Pace, C. and Vella, A. J. (2016). *Xjenza Online*, 4:4–10.

Xjenza Online - Journal of The Malta Chamber of Scientists
www.xjenza.org
DOI: 10.7423/XJENZA.2016.1.01

Research Article

Escalation of Fireworks in Malta: Environmental Forensic Evidence from Perchlorate in Dust Fall

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Abstract. Summer in Malta is accompanied by fireworks as part of the numerous (about 85) religious festivals (*festa*) that occur throughout the period. We attempted to establish whether firework activity is truly following official trade statistics, which imply a decreasing trend, or otherwise. Firework manufacture critically depends on the availability of oxidising agents, two of which (KNO_3, $KClO_3$) being controlled by permits but not potassium perchlorate ($KClO_4$) which is freely available. Recent changes in legislation have slightly decreased the quantity of $KClO_3$ and increased that of KNO_3 and according to official trade statistics, consumption of $KClO_4$ has decreased from 17 t/a in 2010 to $\approx$ 2.5 t/a during 2011–2014. However, from levels of perchlorate in dust fall and rate of total deposition we calculated the quantity of $KClO_4$ used during 2012 and obtained $\geq$ 90 t/a, 36 times the official figure. This situation is serious since the danger of accidental explosion during manufacture escalates as production intensifies. Also, contamination of the environment by firework-waste appears destined to remain high with possible consequent effects on human health.

Keywords: perchlorate, environmental forensics, Malta, fireworks, dust fall

1 Introduction

Fireworks in Malta are synonymous with summer since the three month long period between June and September is the time of celebration of scores of religious feasts (*festa*), each one of which is accompanied by the burning of abundant fireworks over several days per week. In addition, other events of importance and even an annual competitive fireworks festival are adding to the local firework activity. It is a well-established fact that the burning of fireworks lowers air quality and introduces into the air and other environmental phases compounds of toxic metals such as copper, barium, lead and antimony and other chemicals which are harmful to health. Thus, it is of interest to know whether local firework activity is not just 'traditionally intense' but actually rising, as the anecdotal evidence would suggest despite contrary indication from trade statistics.

Manufacture of fireworks is practically controlled by placing limits on the acquisition of oxidising agents, which are key to pyrotechnic compositions. The principal oxidising agents employed in Malta are potassium nitrate (KNO_3), potassium chlorate ($KClO_3$) and potassium perchlorate ($KClO_4$). Local legislation dating back to the early part of the twentieth century has imposed limits on the use of KNO_3 and $KClO_3$. Recently (2014), the limits have been modified by a slight lowering to 1000 kg per year for $KClO_3$ and an increase to 3200 kg per year for KNO_3. There are about 30 firework factories currently active in Malta. Potassium nitrate and chlorate can only be obtained from importers if the proper police permits are presented. On the other hand, perchlorate, imported as the potassium salt ($KClO_4$), has only become popular relatively recently and its acquisition is not limited. According to the National Statistics Office (NSO), imports of $KClO_4$ increased from 2300 kg in 2002 to 17 000 kg in 2010 and then suddenly and inexplicably fell and stabilized at about 2500 kg thereafter. Meanwhile, anecdotal observation of firework activity suggests that these displays have not decreased in number during the last five years or so. It is not known whether the chemical is being (legally) introduced directly into Malta from EU producers whose imports are not reported to the NSO.

The objective of this paper is to establish whether

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firework activity (manufacture and burning) is declining substantially, as the NSO data suggest, or whether the information from trade statistics is incorrect and pyrotechnic activity is actually escalating. Imported fireworks may also be contributing to this activity. This matter is of interest not only because firework burning exerts a negative impact on the quality of the environment (Shi, Zhang, Gao, Li & Cai, 2011; Munster, Hanson, Jackson & Rajagopalan, 2009; Shi et al., 2011; Camilleri & Vella, 2010), especially if it is intensive and prolonged in time, but also because it exposes the general public to greater personal risk associated with illegal manufacture, which takes place covertly in private residences and the legal and dangerous transportation through inhabited areas of significant loads of pyrotechnic articles on their way from factory to burning fields (Vella, Axiak, Delicata & Theuma, 2011).

We have recently shown (Vella, Chircop, Micallef & Pace, 2015) that perchlorate (ClO_4^-) is present in indoor dust in Malta where it appears to derive from the only source of contamination that is significant locally, namely, the burning of fireworks. Twenty eight of 37 indoor dust samples (76%) collected during the period 2011–2013 contained perchlorate at concentrations which ranged from 0.79 ppm to 53 ppm (median value 7.8 ppm). In a study by Wan et al. (2015) on perchlorate in indoor dust from twelve countries, including Greece, USA, Japan and China, it was found that perchlorate levels were typically less than 0.5 ppm and for China, the value was highest at 4.25 ppm: thus, Malta's indoor dust contains almost double the perchlorate in China's dust, making the local situation the worst case of contamination by this chemical reported so far. Perchlorate is also found in local atmospheric dust fall with concentrations that range from 0.52 ppm to 561 ppm (median concentration 6.2 ppm) (Vella et al., 2015). Most significantly, these levels peak during the summer and decrease again during the rest of the year, only to rise again during the next summer period. This temporal variation, which closely follows the firework activity on the island, leaves no doubt about the source of this otherwise rather exotic chemical. Worldwide, the principle use of perchlorate, mainly as the ammonium and potassium salts, is in the manufacture of solid rocket propellant and military explosives although it has other uses, e.g. in air bags, highway flares, tanning and some other manufacturing applications (Urbansky, 1998, 2002; Gu & Coates, 2006). In Malta, potassium perchlorate is imported exclusively for use as an oxidising agent for the manufacture of pyrotechnic compositions. Compositions made from mixtures of potassium perchlorate and aluminium or other metals (magnesium, titanium, magal) are far more stable than those containing potassium chlorate (locally known as "putassa") and for this reason, the latter compositions have recently (2014) been declared illegal in Malta.

The shift from the use of chlorate-metal mixtures towards perchlorate containing systems is expected to reduce the toll exacted by accidents during the manufacture of fireworks which claimed an average of 2.2 lives per annum in the period 1980–2010 (Axiak, Delicata, Theuma & Vella, 2012). On the other hand, the shift will have an impact on the quality of the environment, since firework displays release into the atmosphere unburnt solid residues that precipitate as dusts thus contaminating homes and soils. Dust is a significant source of human exposure to chemicals: the fine particulates present an inhalation risk while coarser material is a risk for ingestion (Christoforidis & Stamatis, 2009; Shi et al., 2011), especially for infants and children who are more prone to transfer matter from hand to mouth (Glorennec, Lucas, Mandin & Le Bot, 2012). Contaminated dust fall also has potential to affect water quality and that of locally grown agricultural produce (Smith, Yu, McMurry & Anderson, 2004; Shi et al., 2007), which become potential sources for consumers of these foods (Lee, Oh & Oh, 2012; Asami, Yoshida, Kosaka, Ohno & Matsui, 2013).

The concern over perchlorate relates to the chemical's known interference with thyroid function: perchlorate blocks the uptake of iodine into the thyroid thus reducing production of thyroxine and triiodothyronine (Wolff, 1998; Greer, Goodman, Pleus & Greer, 2002). Females appear to be more at risk than males, especially if their dietary iodine intake is limited (CDC (Centers for Disease Control and Prevention), 2006). A recent first study of its kind by Taylor et al. (2014), which considered the effects of perchlorate on pregnant women with hypothyroidism and iodine deficiency and the neurodevelopment of their offspring, has provided evidence to show that the chemical affects cognitive development so that their children had lower IQs when measured at three years of age.

In this paper, we attempt to use perchlorate in dust fall as a proxy for the determination of the quantity of fireworks burned in Malta using the rate of total dust fall and the concentration of perchlorate in this dust as the environmental forensic evidence.

2 Materials and Methods

2.1 Collection of field samples

Dust fall was collected from a total of 43 sites from both the main island of Malta and the smaller island of Gozo (Fig. 1), 38 sites of which were sampled during September and October 2011 and 17 sites during March, May, July, August, October and December 2012 and February 2013. Dust fall at each site was collected in four Beatson jars of 10 cm diameter which were exposed for

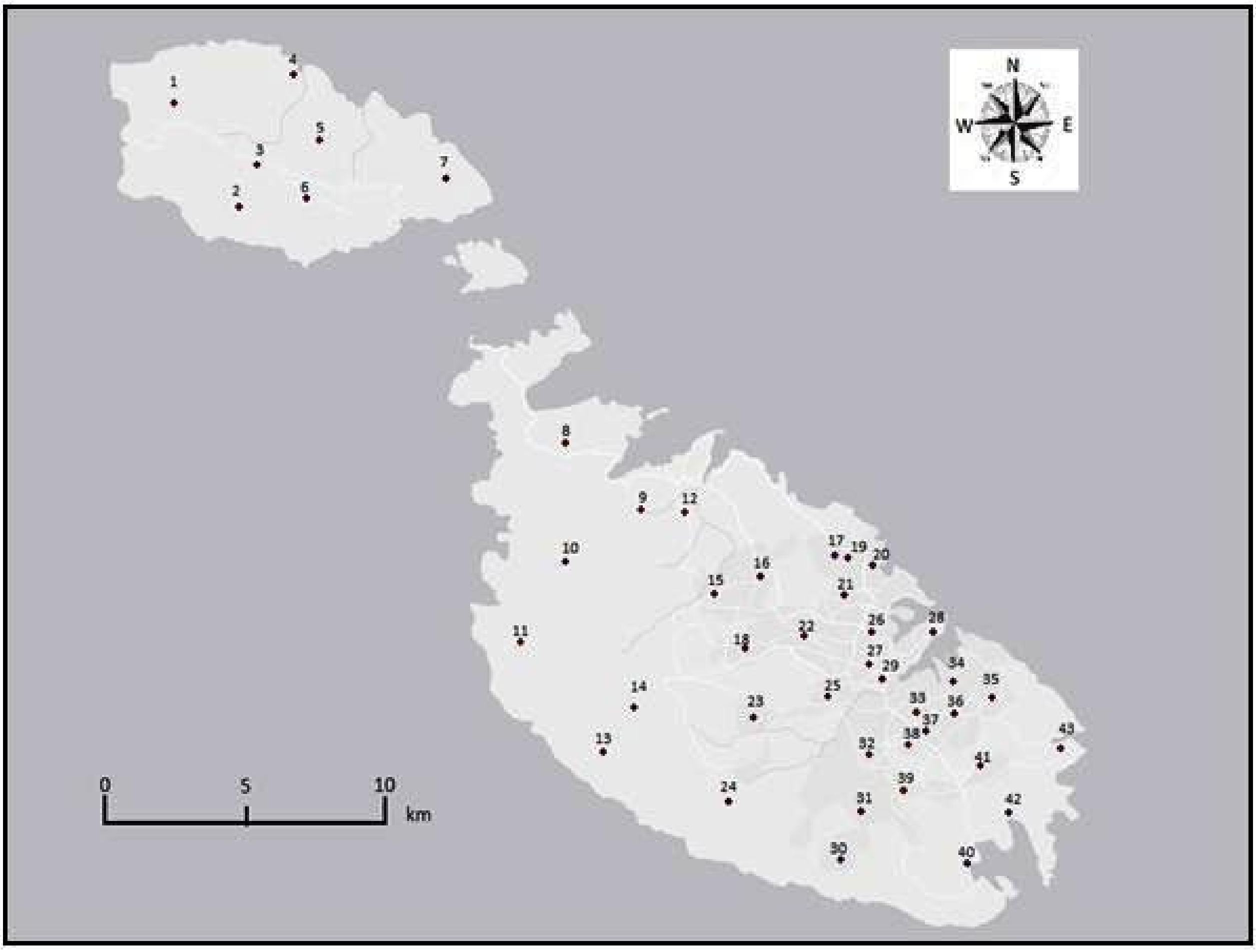

Figure 1: Map of the Maltese Islands showing localities from which dust fall was collected during September 2011 to February 2013.

about 30 days on rooftops of houses, away from walls. After collection, the jars were capped and stored at room temperature pending analysis.

The quantity of dust in each jar was determined by quantitative transfer into tared vials using deionised water which was then evaporated at 100 °C to constant mass. This allowed the rate of dust fall to be measured.

2.2 Measurement of perchlorate in dust fall

The method of analysis of perchlorate in the dust is described in detail elsewhere (Vella et al., 2015) and was based on extraction of the perchlorate in dust using deionised water and analysis of the extract by ion chromatography.

3 Results and Discussion

Fig. 2 shows the rate of dust fall in $mg\,m^{-2}\,d^{-1}$ calculated as monthly means for the period September 2011 to February 2013. It is clear that, for most of the time, Malta experiences considerable dust fall which actually well exceeds $200\,mg\,m^{-2}\,d^{-1}$, this value being the "limit of nuisance" suggested for the UK (Quality of Urban Air Research Group, 1996).

Several sources likely contribute to this coarse atmospheric dust and the major elements present in it (Ca, Si, Fe, Al) as reported by Vella et al. (2015) suggest that it is, in part, transboundary in nature and in part consists of re-suspended settled dust derived from soils and the local limestone terrain by attrition. Another strong local contributor is the quarrying and building industry (Vella & Camilleri, 2005).

The quantity of dust fall is much dependent on wind direction and rainfall: thus, for example, strong western and southern winds transport to Malta dust from the Sahara while rainfall effectively scavenges particulate matter and brings it to earth thus increasing the rate of deposition. For these reasons, the dry summer months July–August 2012 (precipitation in mm was 0.2 and 0.8 respectively) produced very low dust deposition during the period of measurement, although values were still above the mentioned nuisance value. The extremely high values registered during March 2012 and February 2013 were due to dust intrusions from the Sahara desert

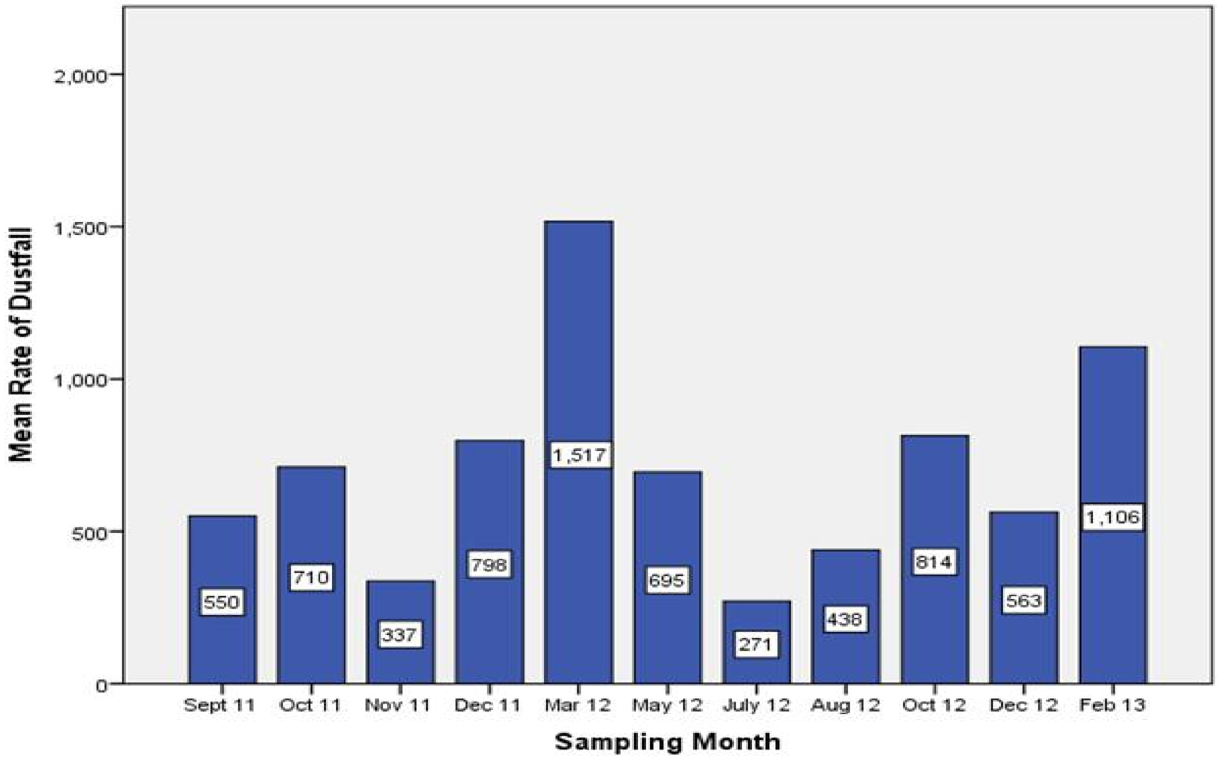

Figure 2: Mean dust fall deposition rates in $mg\,m^{-2}\,d^{-1}$ during September 2011 to February 2013.

which caused significant reductions in visibility according to the Meteorological Office.

We have shown (Vella et al., 2015) that perchlorate levels in dust reached maximum values of about 120 ppm (July 2012) and 40 ppm (August 2012) during the period corresponding to lowest rates of deposition of dust fall. Concentration values dropped to < 20 ppm in the other months and were lowest (< 5 ppm) during February (2013) and March (2012).

Using this information together with the total dust fall data, one could deduce the quantity of potassium perchlorate used in fireworks displays locally if one knew the emission factor for perchlorate of Malta-made fireworks and adopted a box model with specific assumptions regarding the mass balance.

There is a paucity of information with regards to emission factors for perchlorate from fireworks generally and there is none pertaining specifically to Malta manufacturers: so one is obliged to use the best data that is available. Armstrong, Ridley, Guilbeault and Duff (2009) established that for different pyrotechnic articles containing between 2.5 and 28.0% by mass perchlorate, combustion in a sealed container resulted in formation of between 0.001 to 0.017% of residual perchlorate, although the authors observed that these values could be underestimates for real pyrotechnic devices. In another study by Oxley et al. (2009), the emission factors for perchlorate in propellant and explosive articles containing 56.0% and 5.6% by mass perchlorate, respectively, were found to be 0.0022 and 0.0210%; in road flares containing 10% by mass perchlorate, the emission factor was significantly higher, namely 0.17%. In Malta, flash crackers typically contain 63% by mass perchlorate (Camilleri, 2008) but since no information is currently available for the emission factor for the local products, we assumed a probably-inflated value of 0.17%.

Aerial pyrotechnic activity in Malta takes place almost exclusively within the built zone at distances of about 200 m from inhabited areas. Ground displays often occur within town squares or in open spaces within the township. Since the built environment constitutes 22% of the total surface area ($316\,km^2$) of the Maltese Islands, we assumed that, to a first approximation which ignores losses to uninhabited and marine areas, the perchlorate-laden dusts from fireworks deposit equally over an area of about $69.5\,km^2$. Using official perchlorate consumption data for 2012 i.e. 2.5 t, an estimated 4.25 kg of $KClO_4$ or 3.03 kg perchlorate (ClO_4^-) is emitted as unreacted oxidant over a period of about 3 months (July to September), which gives an average predicted value of $0.485\,\mu g\,m^{-2}\,d^{-1}$. Since for July 2012, the average dust fall was $271\,mg\,m^{-2}\,d^{-1}$, the calculated mass fraction of perchlorate in the dust is 1.8×10^{-6}, corresponding to 1.8 ppm. Similarly, for August 2012, the rate of dust fall was $438\,\mathrm{mg\,m^{-2}\,d^{-1}}$ giving a predicted perchlorate concentration of 1.1 ppm.

The experimental mean perchlorate levels in dust for these two months were 120 ppm (July 2012) and 40 ppm (August 2012) which are 67 and 36 times larger, respectively, compared to the modelled values. Given the approximations in the calculation, the two values are reasonably close and clearly indicate that the real quantity of potassium perchlorate being released into the local environment is definitely much larger than the value reported to the National Office of Statistics. If we take, conservatively, the lower of the two estimates, then we conclude that the amount of potassium perchlorate imported annually for firework manufacture is most likely not less than 90 t.

If the value of the emission factor for Malta-made pyrotechnics for perchlorate is lower than the one assumed in this calculation and closer to the value determined by Armstrong et al. and Oxley et al., which pertained to pyrotechnic articles and not road flares, then the discrepancy between the modelled levels based on an import figure of 2.5 t and that actually measured in the dust would increase further. Moreover, even if the assumption that perchlorate is spread uniformly over about a quarter of the territory is relaxed to include the whole of the island's surface area (which is probably unreasonable), the resultant modelled import value (about 22 t) is still ten times larger than the official NSO record.

The fact that significantly more potassium perchlorate seems to be crossing jurisdictional borders than is apparently known to the authorities is of itself a concern, since the chemical is a precursor to powerful explosives and hence a useful commodity for terrorists. Importation of potassium perchlorate into Malta from European manufacturers does not, of course, require clearance by Customs although commercial trading involving the chemical has recently become subject to EU legislation. Regulation EU 98/2013 (OJEU (Official Journal of the European Union), 2013) requires specific official actions to be taken intended to keep tag of movements of (*inter alia*) potassium perchlorate between EU states or into the EU from third countries. This regulation exempts pyrotechnic articles.

Each firework factory is limited in the quantity of articles it can produce by the availability of oxidising agents: thus potassium nitrate regulates black powder production, which limits the quantity of lifting charges that can be manufactured and potassium chlorate, which is an ingredient in a number of manufactures. Both of these oxidising agents are controlled and each factory has access to 3200 kg (KNO_3) and 1000 kg ($KClO_3$) per annum of the chemicals. Given that there are about 30 factories in operation, it is likely that on average, each factory was employing about 3 t or more of additional oxidising agent over that function as provided by KNO_3 and $KClO_3$ and such would have given it potential to increase production by 170% over that without perchlorate use.

If perchlorate has indeed replaced chlorate completely in blasting mixtures, in response to the legal requirement, then the left-over but still utilized potassium chlorate is presumably being re-directed to other uses. Mixing potassium chlorate with charcoal produces a mixture (known as H3) which, due to its extreme sensitivity is illegal to produce but which serves to replace the safer but controlled black powder: this material is a critical component without which fireworks cannot be shot into the air and made to explode to form the familiar shapes associated with the art. If this is actually happening, then another factor is at play which increases the risk of accidental explosion during manufacture.

If utilization of perchlorate by the different factories is uneven, as is more likely, then the increase in consumption and hence in manufacture of articles by the bigger factories would be even larger than 170%. This surge in manufacture is not occurring in larger or improved amenities since fireworks factories have not upgraded or enlarged their facilities due to planning restrictions. This point is important in view of the fact that pyrotechnic manufacturing in Malta has had a rather poor safety record that has been principally attributed to human error, a factor aggravated by excessive and ever-rising production targets.

Apart from the effects on the pyrotechnic community itself resulting from excessive production, there are of course the equally serious effects on the general population. The presence of perchlorate in elevated concentrations in dust, including indoor dust, for at least 3 months per year is exposing the population to potential health risks from a contaminant with well-recognized toxic characteristics. The very young may be most at risk of exposure to perchlorate in dust in view of their propensity for ingestion of such matter. The presence of perchlorate in dust has potential to contaminate other environmental reservoirs such as drinking water and agricultural produce and this would serve to increase consumption of the chemical beyond that already likely present in several items of imported food (Lee et al., 2012).

Environmental pollution is often not easy to tackle when the causative agents are important due to their linkage to production and wealth creation or energy generation. Fireworks are a non-essential activity notwithstanding which however it has significant support from both the 1500-strong pyrotechnic community itself and a sub-set of the general population who fears any interference with the "right of its pastime ('*delizzju*')". The bulk of perchlorate is used for the making of maroons ('*tal-bomba*') and substantial reduction of these noise-making but otherwise colourless fireworks could go some way towards reducing contamination of the environment by perchlorate and other toxic wastes.

Could fireworks be made without the use of perchlorates? The problem is currently under investigation and compositions containing periodates (Moretti, Sabatini & Chen, 2012) and copper iodide (Klapötke, Rusan & Sabatini, 2014) and no $KClO_4$ have been found to perform well for certain applications. Also, organic high-energy materials such as 5-aminotetrazole in place of perchlorate also offer promise (Sabatini & Moretti, 2013). However, although future more environment-friendly ("greener") fireworks are likely to become technically possible, these will probably not be as cheap to manufacture as the chlorine-based pyrotechnics. Also, their adoption locally could present significant challenges to manufacturers, these being largely artisans with limited scientific training. Thus, to protect Malta's environment from further environmental contamination from fireworks, an immediate cap on the use of perchlorate is essential.

4 Conclusion

On the basis of the levels of perchlorate in dust fall and given that this chemical has no other local applications besides fireworks making and burning, a mass balance calculation shows that such manufacture is significantly higher than that which may be deduced from the official rate of importation of the precursors. Our results show that firework manufacture and burning has increased significantly since perchlorate use has become popular and even mandatory for the making of certain firework articles. It appears that factories may have increased their production by 170% over that based on the sole use of the controlled oxidising agents. Excessive manufacturing, coupled with the lack of proper amenity and possibly overcrowded work rooms could lead to accidents and thus obliterate any advantages provided by adoption of the safer chemical for the making of the fireworks. Reducing and limiting the use of perchlorate is the only solution for lowering the significant environmental impact from fireworks in this country which is needed to protect human health from possible deleterious effects from this chemical.

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