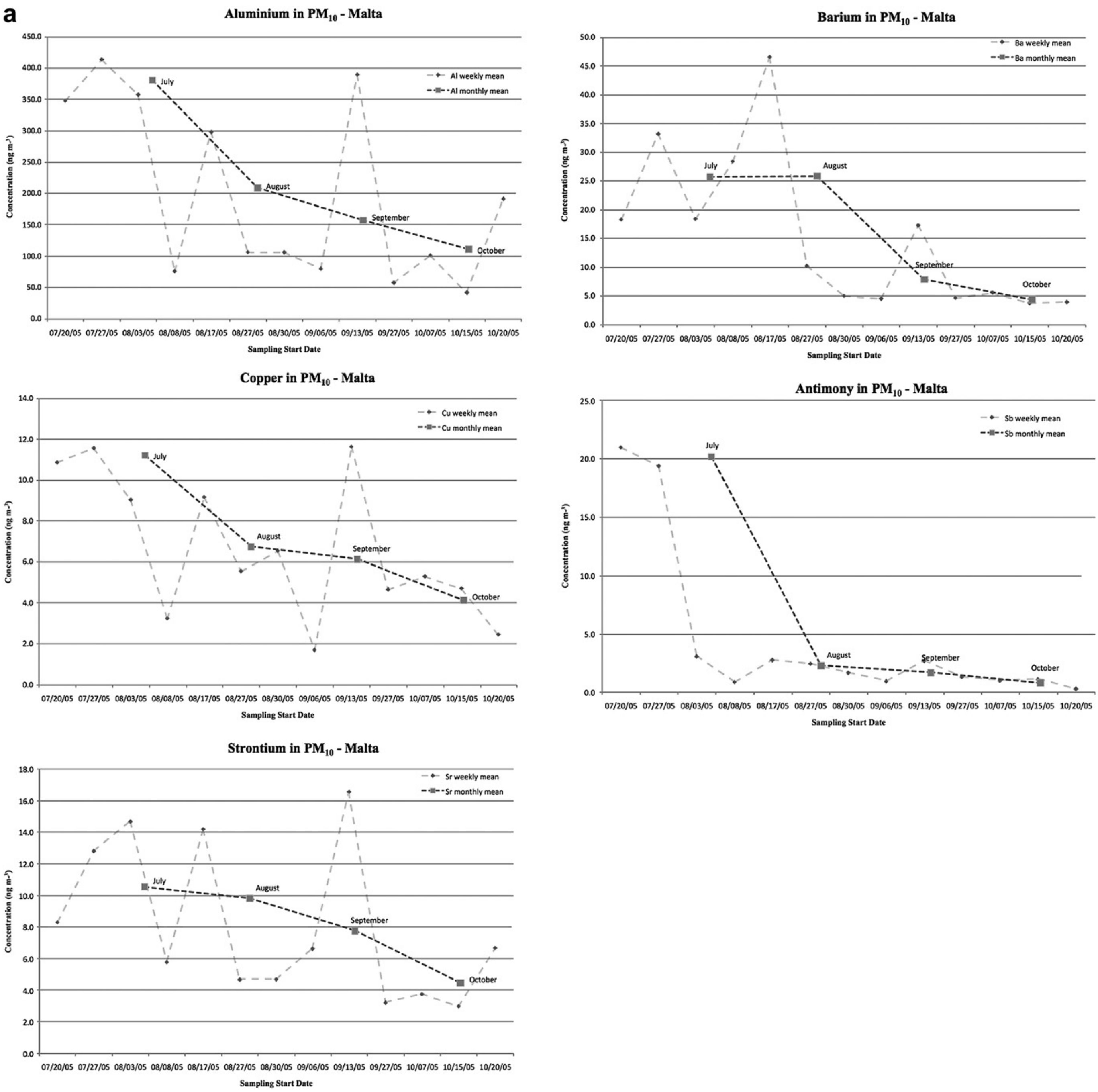

Fig. 3. (a) Weekly mean concentration of Al, Cu, Sr, Ba and Sb in PM_{10} dust in Malta and the associated monthly average concentrations. (b) Weekly mean concentration of Al, Cu, Sr, Ba and Sb in PM_{10} dust in Gozo and the associated monthly average concentrations.

The limestones of Malta contain Al in low abundance (Vella et al., 1997) although soils are likely to be richer and would, in any case, constitute a more important source of the metal than outcropping bedrock and the ubiquitous built environment which is exclusively in limestone (and concrete). Periodically, substantial reddish-brown dust carried by southern winds descends on Malta when PM_{10} levels exceed 90 $\mu g\ m^{-3}$ and reach much higher values (Camilleri, 2004): however, no such (easily visible) dust intrusions were observed and excessive PM_{10} values were not recorded during the sampling period so that we can conclude that the elevated Al concentrations measured during July–August 2005 were not caused by this factor. Al in Malta dust dropped by half from a monthly average of 266 $ng\ m^{-3}$ in July–August to 137 $ng\ m^{-3}$ in September–October and was similarly higher in Gozo during July–August compared to September–October. Al is a main fuel in pyrotechnics and is used in far greater quantity than any of the other four metals especially in flash crackers and maroons which dominate Maltese fireworks: this is confirmed by import trade statistics which show that about 16 000 kg Al powder were imported in 2006 while the combined quantity of the other metals (mainly as nitrates) was less than 2000 kg (NSO, Malta, 2007). Sr is also present in sedimentary rocks and soils and one may expect some contribution to airborne dust from such materials (De Tomasi et al., 2003). In addition, all five metals are also products of non-

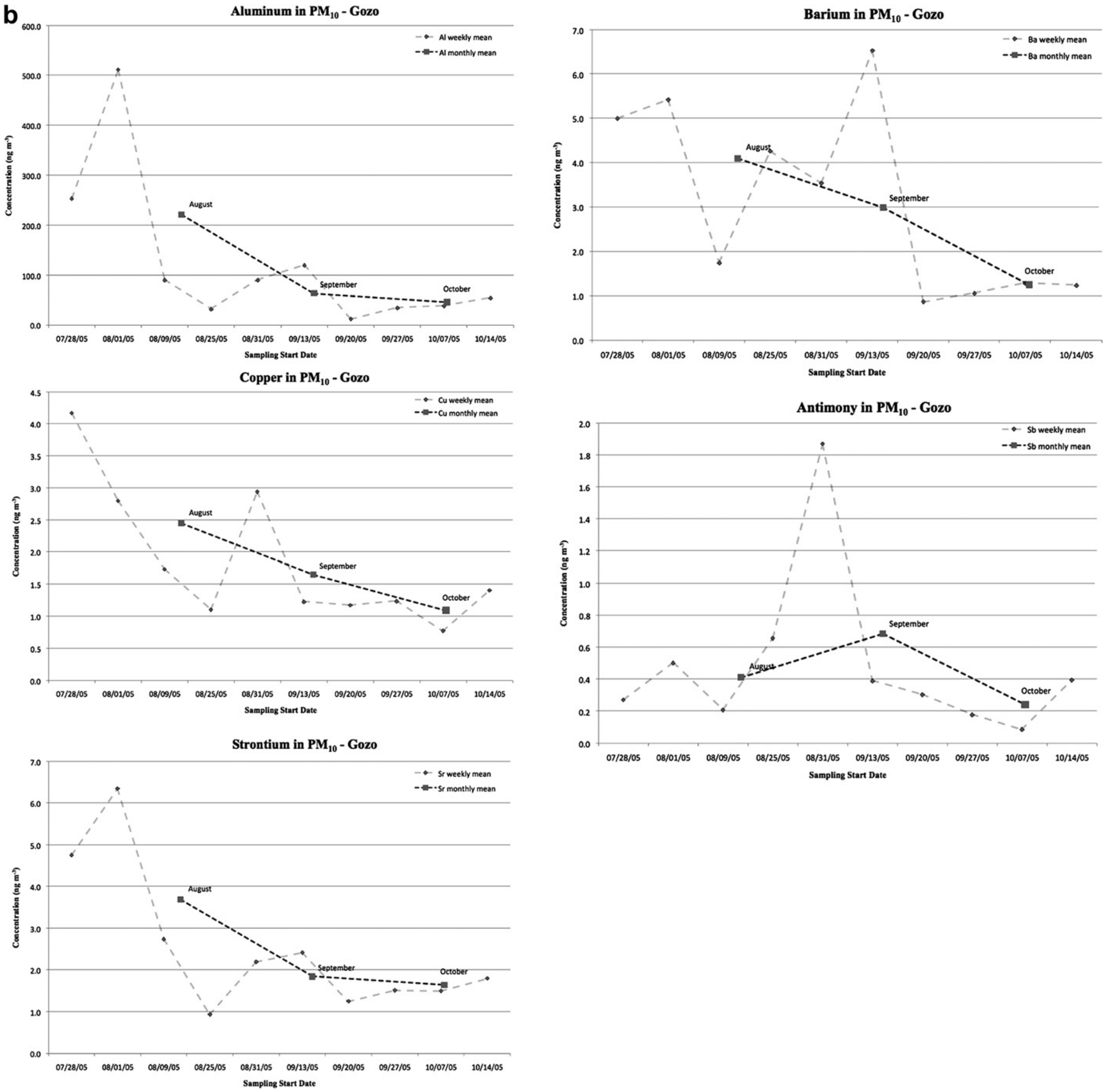

Fig. 3. (*continued*).

Table 2c
Mean daily mass concentration and chemical composition of PM_{10} for Cospicua, Malta.

Sampling date		28/4/06
Mean Daily PM_{10} ($\mu g\, m^{-3}$)		42.0
Mean daily concentration (ng m^{-3})	Cu	4.4
	Sr	4.8
	Ba	2.1
	Al	99.8
	Sb	0.8
Precipitation		–

exhaust emissions from motor traffic: these metals (and several others) are found in particles resulting mainly from brake linings wear, although tyre wear, road surface abrasion and resuspension of road dust are other sources of metallic emission related to motor traffic. This subject has recently been extensively reviewed by Thorpe and Harrison (2008) who state that it is generally accepted that a Cu/Sb mass concentration ratio of 5 in urban dust is indicative of brake-related particles, the value of this ratio for crustal material being 125. In a more recent study by Gietl et al. (2010), the Cu/Sb ratio for a London roadside dust was stated to be, on average, 9.1. The Cu/Sb ratios (and associated standard deviations) generated from our weekly mean values are as follows: for Malta dust, 0.56 ± 0.06 (July); 3.2 ± 0.6 (August), 3 ± 1 (September) and 6 ± 2

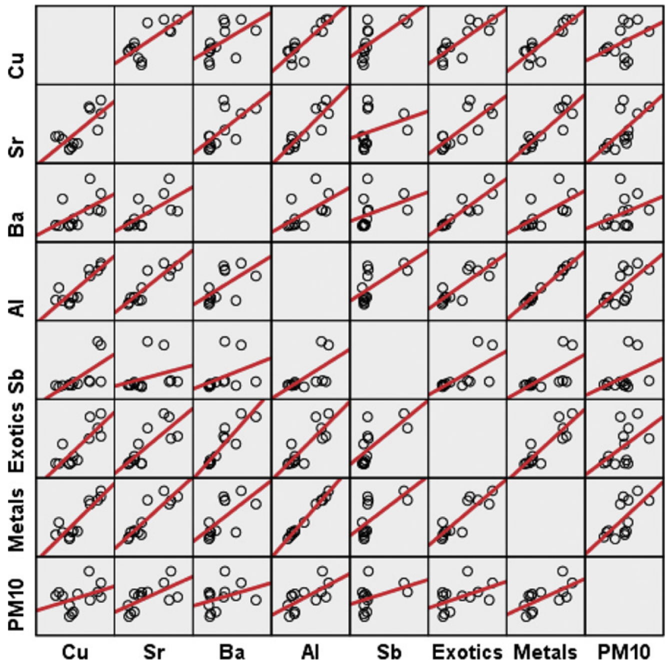

Fig. 4. Graphical correlation matrix for the metals and PM_{10} in Malta dust.

(October). Corresponding values for the Gozo site are: 14 (July, one week only); 4 ± 3 (August); 4 ± 1.5 (September) and 6 ± 3 (October). The low values obtained for Cu/Sb especially for July likely indicate that Zejtun dust collected during this time has a different signature from break liner wear being much richer in Sb: the values for August and September also remain somewhat richer in Sb and only in October does the ratio come closer to the value quoted by, for example, Hjortenkrans et al. (2007) for Stockholm dust (5.3). This value is almost identical to that for Malta dust collected in April 2006 (5.5) and these findings corroborate the conclusion that, during the summer *festa* period, the metallic content of Malta dust with respect to the five metals under study is indeed dominated not by motor traffic but more probably by fireworks.

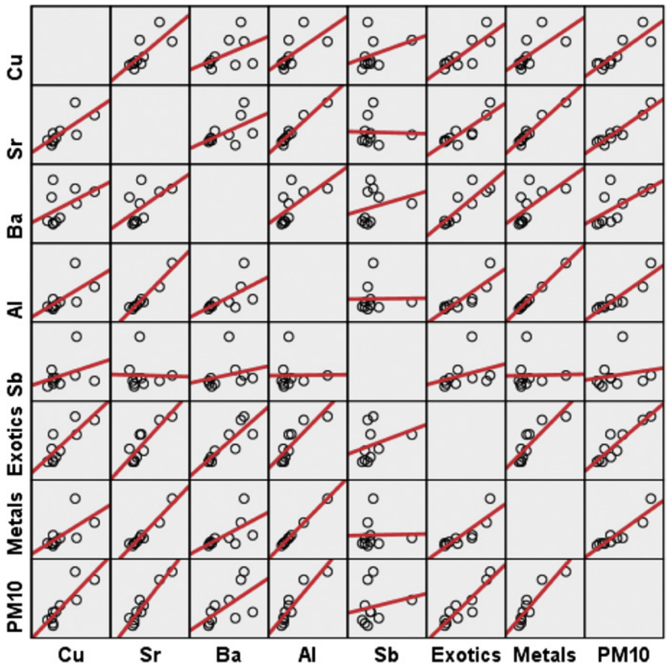

Fig. 5. Graphical correlation matrix for the metals and PM_{10} in Gozo dust.

The dust collected from the background station in Gozo during August and September also seems to have somewhat lower Cu/Sb except for the July value which is rather high. It is notable that in October, the mean Cu/Sb ratios for both the background and the 'target' Malta sites are the same and close to the value presumed to be typical of roadside material.

In the study by Gietl et al. cited earlier (2010), where the concentration of several metals were measured in dust collected from a very heavily-trafficked road in central London and a background urban site from the same city, it was found that the average and median values for Ba in the roadside air were respectively 17.9 and 15.6 ng m^{-3} and those for the background site were 3.90 and 3.65 ng m^{-3}. For Sb, the average and median values in roadside air were 6.73 and 6.24 ng m^{-3} while the background site yielded 1.31 and 1.49 ng m^{-3}. Our corresponding data for Ba for the Zejtun urban background site were 25.9 and 23.4 ng m^{-3} determined using the set of weekly means for July–August and 6.3 and 4.6 ng m^{-3} for September–October. For Sb the corresponding values were 8.2 and 2.9 ng m^{-3} for July–August and 1.3 and 1.1 ng m^{-3} for September–October. Clearly, our values for both metals for July–August are too high and cannot reasonably be ascribed to traffic especially given that Zejtun is an urban background site and traffic passing through this town is not expected to produce temporally-variable or seasonally-dependent emissions. The content of Ba in the single dust sample collected from Cospicua Malta in April (2.1 ng m^{-3}) is even lower than that found in the late summer and is typical of the London background values: this suggests that the effects of the fireworks on the quality of the air in Malta persist even during the latter part of the summer when the quantity of displays has subsided considerably and one questions whether it requires several months for the problem to resolve. During July–August, the ostensibly pristine Gozo rural site (Ba, ng m^{-3}: mean = 4.1, median 4.7) is as contaminated with Ba as the London urban background site and contamination with this metal is only slightly lower during the latter part of the summer (2.4; 1.3 ng m^{-3}). The situation for Sb is only slightly better for Gozo, means and median values for the two periods being (in ng m^{-3}) 0.43, 0.40 and 0.55, 0.35.

Perry (1999) had suggested that fireworks do not pose a significant health hazard as they occur infrequently and detonate at height where pollutants can be dispersed safely. Our findings challenge this conclusion and show that fireworks can indeed affect the air quality negatively so that in densely populated areas such as Malta, where the incidence of pyrotechnic activity is high, the potential for harm to human health probably cannot be ignored. Ba causes bronchoconstrictor effects and interferes with the heartbeat (Hicks et al., 1986; Reeves, 1979); in particulate matter, it is suspected to be responsible for the significant increase in asthma cases during the Fourth of July celebrations on Hawaii (Smith and Vu, 1975) and the Diwali festival in India (Murty, 2000). Ba derived from fireworks is expected to be particularly bioavailable since it consists of water soluble species such as $BaCl_2$, BaO, $Ba(OH)_2$ and residual $Ba(NO_3)_2$ (Steinhauser et al., 2008): in contrast, barite, the mineral in brake liners is insoluble. If, as appears likely, the increase in barium concentration in PM_{10} during the summer festival period is ascribable to fireworks, then the population appears to be exposed to a more harmful species form of Ba than that from road traffic and this exacerbates the contamination problem. Kennedy et al. (1998) have shown that Cu ions in PM_{10} may be responsible for the sensitivity to such particles of asthmatic individuals as established in epidemiologic studies. The presence of antimony in

Malta's inhalable dust may be the most problematic. It is known that Sb_2S_3 oxidises to Sb_2O_3 at 380 °C in air (Melcher and Faullant, 2000) and it is therefore highly likely that conversion of the sulfide used in pyrotechnics to the oxide occurs efficiently during fireworks burning; indeed even the antimony sulfide in brake pads is known to convert to the oxide on frictional heating when brakes are applied to release wear particles (von Uexküll et al., 2005). Although the presence of Sb in the atmosphere is generally attributed to emissions from motor traffic (Thorpe and Harrison, 2008), in light of our findings, it appears that in Malta during the whole summer period, the contribution from fireworks is predominant. The presence of Sb in PM_{10} is worrying even though the health effects of this metalloid are not entirely established, partly because the experimental and epidemiologic data are limited (Hayes, 1997) and also because Sb is frequently present with As so that environmental exposure is often due to both as co-contaminants (Gebel, 1997). Reviewing the available literature, Léonard and Gerber (1996) conclude that the carcinogenic (and mutagenic and teratogenic) risks of Sb are not very important but in a more recent review Cooper and Harrison (2009) are less optimistic and conclude that Sb has 'undoubtedly harmful effects on health and well-being'. Indeed, antimony trioxide (by inhalation) is currently classified as 'possibly carcinogenic to humans (Group 2B)' (IARC, 1989; OSHA, 2003) and the substance has actually been shown to cause lung cancer in female rats (Gebel, 1997).

4. Conclusion

The strong correlations between PM_{10} and Al, Ba, Cu, Sr and Sb in airborne dust, all of which materials are associated with pyrotechnic compositions, but also the magnitude of the concentrations of the metals in PM_{10} when compared with similar data from other countries leaves little doubt that *festa* firework displays are contributing significantly to the dust load and composition in Malta. In this small Mediterranean country, PM_{10} is an air quality issue where EU limit values are frequently exceeded; moreover, while other priority pollutants have been brought under control, the airborne dust situation is not improving (MEPA, 2008). It is concluded that fireworks are exerting a significant negative effect on air quality during the summer-long religious festival season and this factor appears to have been missed as an important and potentially controllable contributor to the PM_{10} problem. Moreover, the metals contributed by fireworks to inhalable dust, and in particular barium and antimony, may represent cause for concern in view of the special health risks posed by these substances.

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