Air quality, health impacts and burden of disease due to air pollution (PM_{10}, $PM_{2.5}$, NO_2 and O_3): Application of AirQ+ model to the Camp de Tarragona County (Catalonia, Spain)

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ABSTRACT

The purpose of this study was to assess the impact to human health of air pollutants, through the integration of different technics: data statistics (spatial and temporal trends), population attributable fraction using AIRQ+ model developed by the WHO, and burden of disease using Disability-Adjusted Life Years (DALYs). The levels of SO_2, NO, NO_2, O_3, H_2S, benzene, PM_{10}, $PM_{2.5}$, CO, benzo(*a*)pyrene and metals, obtained between 2005 and 2017 from the air quality monitoring network across Camp de Tarragona County, were temporally and spatially determined. Health impacts were evaluated using the AIRQ+ model. Finally, the burden of disease was assessed through the calculation of Years of Lost life (YLL) and Years Lost due to Disability (YLD). In general terms, air quality was good according to European quality standards, but it did not fulfil the WHO guidelines, especially for O_3, PM_{10} and $PM_{2.5}$. Several decreasing (NO, NO_2, SO_2, PM_{10} and benzene) and an increasing (O_3) temporal trend were found. Correlation between unemployment rate and air pollutant levels were found, pointing that the economic crisis (2008-2014) was a factor influencing the air pollutant levels. Reduction of air pollutant levels ($PM_{2.5}$) to WHO guidelines in the Camp de Tarragona County would decrease the adult mortality between 23 and 297 cases per year, which means between 0.5 and 7% of all mortality in the area. In this County, for lung cancer, ischemic heart disease, stroke, and chronic obstructive pulmonary disease due to levels of $PM_{2.5}$ above the WHO threshold limits, DAYLs were 240 years. This means around 80 DALYs for 100,000 persons every year -between 2005 and 2017. Population attributable fraction (PAF) and burden of disease (DALYs) methodologies are suitable tools for regional and national policymakers, who must take decisions to prevent and to control air pollution and to analyse the cost-effectiveness of interventions.

Keywords: Air quality, AIRQ+ model, Population attributable fraction, burden of disease, DALYs, Camp de Tarragona County

Highlights

- Pollutants levels from an air quality network were temporally and spatially studied.
- Mortality and health effects due to pollutant levels above WHO standards were assessed.
- If $PM_{2.5}$ fulfil WHO limits, mortality would decrease between 23 and 297 per year.
- DAYLs due to pollutant levels above WHO standards were 80 years for 100,000 individuals.
- Integration of AirQ+ and DALYs is suitable to support decisions to reduce air pollution.

1. Introduction

Air pollution is one of the most challenging environmental problems, which must be faced at local, regional and global scale. Industrial activities, including energy production, road traffic, and household combustion, among others, have been identified as important emission sources of a wide range of pollutants (EEA, 2018; Prüss-Ustün et al., 2016). It is well known that air pollution is associated to adverse health effects such as respiratory and cardiovascular diseases, cancer and even death (Abdo et al., 2016; Ai et al. 2019; Brook, 2007; Chen et al., 2008, 2018; Demetriou et al., 2012; Farhat et al., 2011; Mafrici et al., 2008; Raaschou-Nielsen and Reynolds, 2006; Raaschou-Nielsen et al., 2013; Song et al., 2014; Sun et al., 2016; Sunyer, 2001; Wang et al. , 2019; Wu et al., 2019; Zhang et al. 2020). According to the World Health Organization (WHO), in 2012, 3.0 million of deaths were attributable to outdoor air pollution, while around 6.5 million of deaths (11.6% all world deaths) would attributable to indoor and outdoor air pollution, if they were taken into account together (WHO, 2016a). In a recent study (Stanaway et al. 2018), the attributable number of deaths to air pollution (PM and O_3), for both indoor and outdoor environments, was set at 4.9 million in 2017.

To support decisions of policymakers, it is necessary to quantify the effects of air pollution and the potential changes due to mitigation policies implementation. To measure health outcomes due to air pollution exposure, WHO developed the AIRQ+ model. This model has been applied internationally in Europe, but also worldwide (Al-Hemoud et al., 2018). AIRQ+ measures the contribution of air pollution to the mortality/morbidity using the population attributable fraction (PAF). PAF is the proportional reduction in a population morbidity or mortality, which would occur if exposure to air pollution was reduced to an alternative exposure scenario (WHO, 2016b), e.g. WHO air quality threshold limits (annual mean limits 10 and 20 µg/m^3 for $PM_{2.5}$ and PM_{10}, respectively (WHO, 2006)). As complementary tool, environmental burden of disease using disability-adjusted life years (DALYs) can assess the health impact (mortality and morbidity) that may be attributed to environmental factors such as air pollution (Prüss-Ustün et al., 2016). The DALYs, as a way to compare overall health and life expectancy of different populations, take into account the years of life lost due to a premature death (YLL), as well as the years of healthy life lost due to a disease (YLD) (Devleesschauwer et al., 2014).

The current study has been focused on the Camp de Tarragona County (Catalonia, NE Spain), where since 1960s one of the most important petrochemical industrial complex of south-Europe is placed. In addition to the chemical and petrochemical companies (oil refinery, chlor-alkaly plant, plastic manufacturers, and chemical industries), a hazardous and a municipal solid waste incinerator (HWI and MSWI), an important industrial harbour, as well as a commercial airport, are also present in the same area. Because of the considerable number of potentially contaminant activities, the public concern for the air quality in the zone has promoted the constitution of the “Air quality panel of the Camp de Tarragona” by the Parliament of Catalonia, which is aimed at improving the air quality in the area and at assessing human health effects associated to it (TQACT, 2019).

The aim of present study was to assess the impact to human health of air pollutants in the Camp de Tarragona County between 2005 and 2017, through the integration of different technics (data statistics, population attributable fraction and burden of disease. To achieve this goal, the levels of a number of pollutants (SO_2, NO, NO_2 O_3, H_2S, benzene, PM_{10}, $PM_{2.5}$ and CO), measured by an air quality monitoring network across the Camp de Tarragona County (from 2005 to 2017), were spatially and temporally analysed. In addition, the health impact of some pollutants (NO_2, O_3, PM_{10} and $PM_{2.5}$) was evaluated using the AirQ+ model version 1.3, developed by the WHO (2016b). Population attributable fraction (PAF) of mortality due to pollutant (PM_{10}, $PM_{2.5}$, NO_2 and O_3) levels above the WHO thresholds was also estimated. Finally, the results obtained by PAF methodology were used to evaluate the burden of disease (DALYs) attributable to air pollution (PM_{10}, $PM_{2.5}$, NO_2 and O_3), above the WHO threshold scenario, for the population living in the Camp de Tarragona County.

2. Materials and methods

2.1. Area of study

The Camp de Tarragona County (Fig. 1) is subdivided into three counties: Tarragonès, Alt Camp, and Baix Camp (South of Catalonia, NE Spain). There are two main cities in the area: Reus (103,615 inhabitants in 2017) and Tarragona (131,094 inhabitants in 2017) (IDESCAT, 2019). The total population in this region ranged from 406,000 inhabitants in 2005 to 483,000 inhabitants in 2017 spread in 2703 km^2 (IDESCAT, 2019). The largest petrochemical complex of southern Europe is located in this zone since 1960s. The petrochemical complex is divided in two sub-complexes, the north (petrochemical industries) complex and the south (chemical industries) complex. In addition, there are other potential pollutant activities in the area: a MSWI, a HWI, an industrial harbour, and a commercial airport. Obviously, there are also other diffuse emission sources, such as heating systems, agriculture burning, and a heavy traffic. Climate in the region is typical of Mediterranean basin. The annual mean temperature (2017) was 16°C, with a minimum and maximum temperature of -7°C and 36°C, respectively, and a total accumulated precipitation of 340 mm, more than 70% during the periods of March-April and September-October (Meteocat, 2019). Two different wind regimes are present in the area. One of them with predominant winds blowing from north, following Francolí River valley and affecting Alcover, Perafort, Sant Salvador, Constantí, Tarragona and Laboral stations, and the second one with predominant winds blowing from North-west, affecting Reus and Vilaseca stations (Meteocat, 2019). Moreover, winds regime blowing from south, from the sea, is important in the area during the summer.

2.2. Levels of pollutants

The levels of SO_2, NO, NO_2, O_3, H_2S, benzene, PM_{10}, $PM_{2.5}$, As, metals (Cd, Ni and Pb), benzo[*a*]pyrene and CO were obtained from the Network of Vigilance and Forecast of the Quality of the Air (XVPCA) of the Autonomous Government of Catalonia (Generalitat de Catalunya). Data are available online (AIR QUAILTY, 2019). Table 1 summarizes the main characteristics of each air quality monitoring station. According to the XPVCA (XVPCA, 2019), the sampling and determination of pollutants were carried out following these standards: EN14212 for SO_2; EN14211 for NO and NO_x; EN12341 for PM_{10}; EN14907 for $PM_{2.5}$; EN14626 for CO; EN14625 for O_3; EN14622 for benzene; EN12341, EN14902 and EN15852 for metals (As, Cd, Ni and Pb), and EN12341 and UNE15549 for benzo[*a*]pyrene. Daily and hourly mean levels of pollutants were obtained from January 1, 2005 to December 31, 2017, from the nine stations placed in the Camp de Tarragona (Alcover, Sant Salvador neighbourhood, Perafort, Bonavista neighbourhood, Constantí, Vilaseca, Reus, Tarragona and Laboral educational campus). The location and the pollutants analysed in each station are shown in Table 1 and Fig. 1.

2.3. *Health risk assessment of air pollution (Population attributable fraction).*

Health effects due to exposure to air pollution were quantified using the AirQ+ model (version 1.3) developed by the WHO Regional Office for Europe. AirQ+ model was developed to calculate the magnitude of the burden and impacts of air pollution on health in a certain population. Health effects considered in the current study have been the following: long-term mortality (due to all natural causes), chronic obstructive pulmonary disease (COPD), lung cancer (LC), ischemic heart disease (IHD) and stroke. The model performs calculations that allow quantification of the health effects due to exposure to air pollution. The model is based on the attributable proportion (AP) defined as the portion of a health effect attributable to exposure to air pollution by a population (equation 1):

$$AP = \Sigma((RR-1)\cdot P)/ \Sigma(RR\cdot P) \qquad \text{(equation 1).}$$

where RR is the relative risk for a given health endpoint in a determined exposure to an air pollutant, being P the fraction of population under exposure to the air pollutant.

The amount of health effect attributable to the exposure (IE), and the number of cases attributable to the exposure (NE), can be calculated by means of equations 2 and 3, respectively:

$$IE = I\cdot AP \qquad \text{(equation 2).}$$

$$NE = IE\cdot N_p \qquad \text{(equation 3).}$$

where I is the baseline frequency of health effect in the population under study, while N_p is the size of the population. The parameters used for calculations are summarized in Table 2.

Air pollutants reductions from 2005-2017 levels to the ideal scenario “WHO threshold scenario” were considered. WHO threshold scenario consists in O_3 levels lower than 100 μg/m^3 in 8-hourly mean, and annual means of 20 and 10 μg/m^3 for PM_{10} and $PM_{2.5}$, respectively. For NO_2, a mean annual value of 20 μg/m^3 was taken into account following the recommendations of the HRAPIE project (WHO, 2013). Population data from the Camp de Tarragona -grouped by age- for the period 2005-2017 were obtained from official statistics of Catalonia (IDESCAT, 2019). The incidence of mortality was obtained from the Health Department of Catalonia (Departament de Salut, 2019). Mortality incidence due to all natural causes and for each disease (chronic obstructive pulmonary disease (COPD), mortality due to lung cancer (LC), mortality due to ischemic heart diseases (IHD), and mortality due to strokes) were obtained from the Catalan Health System Observatory (CHSO, 2019). Similar levels of NO_2 and O_3 were found in urban and industrial areas. Health impacts were calculated in urban/industrial areas, using a mean value of the air quality stations. Health outcomes were also calculated in rural areas for O_3, but not for NO_2, because its levels were below the threshold limit (20 µg/m^3). Regarding $PM_{2.5}$, it was only measured in urban and industrial sites, showing similar concentrations. As the levels of $PM_{2.5}$ -from 2005 to 2007- were not measured by air quality stations, they were calculated by multiplying PM_{10} levels and $PM_{2.5}/PM_{10}$ ratio, which was previously set at 0.60. NO_2 concentrations were used to analyse the attribution factor in the mortality due to natural causes, while O_3 concentrations were used to analyse the attribution factor in the mortality because of respiratory diseases. $PM_{2.5}$ levels were used to analyse the attribution factor in mortality due to all natural causes (for adults above 30-years old), mortality due to chronic obstructive pulmonary disease (COPD), mortality due to lung cancer (LC), mortality due to ischemic heart diseases (IHD), and mortality due to strokes. To assess human health impacts using AIRQ+ model, a ratio of 0.60 was used in order to convert PM_{10} levels to $PM_{2.5}$ at the stations where data were lacking

2.4. Burden of disease (DALYs).

Burden of disease was assessed using Disability-Adjusted Life Years (DALYs), which are defined as the loss of healthy life years (Devleesschauwer et al., 2014). They are calculated as the sum of the Years of Life Lost (YLL) due to premature deaths, and the Years Lost due to Disability (YLD) for people living with the health consequences of the respective disease (equations 4-6).

$$DALYs=YLL+YLD \quad \text{(equation 4)}$$

$$YLL=N_d \cdot L \quad \text{(equation 5)}$$

$$YLD=Ic \cdot DW \cdot D \quad \text{(equation 6)}$$

where N_d is the number of deaths, L is the standard life expectancy at the age of death in years, Ic is the number of incident cases, DW is the disability weight factor, which indicates the severity of the disease from 0 (perfect health) to 1 (death) (WHO, 2019), and D is the average duration of the case until remission or death (years). Since no information about adverse health outcomes onset and their duration was found, mean age of diagnosis, and survival rate, were obtained from the scientific literature (Almagro et al., 2002; Chirlaque et al., 2018; Clua-Espuny, 2014; Miravitlles et al., 2009; Moran et al. 2014; Salmerón et al. 2012; Taylor et al., 2019). These data were used according to the following origin criteria: Catalan population, Spanish population, and West-Europe population.

2.5. Statistics

The statistical software package XLSTAT (Version 2015.2.02.18681) and IBM SPSS statistics were used to perform the statistical analysis of data. To evaluate significant differences between groups, a Levene test was performed to evaluate variances. ANOVA (parametric data) or Kruskal-Wallis (non-parametric data) test were subsequently applied. Significance level was set at $p<0.05$. Correlations between variables were assessed using the Pearson's coefficient. Principal components analyses (PCA) with Varimax rotation were applied to reduce the number of variables and to facilitate the patterns recognition. Agglomerative hierarchical clustering (dendrograms) were performed with the Ward method using dissimilarities (Euclidian distances). Annual mean levels were used to determine temporal trends with Mann-Kendall test at significance level of 0.05.

3. Results

3.1. Levels of pollutants

The levels of SO_2, NO, NO_2 O_3, H_2S, benzene, PM_{10}, $PM_{2.5}$ and CO registered in the nine stations located in the Camp de Tarragona from 2005 to 2017 are shown in Table 2 and Fig. 2. Annual air mean pollutants levels trends at a significance level of 0.05 were summarized at Table S1 (Supplementary materials).

Mean annual levels of SO_2 in the nine stations, presented a clear decrease tendency ($p<0.05$) from 2005 (4.9 µg/m^3) to 2017 (2.8 µg/m^3). A similar significant decreasing trend ($p<0.05$) was observed for the four areas: urban, rural, and north complex, meanwhile no significant ($p>0.05$) trends were noted in the South complex. However, between 2015 and 2017 a slight increase was noticed in the north and south industrial complexes. European air quality standards set a SO_2 daily average limit of 125 µg/m^3, a limit that cannot be reached more than 3 times/year (European Union Parlament and Council, 2008). In the Camp de Tarragona and for the period studied, there were between 2-5 exceedances between 2010 and 2017 (especially in the Constantí station), and up to 8 exceedances in the period 2005-2007. WHO air quality Guidelines for SO_2 establishes a daily mean of 20 µg/m^3 (WHO, 2006). In recent years (2012-2017), the WHO guideline was exceeded up to 1 time (except for the Alcover station -placed in a rural area-, where it was exceeded 6 times in 2016). In contrast, between 2005 and 2009, it was exceeded up to 20 times/year, mainly in the urban, north and south complexes.

Regarding NO_2, annual mean levels measured in all stations showed a decrease trend ($p<0.05$) from 23.5 µg/m^3 in 2005, to 18.3 µg/m^3 in 2017. Similar significant ($p<0.05$) decreasing trends were found for the four areas. However, higher levels were detected in the urban and south complex (more affected by urban environment) than the north complex. The rural area presented the lowest NO_2 levels. A similar trend than that of NO_2 was also observed for NO, with a significant decreasing trend ($p<0.05$) from 7.3 µg/m^3 in 2005 to 4.9 µg/m^3 in 2017. The same decreasing trends were found in all the areas, with the exception of the rural zones. where no significant ($p>0.05$) temporal trend was observed. Similar differences than those for NO_2 were observed for the four areas (Urban>South complex>North complex>Rural). European and WHO air quality standards set NO_2 annual and daily mean limits of 40 μg/m^3 and 200 μg/m^3, respectively (European Union Parlament and Council, 2008; WHO, 2006). For the period evaluated, the annual limit was not reached in any station of the Camp de Tarragona. In turn, the daily limit was reached few times (1-3) per year in the South complex (Bonavista and Laboral stations), but it never surpassed the 18 exceedances/year allowed by the legislation.

Ozone (O_3) was analysed only in five stations (Alcover, Constantí, Vilaseca, Reus and Tarragona). Ozone presented a significant ($p<0.05$) increasing trend over the years from 53.6 μg/m^3 in 2005 to 59.8 μg/m^3 in 2017. This increasing trend was also significant ($p<0.05$) for rural, urban and North complex, but did not reach the level of significance ($p>0.05$) in the South complex between 2005 and 2007. Rural area (Alcover station) showed higher O_3 levels than those found in the rest of stations (Constantí, Vilaseca, Reus and Tarragona), which showed similar levels among them. According to the European legislation, the maximum daily 8-hours mean (120 μg/m^3) was exceeded between 16 and 20 times in the Alcover station, and below 8 in the other four stations. However, this number of exceedances does not surpass the 25 exceedances allowed by the EU air quality standards (European Union Parliament and Council, 2002). Taking into account WHO (2006) guidelines, the 8-hour limit (100 μg/m^3) was reached 25 times in Alcover.

For H_2S, no significant ($p>0.05$) temporal trends were observed. No spatial differences were noticed despite the high annual levels registered in 2007 and 2009 in the North complex. The concentrations were between 1 and 2 μg/m^3. Nowadays, there is no European or WHO directives that establish maximum concentrations for this pollutant. However, Spanish legislation (Real Decreto 102, 2011) set at 40 μg/m^3 daily limit, and 100 μg/m^3 half-hourly limit. Some exceedance in half-hourly limit, especially in Constantí, Perafort and Laboral stations- before 2011- were noticed. However, in general terms these limits were accomplished.

Benzene levels did not show significant trends ($p>0.05$) along all period, with the exception of the North complex (Constantí), where a significant ($p<0.05$) decreasing trend was found between 2005 and 2017. Between 2005 and 2010, in the Constantí station, benzene registered higher levels ($p<0.05$) (between 3.7 and 6.2 μg/m^3) than those during in the period 2011-2017. Without considering this period and station, benzene levels were far below the air quality standard annual limit set in 5 μg/m^3 (European Union Parlament and Council, 2008).

Regarding PM_{10}, a clear decrease was observed between 2005 (46.5 μg/m^3) and 2013 (16.9 μg/m^3), followed by periods with constant concentrations: 19.1-23.4 μg/m^3. PM_{10} presented a significant ($p<0.05$) decreasing trend between 2005 and 2017, if all stations were considered and for each area (rural, urban, North and South complex). No differences ($p>0.05$) were noticed in PM_{10} levels regarding the location of each station. $PM_{2.5}$ levels were measured since 2008. Mean annual $PM_{2.5}$ concentrations did not show a significant ($p>0.05$) trend, ranging from 10.7 to 11.8 μg/m^3 between 2008 and 2017, respectively. In both, North and South complex, similar levels ($p>0.05$), and no significant ($p<0.05$) trends, were observed. The annual $PM_{2.5}/PM_{10}$ ratio was similar in all stations and ranged from 0.40 to 0.71. Mean ratio for the 2008-2017 period, when both PM were analysed, was set between 0.58 and 0.64, depending on the station. Regarding European air quality standard, annual PM_{10} limit and daily limit are 40 and 50 μg/m^3, respectively. The daily limit cannot be exceeded more than 35 times/year (European Union Parlament and Council, 2008). Several stations exceeded air quality standard annual limits for PM_{10} in 2005 and 2006. Thus, Vilaseca station in 2015, and Reus and Constantí stations in 2006, exceeded more than 35 times the daily limit (50 μg/m^3). Otherwise, the WHO established air quality guidelines for annual mean PM_{10} levels of 20 μg/m^3 (WHO, 2006). This level was exceeded in almost all stations in the Camp de Tarragona. For $PM_{2.5}$, European air quality standard is set at annual mean levels of 25 μg/m^3 (European Union Parlament and Council, 2008), while the WHO air quality guideline established annual and daily mean levels of 10 and 25 μg/m^3, respectively (WHO, 2006). Similarly, to PM_{10}, $PM_{2.5}$ concentrations in the Camp de Tarragona were also below the European standard, but they did not fulfil the WHO guidelines for annual and daily mean levels.

Only three stations (Alcover, Reus and Tarragona) measured CO for all the studied period (2005-2017). No significant ($p>0.05$) temporal trends were noted despite that the mean levels decreased from 0.4 mg/m^3 in 2005 to 0.2 mg/m^3 in 2017. Similar levels were found in these three stations. No exceedances of the 8-hourly limits (10 mg/m^3) were detected between 2005 and 2017 in these three stations (European Union Parlament and Council, 2008).

In addition to these pollutants, the concentrations of benzo(*a*)pyrene were analysed discontinuously (14% of days) in one station (Constantí) since 2008, while metals (As, Cd, Ni and Pb) were analysed discontinuously (14% of days) in Vilaseca, Reus and Constantí stations. Benzo(*a*)pyrene registered an annual mean value between 0.12 and 0.18 ng/m^3, which is below the annual mean limit of 1 ng/m^3 established by the European legislation. Nowadays, the WHO has not set air quality guidelines for benzo(*a*)pyrene. Regarding metals, in Constantí and Vilaseca stations, the mean annual levels in the period 2005-2017 ranged between 0.5 and 2.1 ng/m^3 for As, 0.1 to 3.1 ng/m^3 for Cd, 2.4 to 7.4 ng/m^3 for Ni, and 2.4 to 12.4 ng/m^3 for Pb. According to the European legislation (European Union Parlament and Council, 2008, 2004), the annual mean levels of these metals cannot exceed 500 ng/m^3, 20 ng/m^3, 5 ng/m^3 and 6 ng/m^3 for Pb, Ni, Cd, and As, respectively. As for benzo(*a*)pyrene, metal levels were below the European annual threshold, taken into account that they were analysed only 14% of days.

High positive Pearson's correlation values were found between NO and NO_2 (0.820; $p<0.01$) and between NO_2 and NO, and PM_{10} (0.551 and 0.768, respectively, both at $p<0.01$), while there were and negative correlations between NO and O_3 (-0.616; $p<0.01$) and NO_2 and O_3 (-0.788; $p<0.01$) (Table 3). In addition, moderate but significant ($p<0.01$) negative correlations were found between O_3 and PM_{10}. Finally, moderate although significant ($p<0.01$) positive correlations were found between nitrogen oxides (NO and NO_2) and particulate matter (PM_{10}).

Figure 3 depicts principal components analysis (PCA) and dendrograms of 2005-2017 annual mean levels of pollutants measured in the Camp de Tarragona stations. As not all pollutants were measured in all stations, therefore, three PCA and dendrograms were carried out to cover all the stations and most analysed pollutants. $PM_{2.5}$ was not included because it was analysed only in few stations, showing a high linear dependency with PM_{10}. Moreover, CO was not taken into account because of similar levels in the three stations measured. Three analysis (PCA plus dendrogram) were done. The first one (Fig. 3a) included the 9 stations and takes into account NO_2, NO and H_2S. The second one (Fig. 3b), with 6 stations, includes NO_2, NO, SO_2, H_2S and PM_{10}. Finally, the last one (Fig. 3c), considers NO_2, NO, SO_2, H_2S and O_3 in 5 stations.

3.2. Attributable proportion and burden of disease (DALYs).

Population attributable fraction (PAF) methodology was applied to the Camp de Tarragona between 2005 and 2017, using the AirQ+ model.

Figure 4a depicts cases of mortality due all natural causes attributable to air pollutants ($PM_{2.5}$, NO_2 and O_3) above the WHO threshold scenario in the Camp de Tarragona. Excess cases of mortality due $PM_{2.5}$ exposure ranged from 297 (with a 95% confidence interval between 196 and 389 (196, 389; CI95%)) in 2005, to 23 (15, 30; CI95%) in 2016. This corresponds to 7% in 2005 and 0.5% in 2016 of total mortality cases in the area. In the period 2008-2017, when $PM_{2.5}$ levels were directly analysed and not estimated from PM_{10}, excess attributable mortality cases ranged from 23 (15, 30; CI95%) to 137 (90, 181; CI95%) in 2016 and 2015, respectively. Regarding NO_2, in urban and industrial areas, the excess of mortality cases ranged from 71 (33, 108 CI95%) in 2005 to 1 (1, 1; CI95%) in 2017. No cases were noticed in 2013, 2014 and 2016, because environmental levels were below the cut off limit (20 $\mu g/m^3$). The proportion of attributable cases to NO_2 levels above 20 $\mu g/m^3$ was 1.4%. The proportion of attributable cases NO_2 levels was only calculated in urban areas cause in rural areas annual mean levels were below WHO threshold limit (20 $\mu g/m^3$). Finally, for O_3, attributable excess mortality due to respiratory diseases cases were almost constant between 6 (2, 10; CI95%) and 9 (3, 15; CI95%) in urban and rural areas, respectively. More than 80% of attributable mortality cases due to O_3 belongs to the urban area. Despite in rural areas the O_3 levels were higher than those in urban and industrial areas, only around 10% of population of the Camp de Tarragona is living in rural areas. However, the percentages of attributable cases of mortality due to respiratory causes because of the concentrations of O_3 -above the WHO guidelines- were around 1.75% in urban areas, and 3.70% in rural areas. From 2005 to 2007, $PM_{2.5}$ were not directly measured, but calculated from PM_{10}. For this reason, we have considered only data of $PM_{2.5}$ from the last ten years (2008-2017). Mortality cases in adults older than 30 years, attributable to levels above the annual WHO threshold (10 $\mu g/m^3$, ranged from 23 (15, 30; CI95%) to 137 (90, 181; CI95%), which means percentages of 0.5 and 2.85% of mortality by natural causes. Figure 4b depicts the number of mortality excess cases due COPD, LC, IHD and stroke attributable to air pollution ($PM_{2.5}$) above the WHO threshold. In the last ten years (2008-2017), the annual mean excess cases of mortality were 4 (2, 6; CI95%) due to COPD; 2 (1, 4; CI95%) due to LC; 5 (3, 10; CI95%) due to IHD, and 1 (1, 2; CI95%) due stroke.

Based on all natural causes mortality above 30-years old, and assuming a mean mortality age of 56 year (mean between 30 and life expectancy in the region, 82), the YLL in the Camp de Tarragona due to $PM_{2.5}$ above the WHO threshold scenario, was between 7842 (5178, 10274; CI95%) years in

2005, and 570 (372, 755; CI95%) years in 2016. Consequently, mean YLL per 100,000 individuals was 823 years (541, 1084; CI95%) for the 2005-2017 period, and 483 years (316, 638; CI95%) for the 2008-2017 period, when the concentrations of $PM_{2.5}$ were measured. NO_2 levels above the WHO threshold supposes a mean of 163 YLL/100,000 individuals (77, 251; CI95%) in urban areas of the Camp de Tarragona County for the 2005-2017 period. Regarding O_3, the mean YLL per 100,000 individuals due to levels above the WHO threshold scenario was 40 (14, 68; CI95%) for the 2005-2017 period. Pollutant (NO_2, $PM_{2.5}$ and O_3) levels above the WHO threshold resulted in an YLL mean value of 686 (407, 953; CI95%) years/100,000 subjects for the 2008-2017 period. It should be noticed that summing the mortality caused by these air pollutants could induce to a double counting because some pollutants here considered are usually highly correlated. Using the mortality cause for each disease obtained from the AIRQ+ model, as well as survival rates and age of diagnosis/occurrence found in the literature, YLD were calculated. In the Camp de Tarragona, mean lost due to disability (YLD) were 19.7 years/100,000 individuals (10.7, 32.5; CI95%), or 60.2 years for all the Camp de Tarragona population (32.6, 99.5; CI95%) in the 2005-2017 period, for LC, IHD, stroke and COPD. Taken into account both results, YLL and YLD, DALYs in the Camp de Tarragona for lung cancer, ischemic heart disease (IHD), stroke and chronic obstructive pulmonary disease (COPD), due to $PM_{2.5}$ levels above the WHO threshold limits, was 240, with 25% coming from YLL. It means around 80 DALYs for 100,000 subjects (78 years/100,000 subjects) with minimum and maximum values of 16 years/100,000 subjects in 2016, and 264 years/100,000 subjects in 2005, respectively.

4. Discussion

4.1. Levels of pollutants

Based on the levels of the pollutants (NO_2, NO, H_2S, PM_{10}, $PM_{2.5}$, CO, benzene, metals and benzo[*a* pyrene]) analysed in the Camp de Tarragona, the air quality is good according to the European thresholds. However, it did not fulfil the WHO guidelines, especially for O_3 in rural area, PM_{10} and $PM_{2.5}$. The high O_3 levels registered in Alcover (rural area) are from local origin, due to the orography of the zone (Jiménez and Baldasano, 2004). Typical winds in the area, from sea to land, move O_3 or its precursors (NO_2 and VOCs), produced in urban areas (Tarragona) and in the petrochemical complex. They could have been accumulated in the Alcover area because of the presence of northern mountains. For both PM_{10} and $PM_{2.5}$, in general terms, EU standard were accomplished with few exceptions, by contrast, WHO air quality guidelines were not fulfilled in almost all stations.

WHO air quality guidelines and EU air quality standard have different roles. WHO air quality guidelines are based on scientific evidence to protect human health. However, EU standards can have legal consequences once implemented if they are not accomplished. Moreover, EU air quality standards take into account not only scientific evidences, but also economic, technical, and political aspects (Schneider et al., 2004). For the purpose of the current study, WHO air quality guidelines were taken into account and as commented previously, O_3 in rural area, PM_{10} and $PM_{2.5}$ surpassed the WHO guidelines continuously between 2005-2017 in the Camp de Tarragona County.

$PM_{10}/PM_{2.5}$ ratios, ranging from 0.58 to 0.64, were similar to those proposed by the WHO (2013) ($PM_{2.5}/PM_{10}$=0.65), being equivalent to those found in the literature for the Mediterranean European region (CAFE, 2004; Contini et al., 2014; Ostro, 2004; Querol et al., 2004, 2008). Consequently, as previously mentioned in the Materials and Methods section, to assess human health impacts using AIRQ+ model, a ratio of 0.60 was used in order to convert PM_{10} levels to $PM_{2.5}$ at the stations where data were lacking.

It should be taken into account that between 2005 and 2017 several significant decreasing trends were found especially for nitrogen oxides, SO_2, PM_{10}, and benzene, meanwhile an increasing trend was registered for O_3. Similar trends were registered in other European studies (Borge et al. 2019; Fenech et al. 2020; Font et al. 2019).

As expected, positive correlations between PM_{10} and $PM_{2.5}$ and between nitrogen oxides (NO_2 and NO) were found sources. Correlations between nitrogen oxides (NO and NO_2) and particulate matter (PM_{10}) were probably due to the common sources of these pollutants: traffic, heating systems, industries, and other combustion processes (Núñez-Alonso et al. 2019). However a negative correlation were found between O_3 and other pollutants (NO, NO_2 and PM_{10}). As suggested by some authors (Jia et al., 2017; Li et al., 2011), particulate matter absorb solar radiation, and in consequence, O_3 formation is reduced. Negative correlations between nitrogen oxides (NO and NO_2) and O_3 have been reported in previous studies (Beckerman et al., 2008; Fecht et al., 2016; Kumar et al., 2015). Nitrogen oxides, O_3 and VOC cycles during the day, nights, as well as differences in weekend and weekday, would explain these correlations (Alghamdi et al., 2014; Han et al., 2011).

The period of time (2005-2017) here studied includes the years of recent economic crisis, which in Catalonia meant an increase of the unemployment rate from 8.9% in 2008 to 16.2% in 2009, reaching a maximum of 23.1% in 2013. After that, the unemployment rate decreased to 13.4% in 2017. It is a clear indicator of the economic activity in the Camp de Tarragona, being related with the air quality. For this reason, Pearson's correlation was performed between annual mean values of air pollutants and unemployed rate as an indicator of the economic crisis (Table S2). Unemployment rate correlated negatively and significantly ($p<0.05$) with almost all pollutants NO_2 (-0.627), NO (-0.850), SO_2 (-0.760), PM_{10} (-0.811) and benzene (-0.617), but positively with O_3 (0.824). Decreased trends were observed in PM_{10}, NO_2 (especially in urban areas), NO, SO_2, H_2S (in north complex) and benzene (in north complex). These decreasing trends started in 2009, just when the economic crisis started in Spain. Some of these levels increased again in 2014, when the economic crisis began to remit. Similar trends and impacts of economic crisis in air quality and air pollutants emission were found in European countries and US (Monteiro et al. 2018, Pacca et al. 2020; Squizzato et al. 2019; Tong et al. 2016, Tzima et al. 2018).

Regarding spatial analysis (Figure3), Vilaseca and Reus stations, affected from wind blowing NO-SE direction, appear close together in PCA and dendrograms (Figures 3b and 3c). These two stations were impacted by the urban environment and were less affected by South and North industrial complexes because of prevalent wind directions. Tarragona, an urban station, was represented alone (Figures 3a, 3b and 3c) and showed a high correlation with NO_2. The Laboral and Bonavista stations appeared together since they are placed together and influenced by the South complex, urban background and the harbour (Figures 3a and 3b). Perafort and Alcover stations, both rural, are influenced by the industrial activities and appeared close together (Fig. 3c), with a negative correlation with nitrogen oxides. Alcover station appeared highly correlated with O_3 (Fig. 3a). As above commented, Alcover is an O_3 hotspot due to prevalent winds and orography. Finally, Constantí station did not show similarities with the rest of stations. Constantí station showed a high positive correlation with SO_2 and H_2S (Fig 3). This station is placed in the Francolí River valley, which is highly affected by the North complex, the MSWI, Tarragona city and Harbour, since prevalent winds blow from North and South.

In addition to pollutant previously reported, benzo(*a*)pyrene, the only polycyclic aromatic hydrocarbon (PAH) analysed, was measured only in Constantí station each year during the 14% of days. Similarly, metal (As, Cd, Ni and Pb) levels, were only analysed in three stations (Reus, Constantí and Vilaseca) at 15% of the days/year. In turn, other pollutants such as VOCs, (i.e., 1,3-butadiene, acrylonitrile and vinyl chloride), metals (V), or other PAHs, characteristics of industrial sources present in Camp de Tarragona (Chen et al., 2016; Domínguez-Morueco et al. 2015; Kampeerawipakorn et al., 2017; Kwon et al., 2016; Visschedijk et al., 2013) are not measured by the air quality network. However, there are previous data and monitoring studies in the area, which are focused on specific pollutants such PAHs and VOCs (Domínguez-Morueco et al., 2019, 2017; Ramírez et al., 2012, 2011), or on specific contaminants (PCDD/Fs, PCBs, metals) and industrial sources such as HWI (Bordonaba et al., 2011; Mari et al., 2013, 2014; Nadal et al., 2009a; Vilavert et al., 2010), MSWI (Vilavert et al., 2012, 2014, 2015) and chemical and petrochemical complexes (Domínguez-Morueco et al., 2017, 2019; Gallego et al. 2018; Nadal et al., 2009b, 2011; Ras et al., 2009). Based on these results, specific pollutants, such as metals, COVs and PAHs, which are representative of industrial activities in the area, should be measured with a higher frequency in order to improve the Camp de Tarragona air quality monitoring network.

4.2. Attributable proportion and burden of disease (DALYs).

In the present study, mortality due all-natural causes attributable to $PM_{2.5}$ above the WHO threshold scenario in the Camp de Tarragona ranged from 6.9 to 41.7 cases/100,000 individuals. These results were higher than in other Mediterranean cities such as Perpignan, Marseille, Nice and Livorno (3.78, 7.07, 9.71, and, 10.67 cases/100,000 individuals, respectively), but similar or even lower than those found in other Mediterranean cities such as Naples or Rome (55.20 and 34.75 cases/100,000 individuals, respectively) (Sicard et al. 2019). However, for mortality due to respiratory diseases attributable to O_3 above the WHO threshold scenario, the same Mediterranean cities (ranging from 1.77 to 5.17 cases/100,000 individuals for Rome and Livorno, respectively) showed slightly higher values than those of the current study (range 1.34 to 1.84 cases/100,000 individuals) (Sicard et al. 2019).

Results for YLL (823, 163 and 40 YLL/100,000 individuals for $PM_{2.5}$, NO_2 and O_3, respectively) are in agreement with the European air quality report that, for Spain, estimated 658, 209 and 43 YLL/100,000 individuals due to $PM_{2.5}$, NO_2 and O_3, respectively (EEA, 2018).

Due to the lack of data for the region, to calculate YLD, attributable to air pollutant levels above the WHO guidelines for the population of the Camp de Tarragona, was not possible. However, to calculate the DAYLs (YLL+YLD) of COPD, LC, IHD and stroke due to air pollutant levels above WHO guidelines (Fig. 5), several approaches were done (survival rate, average age disease detection, and time with the diseases were obtained from the scientific literature) (Almagro et al., 2002; Chirlaque et al., 2018; Clua-Espuny, 2014; Miravitlles et al., 2009; Moran et al. 2014; Salmerón et al. 2012; Taylor et al., 2019). In the Camp de Tarragona, for lung cancer, ischemic heart disease (IHD), stroke and chronic obstructive pulmonary disease (COPD) due to $PM_{2.5}$ levels above the WHO threshold limits, DAYLs was 240 years, with a 25% coming from YLL. This means around 80 DALYs for 100,000 individuals (78 years/100,000 individuals) annually between 2005 and 2017. According to the WHO (2016c), in Spain in 2012, DALYS from due ambient air pollution was set in 286 years/100,00 individuals, with uncertainty intervals (97.5% CI) between 53 and 469 years/100,00 individuals. The results of present study, even in the low range of results reported by the WHO, are of the same order of magnitude.

This study present some limitations, for example, acute health effects for O_3 or PM outbreak episodes were not taken into account. In the present investigation, only health effects of chronic exposure to air pollutant levels were assessed. It is well known that health effects due to acute exposure are not negligible in terms of health outcomes (Kloog et al. 2012), which should be taken into account in future research. However, the lack of health outcomes data with enough temporal resolution makes impossible to assess acute health effects to air pollutants peak levels. It should be taken into account that PM shape and chemical components differs in each location and period, and consequently, health effects may be different (Sicard et al. 2019). In addition, concentrations were measured in air quality monitoring stations for all the territory, and they did not take into account the real pollutant levels indoor (where usually people expend more time), or other microenvironments. The use and integration of agent based modelling, remote sensing, and personal sensors could help to solve these limitations (Reis et al., 2015). Finally, individual differences among subgroups, such as elderly, children or chronic patients, are an important issue to take into account and it should be also considered in further studies.

5. Conclusions

In general terms, according to the pollutant levels analysed by air quality network in the Camp de Tarragona County, the air quality was good based on the European air quality standards. However, it did not fulfil the WHO guidelines, especially for O_3, PM_{10} and $PM_{2.5}$. An economic indicator such as the unemployment rate significantly correlates (negatively) with several air pollutant (NO_2, NO, SO_2 and PM_{10}) levels that remark the influence of the economic crisis (2009-2014) in air pollutant levels. Reduction of air pollutants levels ($PM_{2.5}$) in the Camp de Tarragona to WHO guidelines should reduce the adult mortality between 23 and 297 cases per year. It means between 0.5 and 7% of all mortality in the area. Lack of data (metals, PAHs and VOCs), or incomplete data (benzo(*a*)pyrene and metals were only analysed in 14% and 15% of the days, respectively) are the main weak points of the air quality network in the Camp de Tarragona. Moreover, specific pollutants such as metals, VOCs and PAHs, which are representative of industrial activities in the zone, should be more frequently measured to improve the air quality monitoring network. The integration of population attributable fraction (PAF) and burden of disease (DALYs) methodologies, as well as statistical tools to find temporal and spatial trends, is a suitable approach to assess the impact to human health. This approach should help to regional and national policymakers, who must take/support decisions to prevent and control air pollution and to analyse the cost-effectiveness of interventions.

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Table1. Air quality monitoring stations in the Camp de Tarragona

Station	Coordinates UTM 31 North	Altitude (m)	Pollutants analysed	Area
Sant Salvador	352405, 4558155	57	SO_2, NO, NO_2, H_2S	North complex
Perafort	352230, 4561950	97	SO_2, NO, NO2, H_2S, Bz	Rural
Bonavista	348300, 4553400	39	SO_2, NO, NO2, H_2S, PM_{10}	South complex
Constantí	350550, 4557690	56	SO_2, NO, NO_2, H_2S, O_3, Bz, PM_{10}, $PM_{2.5}$, metals, B[*a*]p	North complex
Vilaseca	344920, 4553050	41	SO_2, NO, NO_2, H_2S, O_3, PM_{10}, metals	South complex
Reus	342355, 4557402	102	SO_2[a], NO, NO_2, H_2S, O_3, PM_{10}, CO, metals[b]	Urban
Tarragona	352474, 4553482	13	SO_2, NO, NO_2, H_2S, O_3, Bz, PM_{10}, CO	Urban
Laboral	349010, 4552030	5	SO_2, NO, NO_2, H_2S, Bz, PM_{10}, $PM_{2.5}$	South complex
Alcover	347665 , 4571462	238	SO_2, NO, NO_2, H_2S, O_3, CO	Rural

Bz: Benzene; B[*a*]p: Benzo(*a*)pyrene, Metals: As, Cd, Ni, and Pb.
[a] Data available from 2005 to 2012; [b] Data available from 2005 to 2015

Table 2. Yearly means (standard deviations) of pollutants in the Camp de Tarragona from 2005 to 2017 (AIRQUALITY, 2016) and European air quality standards (European Union Parlament and Council, 2008) and WHO guidelines (WHO, 2006).

		NO_2	**NO**	**SO_2**	**PM_{10}**	**$PM_{2.5}$**	**O_3**	**H_2S**	**CO**	**Bz**
2005		23.5 (6.5)	7.3 (3.7)	4.9 (1.7)	46.5 (7.8)	NA	53.6 (6.4)	1.4 (0.5)	0.4 (0.0)	1.1 (0.1)
2006		21.0 (5.7)	7.0 (2.5)	3.7 (1.0)	38.5 (3.5)	NA	55.0 (7.3)	1.6 (0.4)	0.3 (0.1)	1.5 (0.4)
2007		21.6 (6.6)	6.5 (3.1)	4.7 (1.7)	32.5 (2.1)	NA	54.6 (7.7)	1.7 (0.6)	0.3 (0.1)	1.4 (0.6)
2008		20.1 (5.6)	6.4 (2.6)	3.2 (1.2)	29.3 (3.1)	10.7 (2.4)	53.8 (2.5)	1.6 (0.4)	0.3 (0.1)	1.3 (0.5)
2009		21.6 (5.8)	5.8 (2.6)	4.0 (1.7)	27.4 (1.2)	12.5 (0.7)	55.9 (5.9)	1.7 (0.8)	0.3 (0.1)	1.1 (0.4)
2010		21.3 (6.1)	5.3 (2.4)	3.1 (1.5)	22.0 (1.8)	12.2 (2.5)	58.7 (6.2)	1.6 (0.3)	0.3 (0.1)	1.1 (0.6)
2011		20.2 (5.2)	5.4 (2.5)	3.7 (1.5)	25.5 (1.3)	13.9 (1.9)	57.1 (7.0)	1.4 (0.3)	0.3 (0.1)	1.3 (0.4)
2012		19.1 (5.6)	5.1 (2.2)	2.4 (0.8)	22.4 (2.6)	14.0 (2.5)	58.9 (6.6)	1.4 (0.3)	0.3 (0.0)	1.0 (0.4)
2013		17.8 (4.6)	4.9 (2.0)	2.2 (0.5)	16.9 (2.3)	10.8 (1.6)	61.7 (6.8)	1.4 (0.3)	0.3 (0.0)	1.2 (0.5)
2014		18.0 (4.3)	5.0 (1.7)	2.3 (0.5)	19.4 (3.1)	11.3 (0.5)	58.0 (6.7)	1.4 (0.3)	0.2 (0.0)	1.2 (0.4)
2015		19.5 (4.9)	5.7 (2.0)	2.8 (1.1)	23.4 (2.0)	14.7 (1.2)	58.3 (7.0)	1.5 (0.3)	0.2 (0.0)	1.2 (0.4)
2016		17.9 (4.7)	5.3 (1.8)	3.0 (1.1)	19.1 (2.8)	10.8 (0.6)	57.5 (6.8)	1.6 (0.3)	0.3 (0.0)	1.1 (0.2)
2017		18.3 (5.1)	4.9 (1.7)	2.8 (0.9)	20.7 (1.4)	11.8 (0.9)	59.8 (6.8)	1.7 (0.3)	0.2 (0.0)	1.2 (0.4)
WHO guidelines	**annual**	40	-	-	20	10		-	-	-
	24-hour	200[a]	-	20	50	25	100[b]	-	10[b]	-
EU standards	**annual**	40	-	-	40	25		-	-	5
	24-hour	200[a]	-	125	50	-	120[b]	-	10[b]	-

Bz: Benzene; NA: Not Available. [a] 1 hour mean; [b] 8 hour-mean. Levels of NO_2, NO, SO_2, PM_{10}, $PM_{2.5}$, O_3, H_2S and benzene in µg/m^3. Levels of CO in mg/m^3.

Table 3. Pearson's correlations of annual mean pollutants analysed in all stations of air quality network.

	NO_2	**NO**	**SO_2**	**PM_{10}**	**$PM_{2.5}$**	**O_3**	**H_2S**	**CO**	**Bz**
NO_2	1.000	0.820**	-0.132	0.551**	0.063	-0.788**	0.128	0.289*	0.169
NO		1.000	-0.034	0.768**	-0.024	-0.616**	-0.053	0.395*	-0.038
SO_2			1.000	0.281	-0.112	0.084	0.171	0.289	0.257
PM_{10}				1.000	0.431*	-0.340**	0.040	-0.118	0.395
$PM_{2.5}$					1.000	0.130	-0.018	0.024	-0.114
O_3						1.000	-0.264*	-0.477**	-0.033
H_2S							1.000	-0.073	0.343*
CO								1.000	-0.187
Benzene									1.000

Bz: Benzene; * Significant correlation at $p<0.05$, **Significant correlation at $p<0.01$

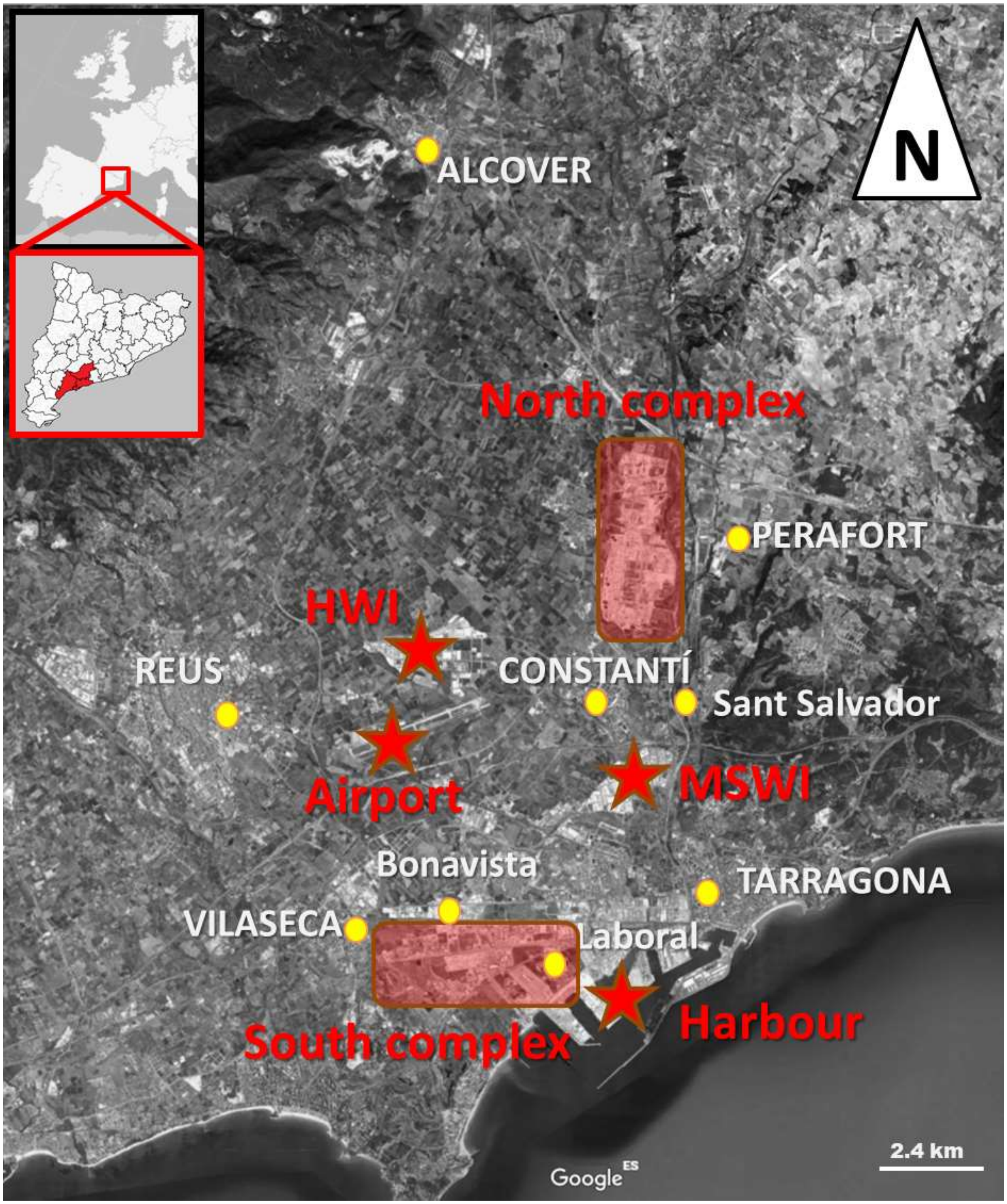

Fig. 1. Camp de Tarragona map with emission sources marked in red and air quality network stations marked in yellow circles. Principal municipalities are in capital letters.

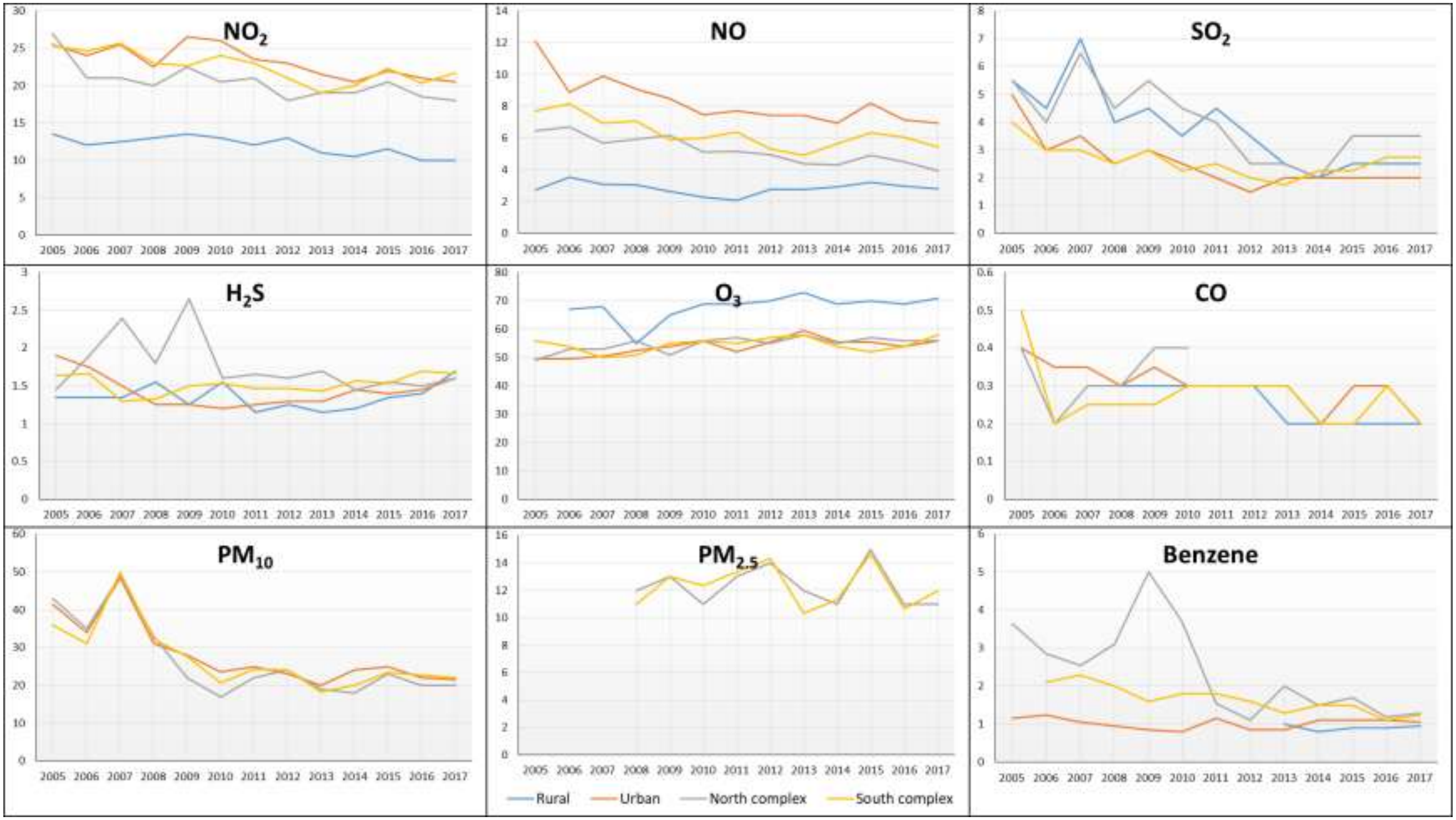

Fig. 2. Mean annual levels between 2005 and 2017 in the Camp de Tarragona of air pollutants measured in 9 stations clustered in four areas (rural, urban, north and south industrial complex). Levels of SO_2, NO, NO_2, O_3, H_2S, Benzene, PM_{10} and $PM_{2.5}$ in µg/m^3. Levels of CO in mg/m^3.

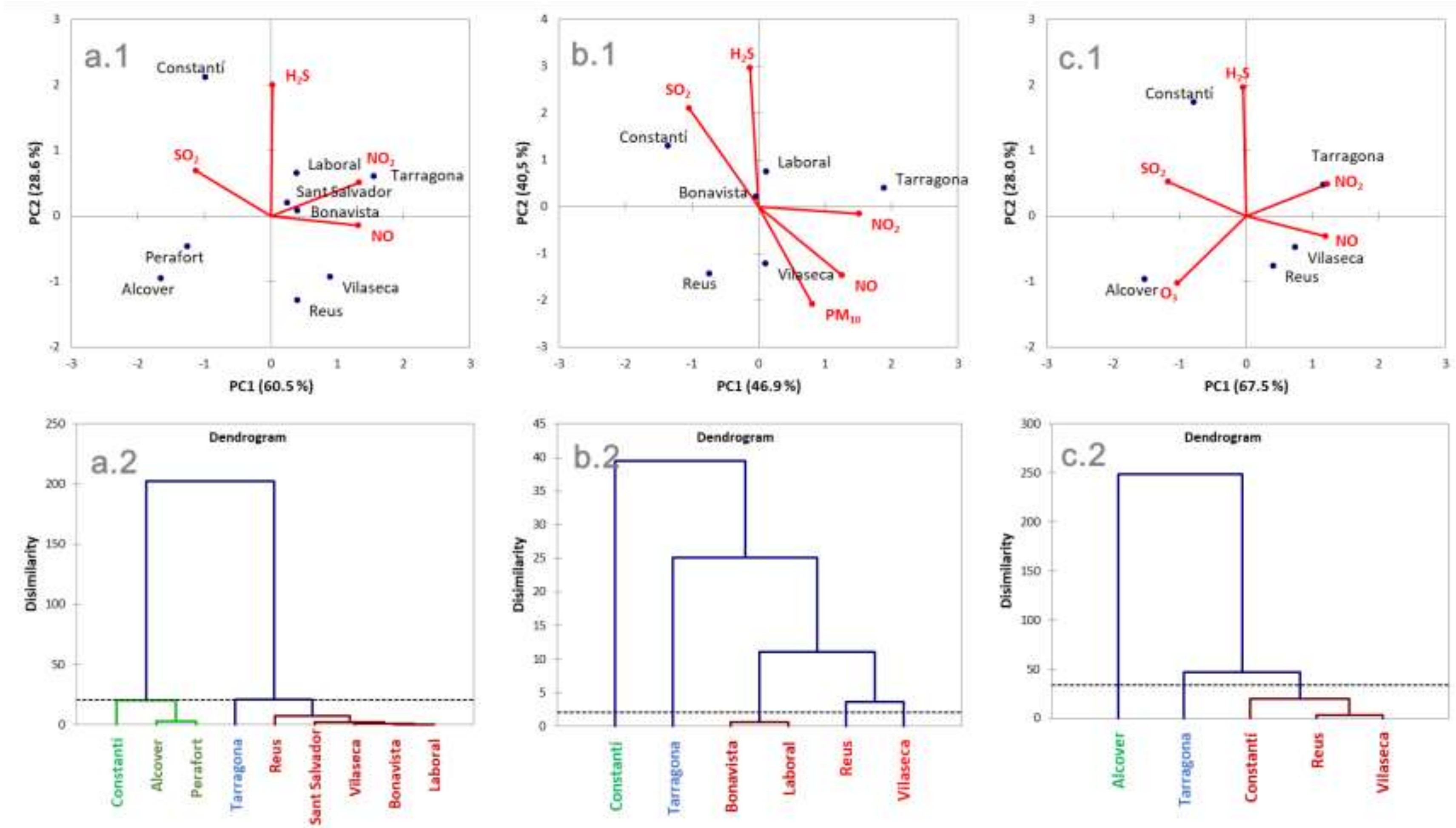

Fig. 3. Principal components analysis (1) and dendrograms (2) of annual mean pollutant levels in stations of the Camp de Tarragona. As not the same pollutants were measured in each station, different pollutants were included in different graphs: NO_2, NO, SO_2 and H_2S in (a); NO_2, NO, SO_2, H_2S and PM_{10} in (b), and NO, NO_2, SO_2, H_2S and O_3 in (c).

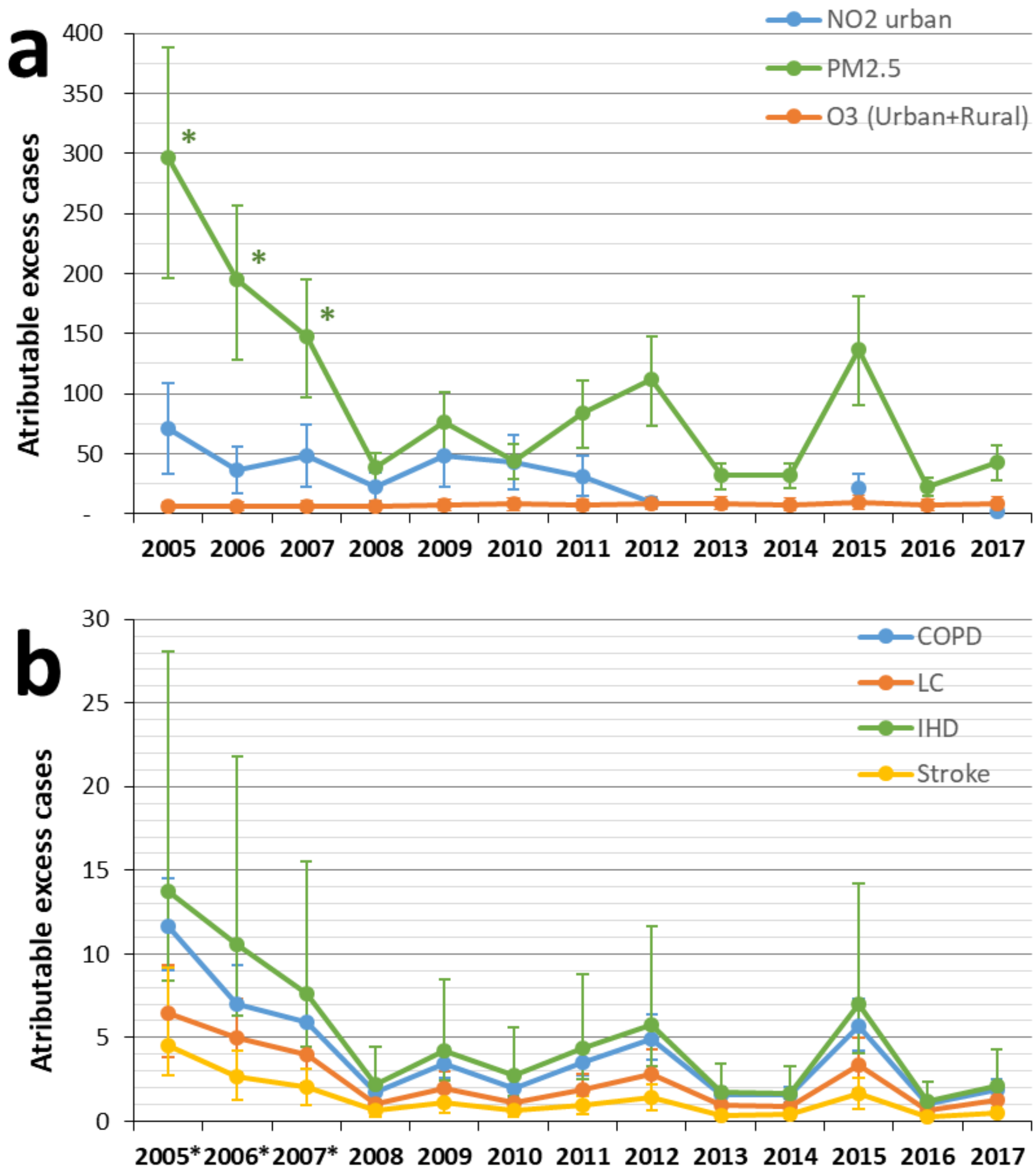

Fig. 4. Attributable excess cases of mortality due to: a) all natural causes and b) chronic obstructive pulmonary disease (COPD), lung cancer (LC), ischemic heart disease (IHD) and stroke, attributable to air pollution above the WHO threshold scenario in the Camp de Tarragona (2005-2017). Points and error bars are central values and 95% confidence intervals, respectively. * Indicates that $PM_{2.5}$ levels from 2005 to 2007 were calculated from PM_{10}.

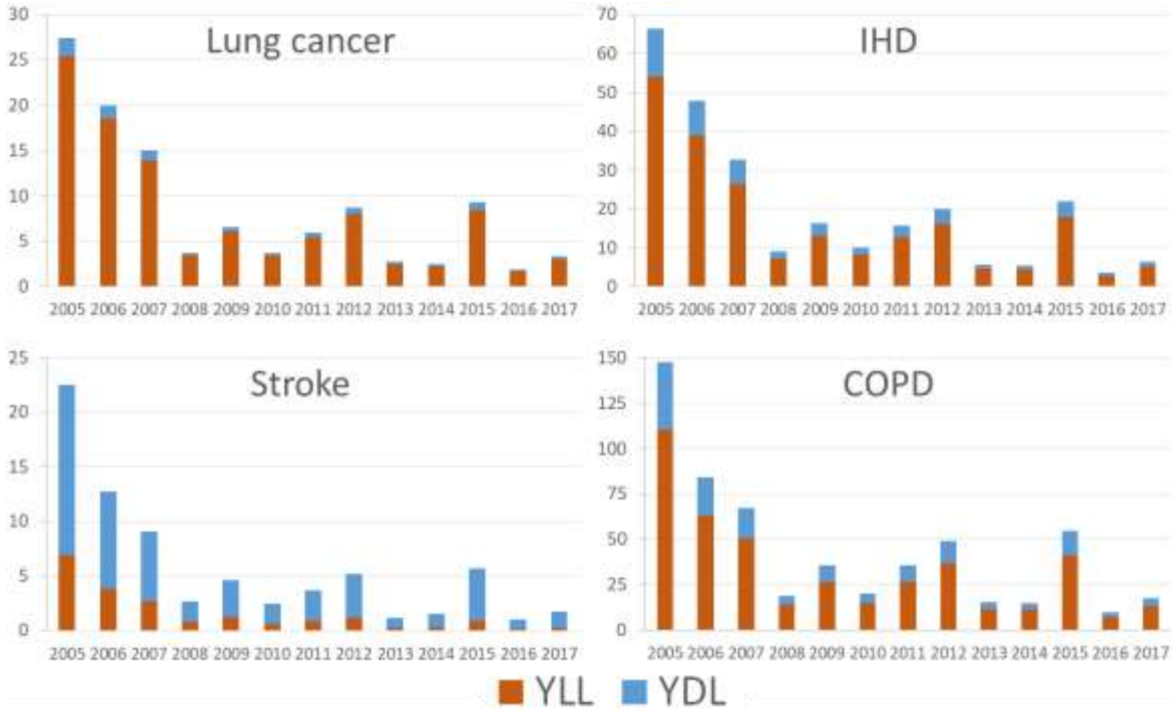

Fig. 5. Disability-Adjusted Life Years (YLDs plus YLLs), per 100,000 inhabitants for lung cancer, ischemic heart disease (IHD), stroke, and chronic obstructive pulmonary disease (COPD) due to $PM_{2.5}$ levels above the WHO threshold limits in the Camp de Tarragona during the 2005-2017 period.