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Temporal change and impact on air quality of an energy recovery plant using the M-BACI design in Gipuzkoa

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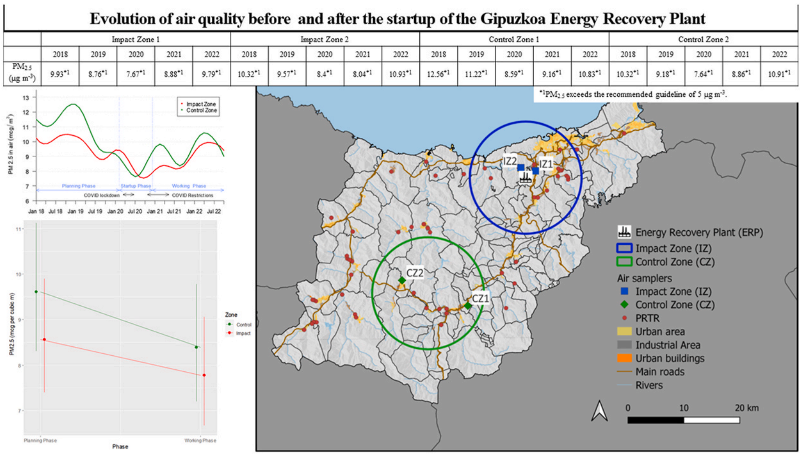
HIGHLIGHTS

- Air quality assessment with BACI design after the start-up of an ERP.
- $PM_{2.5}$ and trace elements levels remained below pre-ERP levels.
- Selenium was the only pollutant with increased levels in the post-ERP period.

GRAPHICAL ABSTRACT



Evolution of air quality before and after the startup of the Gipuzkoa Energy Recovery Plant

	Impact Zone 1					Impact Zone 2					Control Zone 1					Control Zone 2				
	2018	2019	2020	2021	2022	2018	2019	2020	2021	2022	2018	2019	2020	2021	2022	2018	2019	2020	2021	2022
$PM_{2.5}$ ($\mu g\ m^{-3}$)	9.93^{*1}	8.76^{*1}	7.67^{*1}	8.88^{*1}	9.79^{*1}	10.32^{*1}	9.57^{*1}	8.4^{*1}	8.04^{*1}	10.93^{*1}	12.56^{*1}	11.22^{*1}	8.59^{*1}	9.16^{*1}	10.83^{*1}	10.32^{*1}	9.18^{*1}	7.64^{*1}	8.86^{*1}	10.91^{*1}

$^{*1}PM_{2.5}$ exceeds the recommended guideline of 5 $\mu g\ m^{-3}$.

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ABSTRACT

A significant concern in our society is the potential impact on both health and the environment of air pollutants released during the incineration of waste. Therefore, it is crucial to conduct thorough control and monitoring measures. In this context, the objective of this research was to study the evolution of particulate matter ($PM_{2.5}$) and associated trace elements during the period before and after the installation of an Energy Recovery Plant (ERP). For that, a descriptive and temporal analysis of $PM_{2.5}$ concentration and composition were performed on two similar areas (impact/control) using the Before-After/Control-Impact (BACI) design and two periods (before

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BACI design
ERP

from January 01, 2018 to February 06, 2020 and after from December 10, 2020 to September 30, 2022). Results showed a decrease in the levels of $PM_{2.5}$ and associated trace elements is observed in the impact zone (IZ) and in the control zone (CZ) throughout the study period. In the case of $PM_{2.5}$, the most notable decrease occurred in the period of the start-up of the ERP, a period that coincides with the confinement and restrictions of COVID, with a subsequent increase in both zones, without reaching the levels observed in the period prior to the start-up of the ERP. Selenium is the only trace element that increases significantly in the IZ. In conclusion, a decrease is observed for all pollutants except selenium in both zones, although less pronounced in the IZ. Since selenium already showed an upward trend in the phase prior to the start of the ERP, it is necessary to investigate its evolution and find out the possible cause.

1. Introduction

Eurostat's 2023 data reveals that each citizen in the EU-28 generates 530 kg of municipal waste. The primary methods of waste management encompass recycling, composting, wastewater treatment, landfill, and incineration. The latter covers 26% of municipal waste treatment, mostly in energy recovery plants (ERP) where solid waste undergoes combustion to generate electricity (Tait et al., 2020), reducing waste volume by about 90%. However, it is important to note that this process also releases various atmospheric pollutants, particulate matter (PM), acids and other gases, carbon compounds, greenhouse gases, dust and heavy metals (Tait et al., 2020).

$PM_{2.5}$ is a type of particle with a diameter of less than 2.5 μm characterized by its small diameter, large relative surface area and high ability to absorb toxins (Wang et al., 2021). This particulate matter is classified as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC) and is currently the most significant air pollutant affecting public health (Cohen et al., 2017). In 2021, the European Environment Agency (EEA) reported that the premature deaths attributable to $PM_{2.5}$ were 253,000 in the EU (EEA, 2023). Notably, Italy, Poland, Germany, Romania, and Spain recorded the highest absolute numbers of premature deaths attributed to $PM_{2.5}$. In compliance with the directive, since 2020 the limit of annual mean levels for $PM_{2.5}$ in Europe is set at 20 μg per cubic meter (μg m^{-3}) (Directive, 2008/50/EC). In the year 2021, the World Health Organization (WHO) set a more stringent annual mean recommendation of 5 μg m^{-3}, since it is estimated that the increase in mortality is 6–13% for each increase of 10 μg m^{-3} of $PM_{2.5}$ (Pope et al., 2018). In line with efforts to combat air pollution, the European Commission's Zero Pollution 2050 plan outlines interim targets aligning more closely with WHO recommendations, proposing a reduction of over half in the annual limit value for $PM_{2.5}$ to work towards achieving zero air pollution by 2050 and consequently reducing by more than 55% premature deaths caused by air pollution (European Commission, 2021).

$PM_{2.5}$, with a diameter less than 2.5 μm, can cross the alveolar-capillary membrane and enter the blood system (Schwartz and Neas, 2000), subsequently accessing different tissues and organs (Thurston et al., 2021). Chronic exposure to these particles heightens the risk of developing cardiovascular and respiratory diseases, along with an increased likelihood of lung cancer (De Bont et al., 2022; Du et al., 2016; Li et al., 2018). Based on scientific evidence, a short and long-term morbidity and mortality effect from short- and long-term exposure to particulate matter has been observed (Ali et al., 2013; Burnett et al., 2014; Schwartz al., 2018).

$PM_{2.5}$ particles comprise both liquid and solid components, including sulphates, nitrates, black carbon, organic chemicals, and metals. Particulate matter contains trace elements such as arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb) and selenium (Se) among others (Chen et al., 2021; Cusack et al., 2012). Some of these trace elements can be toxic. According to the WHO and the Agency for Toxic Substances and Disease Registry (ATSDR), chromium, cadmium and lead are carcinogenic and neurotoxic. Acute and/or chronic metal contamination can cause health problems such as nerve disorders, cancer, respiratory failure, asthma, abnormalities in fetuses, effects on behavior and intelligence, kidney damage and even death (Hou et al., 2019; Jan et al., 2015).

Research on air quality usually consists of a comparison of means and/or medians without considering temporal variability. Moreover, when temporality is taken into account, the samples are not taken daily and only the trend of the pollutants is studied (whether it decreases or increases) without comparing it to control sites. In order to overcome these limitations, we opted for a model commonly employed in ecology studies known as the Before-After/Control-Impact design or BACI. To the best of our knowledge, it has not been used so far to study the potential impact of an ERP on air quality. This model allows us to assess, taking into account temporal variability, whether observed changes in the air can be linked to the placement of a new emission point. This design is frequently used to assess the effects of environmental perturbations on ecological systems when the timing and location of the impact area are known, and pre-data are collected. It is considered an optimal approach for isolating the effects from natural variability (Chevalier et al., 2019; Smokorowski and Randall, 2017). The fundamental concept involves comparing the state of a system with an intervention to the state of a system where the intervention does not occur. The establishment of an ERP in the province of Gipuzkoa (Basque Country, Spain) provided an opportunity to analyze the potential effects of the plant on air quality. To this end, air quality data were collected before and after the operation of the plant in two zones, allowing the utilization of the BACI methodology to assess the ERP's impact.

To our knowledge, this is the first study in which this methodology has been used to analyze the impact of an EPR on air quality. In particular, we have only found one other study that has monitored dioxin levels in environments prior to the start-up of an EPR plant (Caserini et al., 2004). However, we have not found similar papers related to analysis of particles and trace elements. Therefore, our study is particularly relevant to understanding the impact of an EPR on air quality.

This study has two main aims. The first aim is to determine the changes in air quality measured at immission points concerning $PM_{2.5}$ and associated trace elements before and after the startup of the Zubieta ERP installed in the province of Gipuzkoa (Basque Country, Spain). The second aim is to use BACI design methodology to evaluate the changes observed in air quality due to the implementation of the ERP.

2. Materials and methods

The analysis includes assessing the levels of $PM_{2.5}$ and associated trace elements across distinct study periods: the period prior to the start-up of the ERP, the start-up period, and the period of full operation of the ERP. The study delves into the temporal change and trends of these air quality parameters. Furthermore, the research involves a comparison of $PM_{2.5}$ and trace element values before and after the start-up, as well as between the control and intervention areas, using a BACI design. This methodology aids in systematically evaluating the impact of the ERP on air quality, considering both the temporal changes and the spatial variations between the control and intervention zones.

2.1. Study design

The research was conducted in two industrial environments located in the province of Gipuzkoa (Basque Country, Spain) (Fig. 1). The “Impact Zone (IZ)," where the Energy Recovery Plant (ERP) is situated, encompasses four industrial complexes engaged in iron and steel activities, as documented in the Spanish Register of Emissions and Pollutant Sources (PRTR). The selection of the “Control Zone (CZ)" located 28 km away from the ERP, was based on ensuring a comparable presence of metallurgical industry (with an important steel industry), and with six industrial complexes documented in the Spanish Register of Emissions and Pollutant Sources (PRTR). Both areas had similar characteristics in terms of population and topography, characterized by narrow valleys with little aerial dispersion and intense traffic (Lertxundi et al., 2010).

Air samples collection began in September 2017, but for this study, only the data collected between January 01, 2018 and 09/30/2022 was considered, which was divided into three periods depending on the period in which the ERP was:

1. **Period prior to the start-up of the ERP** (01/01/2018–06/02/2020)
2. **Period of start-up of the ERP** (07/02/2020–09/12/2020)
3. **Period of full operation of the ERP** (10/12/2020–09/30/2022)

2.2. Data collection and laboratory analysis

Concentrations of $PM_{2.5}$ and 16 trace elements were determined. The trace elements analyzed were: arsenic (As), barium (Ba), cadmium (Cd), cerium (Ce), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), nickel (Ni), palladium (Pd), selenium (Se), vanadium (V), and zinc (Zn). PM2.5 and trace element concentrations were collected daily using Digitel DAH 80 high-volume samplers. Four samplers were located at immission points in the study area: two in the CZ (CZ1 and CZ2) and two in the IZ (IZ1 and IZ2) (Fig. 1). Samplers operated at a flow rate of 30.0 ± 1.5 m3 h-1 under ambient conditions for 24 h, ensuring a quantification limit of 2 $\mu g\ m^{-3}$. Particles were collected on Whatman QMA 150-mm quartz-fibre filters. Filters were stored in controlled laboratory conditions, maintaining ambient temperature (20 ± 1 °C) and relative humidity (50 ± 5%) for a minimum of 48 h.

Particle mass was determined using a gravimetric method on calibrated scales with a maximum uncertainty of 0.09 mg (K = 2). Trace element content was analyzed by digesting the filters with concentrated nitric acid at 220 °C for approximately 20 h. The digested samples were processed using an inductively coupled plasma mass spectrometry (ICP-MS) system (Agilent 7500a) equipped with a Babington nebulizer, a collision cell, and a Shieldtorch for low-temperature operations. Maintenance, verification, and calibration of the instrumental analysis equipment and physicochemical tests were performed under the ISO 17025 Quality Management System.

2.3. Statistical analysis

We divided the analytical work of this project into three parts: descriptive analysis, time-series plotting and BACI analysis, which are complementary to each other. The descriptive analysis and time-series plotting utilized air quality data from three distinct periods. For assessing the impact of the ERP on air quality using the BACI methodology, we focused on data from two specific periods: the period before the ERP startup and the period of full ERP operation. Descriptive statistics were computed for $PM_{2.5}$ and the 16 trace elements across all monitoring stations. We also cross-tabulated descriptive statistics for

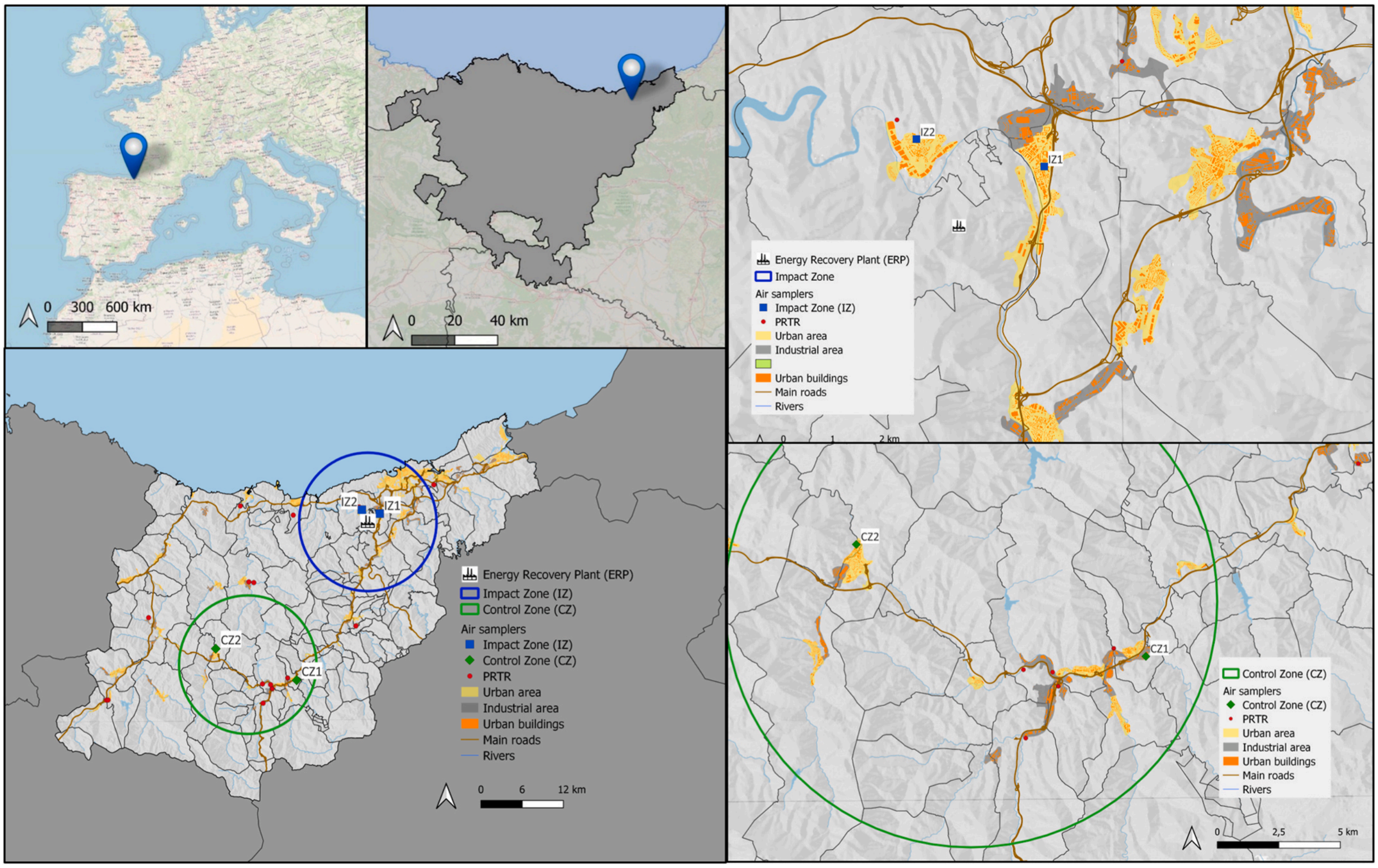

Fig. 1. Map of the studied area.

$PM_{2.5}$ and trace elements, using each combination of monitoring station and period. In order to evaluate the trend and current regulation, the mean annual levels of $PM_{2.5}$ and trace elements were computed for each station and year. The year 2022 only considers samples until the 30th of September (272 days). The correlation between each of the pollutants studied in each zone (*control* and *impact*) and period (*before* or period prior to start-up (1) and *after* or the period of operation at full capacity (3)) was also calculated to identify common emission sources.

For plotting time-series data, smoothing techniques were applied to graphically show both the seasonality and trends of the quantified 17 pollutants over the period from January 2018 to September 2022. To smooth and plot mean curves, we used functions of the R package fdapace (Zhou et al., 2022). The bandwidth values for the smoothed mean functions were automatically determined based on generalized cross-validation (Silverman, 1986). On an empirical basis, a Gaussian smoothing kernel was selected (Wand and Jones, 1994; Bowman and Azzalini, 1997).

To detect a possible impact in the $PM_{2.5}$ and trace elements levels due to the ERP, we used a Before-After/Control-Impact (BACI) design (Green, 1979; Bernstein and Zalinski, 1983; Stewart-Oaten et al., 1986; Underwood, 1992), a design later reviewed by Underwood (1993) and Stewart-Oaten and Bence (2001). For this purpose, we used a <Phase> factor, representing <time>, i.e., temporal variation; this factor had two levels (<Before> and <After>). The Before level (Period prior to the start-up of the ERP) covered the period from January 01, 2018 to February 06, 2020 (25 months); the After level (Period of full operation of the ERP) included the period from December 10, 2020 to 09/30/2022 (22 months). The period corresponding to the start-up of the ERP (with or without some waste combustion) was excluded from the BACI analysis, since the ERP was not at full operation and, besides, this period was coincident with various confinement periods and movement restrictions due to the COVID pandemic. Likewise, we used a <Zone> factor with two levels (<Control> and <Impact>), representing spatial variation; the Control level included the results from the two sensors located in the CZ (CZ1 and CZ2) and the Impact level included the results from the two sensors in the IZ (IZ1 and IZ2). Thus, data related to the four sample sites (two in the CZ and two in the IZ) were analyzed.

To analyze the above BACI design, we applied Gaussian linear mixed modelling (Bolker et al., 2009; Harrison et al., 2018), using not only the said Zone and Phase factors as fixed factors but also four random terms (station, season, month, and date, with date nested within month, and month nested within season). These random terms were planned to control for station-to-station, day-to-day, month-to-month, and season-to-season variation in $PM_{2.5}$. Because mixed modelling is robust to sparse datasets (Pinheiro and Bates, 2000), this technique is applicable to the present BACI analysis. Data analysis was applied mainly via functions of the packages lme4 (Bates et al., 2015) and lmerTest (Kuznetsova et al., 2017) in R software (R Core Team, 2017).

3. Results

3.1. Descriptive statistics

In overall, 5564 samples of $PM_{2.5}$ and 5476 samples of trace elements were collected across all monitoring stations and periods (Table S1 at Supplementary material). Table 1 includes the annual means for $PM_{2.5}$ and trace elements in all the monitoring stations. In general, higher values are observed in the control zone than in the impact zone from the beginning of the study until 2022. The annual means legal limits for $PM_{2.5}$ established in Spain (BOE-A-2011-1645) (20 µg m^{-3}) are not exceed. However, the WHO guideline recommendation for $PM_{2.5}$ (5 µg m^{-3}) is exceeded every year in the four stations (CZ1, CZ2, IZ1 and IZ2). Moreover, in CZ1 the described objective of the air quality directive for 2030 (10 µg m^{-3}) is exceeded in the years 2018, 2019 and 2022, in CZ2 for the years 2018 and 2022 and in IZ2 for the years 2018 and 2022. Metals in $PM_{2.5}$ are not regulated; however, the European directive

Table 1

Annual means (from 2018 to 2022) for $PM_{2.5}$ and all trace elements. IZ: Impact zone, CZ: Control zone.

	IZ1					IZ2					CZ1					CZ2				
	2018	2019	2020	2021	2022	2018	2019	2020	2021	2022	2018	2019	2020	2021	2022	2018	2019	2020	2021	2022
$PM_{2.5}$ (µg m^{-3})	9.9*1	8.7*1	7.6*1	8.8*1	9.7*1	10.3*1	9.5*1	8.4*1	8.0*1	10.9*1	12.5*1	11.2*1	8.5*1	9.1*1	10.8*1	10.3*1	9.1*1	7.6*1	8.8*1	10.9*1
Arsenic (ng m^{-3})	0.2	0.3	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.8	0.7	0.5	0.6	0.6	0.2	0.2	0.2	0.2	0.2
Barium (ng m^{-3})	2.1	2.3	2.2	2.3	2.2	2.3	2.4	2.2	2.2	2.2	2.6	2.5	2.2	2.3	2.4	2.5	2.2	2.2	2.5	2.3
Cadmium (ng m^{-3})	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.5	0.2	0.2	0.3	0.2	0.2	0.2	0.2	0.2
Cerium (ng m^{-3})	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.1	0.1	0.1	0.0	0.0	0.1	0.1	0.1	0.1	0.0	0.1	0.1
Chromium (ng m^{-3})	2.6	2.9	2.2	2.4	2.3	4.5	2.6	2.1	2.1	2.2	27.0	54.8	11.2	7.4	11.9	4.2	3.6	3.6	3.2	2.8
Cobalt (ng m^{-3})	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.0	0.2	0.4	0.1	0.0	0.1	0.0	0.0	0.0	0.0	0.0
Copper (ng m^{-3})	4.5	4.7	3.8	3.6	3.7	4.4	3.6	2.9	3.1	3.1	29.8	46.1	19.0	15.7	17.4	5.9	4.5	4.5	4.3	3.9
Iron (µg m^{-3})	0.0	0.1	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.1	0.3	0.4	0.1	0.1	0.2	0.1	0.1	0.0	0.1	0.1
Lead (µg m^{-3})	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Manganese (ng m^{-3})	4.7	5.3	4.8	12.0	4.9	7.5	4.9	4.5	5.1	5.4	29.5	42.7	14.3	22.7	15.1	6.4	6.1	6.1	5.9	6.1
Mercury (ng m^{-3})	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Nickel (ng m^{-3})	2.2	2.1	2.0	2.2	2.0	2.7	2.0	2.0	2.1	2.0	17.8	33.5*2	6.8	4.5	7.0	2.7	2.3	2.4	2.2	2.1
Palladium (ng m^{-3})	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Selenium (ng m^{-3})	0.3	0.3	0.2	0.3	0.3	0.5			0.7	1.4	0.3	0.3	0.2	0.2	0.3	0.2	0.2	0.2	0.2	0.2
Vanadium (ng m^{-3})	0.8	0.8	0.3	0.3	0.3	0.8	0.7	0.3	0.3	0.4	0.7	0.7	0.2	0.3	0.4	0.6	0.6	0.2	0.3	0.3
Zinc (µg m^{-3})	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0

*1$PM_{2.5}$ exceeds the recommended guideline of 5 µg m^{-3}*2Nickel exceeds the legal European objective value of 20 ng m^{-3} in PM_{10}.

regulates the average annual concentration of some of them in PM_{10}. When transforming the concentration of $PM_{2.5}$ to PM_{10}, taking as reference the $PM_{10}/PM_{2.5}$ correlations observed in 2014 at the M^{a} Díaz Environmental station (Bilbao) (Gobierno Vasco, 2014), located in an urban environment, the estimate of the average value of Ni in PM_{10} was higher than 20 ng m^{-3} established in the European Directive for PM_{10}. The only other pollutants with objective values in the PM_{10} fraction are arsenic (6 ng m^{-3}) and cadmium (5 ng m^{-3}) which are not exceeded. The legal limit for lead (0.5 μg m^{-3}) is not surpassed either. It is important to note that for the rest of studied pollutants there are no legal limits or objective values.

Cross-tabulated descriptive statistics for $PM_{2.5}$, considering each combination of monitoring station and period, are available in Table 2. In the case of $PM_{2.5}$, the most notable decrease occurred in the period of the start-up of the ERP, a period that coincides with the confinement and restrictions of COVID, with a subsequent increase in both zones, without reaching the levels observed in the period prior to the start-up of the ERP. Table S2 (Supplementary material) provides cross-tabulated statistics for all trace elements.

Fig. 2 includes the correlation plots for each combination of zone and periods. The correlation values between chromium, cobalt, copper, iron, nickel, and manganese are high (greater than 0.75) in the control zone before the ERP starts working and lower in the after period. In the impact zone the correlation between cerium and iron is high (0.73) before the ERP starts working and in the working phase (0.79), as well as that between arsenic and copper (0.77) in the posterior period.

3.2. Time-series plotting

Smoothing techniques were utilized to visually depict the seasonality and trends of 17 pollutants spanning from January 2018 to September 2022 (Figs. 3 and 4). Prior to the implementation of the ERP (January 1, 2018, to June 2, 2020), $PM_{2.5}$ levels showed a declining trend in both the CZ and IZ, with a more pronounced decrease in the CZ, reaching its lowest point in the middle of the startup phase around July 2020. Subsequently, the trend reversed in both locations, rising steadily until the conclusion of the study period in October 2022. Throughout this timeframe, $PM_{2.5}$ exhibited a seasonal pattern in both zones, with peak levels observed at the end of winter and troughs at the end of summer (Fig. 3). The behavior of trace elements generally mirrored that of $PM_{2.5}$ to some extent (Fig. 4). Most trace elements displayed seasonal variations similar to $PM_{2.5}$. Chromium, cobalt, copper, iron, lead, manganese, nickel, and zinc demonstrated comparable patterns, whereas arsenic, palladium, vanadium, and others exhibited distinct seasonal characteristics. Notably, selenium showed an increasing trend only in the impact zone since the study's commencement, differing from the control zone.

3.3. BACI analysis

A Before-After/Control-Impact (BACI) analysis was applied to $PM_{2.5}$ original measurements. Controlling for station-to-station, day-to-day, month-to-month, and season-to-season variation, we find that the fixed factors zone, phase, as well as the interaction between them, explained only $R\text{-}^{2}_{\text{adj.}} = 2.3\%$ in the observed variation in $PM_{2.5}$. The hypothesis test for the interaction term (Zone x Phase), which represents the so-called BACI effect, gives $F_{1,\ 3193} = 6.9$, with p-value = 0.008 (Table 3).

The estimate for the BACI effect of $PM_{2.5}$ is found to be −0.43 μg m^{-3} with 95% confidence interval (−0.49, −0.38) (Table 3 and Fig. 5). In the context of an overall decrease of $PM_{2.5}$ immission levels, the emission in the CZ is decreasing faster than in the IZ (the BACI effect tells us that there is a "deficit" of reduction in the impact zone of −0.48 μg m^{-3}). The estimates for the BACI effects of the other pollutants are all negative (except for vanadium which is not statistically significant) implying there is "deficit" of reduction in the impact zone for all pollutants. The estimates are included in Table 3 and their BACI comparisons in Fig. S1 (Supplementary material).

4. Discussion

Given the negative health effects of air pollutants, it is necessary to assess whether the implementation of an ERP affects air quality in its vicinity. Using the BACI methodology, which considers a before and after scenario and includes a control area, we have analyzed the impact of an ERP on air quality (PM2.5 and trace elements).

The results of the present study indicate that at no point in time or zone, did the mean levels of $PM_{2.5}$ (<12.56 μg m^{-3}) exceed the legal limit of 20 μg m^{-3}. However, the WHO guidelines for 2030 were exceeded in the CZ and almost in the IZ after the winter of 2022. The mean levels of $PM_{2.5}$ had a clear decreasing trend in both the CZ and IZ throughout the entire period prior to the start-up of the ERP and the first half of the start-up period. By contrast, these mean levels tended to increase in both the CZ and IZ during the second half of the start-up period and the entire period of full operation. Nonetheless, the mean levels were higher in the CZ compared to the IZ, except for a short period during the first half of the start-up period. Although a reduction has been seen, this improvement has not been as strong in the exposed area compared to the control area, and it cannot be ruled out that it is because there is a new source of exposure, the ERP.

The average annual concentrations of $PM_{2.5}$ and trace elements obtained in this study were similar to those reported in 2021 by the air quality network of the Basque autonomous community (Gobierno Vasco, 2022). These levels were comparable to or lower than those found in both indoor and outdoor environments of other European cities (Götschi et al., 2002; Vallius, 2005; Kaleta and Kozielska, 2022). Our results also aligned with findings from two studies that analyzed air

Table 2
Descriptive statistics for $PM_{2.5}$ (μg m^{-3}) for each zone and period of the study. IZ: Impact zone, CZ: Control zone.

Station	Period of the ERP	*n*	Min	Max	Range	Mean	StDev	Median	IQR
IZ1	Prior to the start-up	728	2.0	34.0	32.0	9.3	4.5	9.3	6.0
	Start-up	230	1.0	26.0	25.0	7.5	3.8	7.5	4.0
	Full operation	511	1.0	27.0	26.0	9.0	4.6	9.0	5.0
IZ2	Prior to the start-up	550	1.0	56.0	55.0	10.5	5.9	10.5	7.0
	Start-up	266	1.0	55.0	54.0	8.2	5.5	8.2	5.0
	Full operation	500	2.0	41.0	39.0	9.0	5.3	9.0	6.0
CZ1	Prior to the start-up	732	2.0	36.0	34.0	12.0	5.8	12.0	7.0
	Start-up	300	2.0	22.0	20.0	8.0	3.8	8.0	5.0
	Full operation	501	2.0	57.0	55.0	9.7	5.2	9.7	6.0
CZ2	Prior to the start-up	520	1.0	68.0	67.0	9.9	5.5	9.9	6.3
	Start-up	221	2.0	23.0	21.0	7.3	3.8	7.3	5.0
	Full operation	505	2.0	59.0	57.0	9.6	5.8	9.6	6.0

n: sample size; Min: minimum; Max: maximum; StDev: standard deviation; IQR: interquartile range.

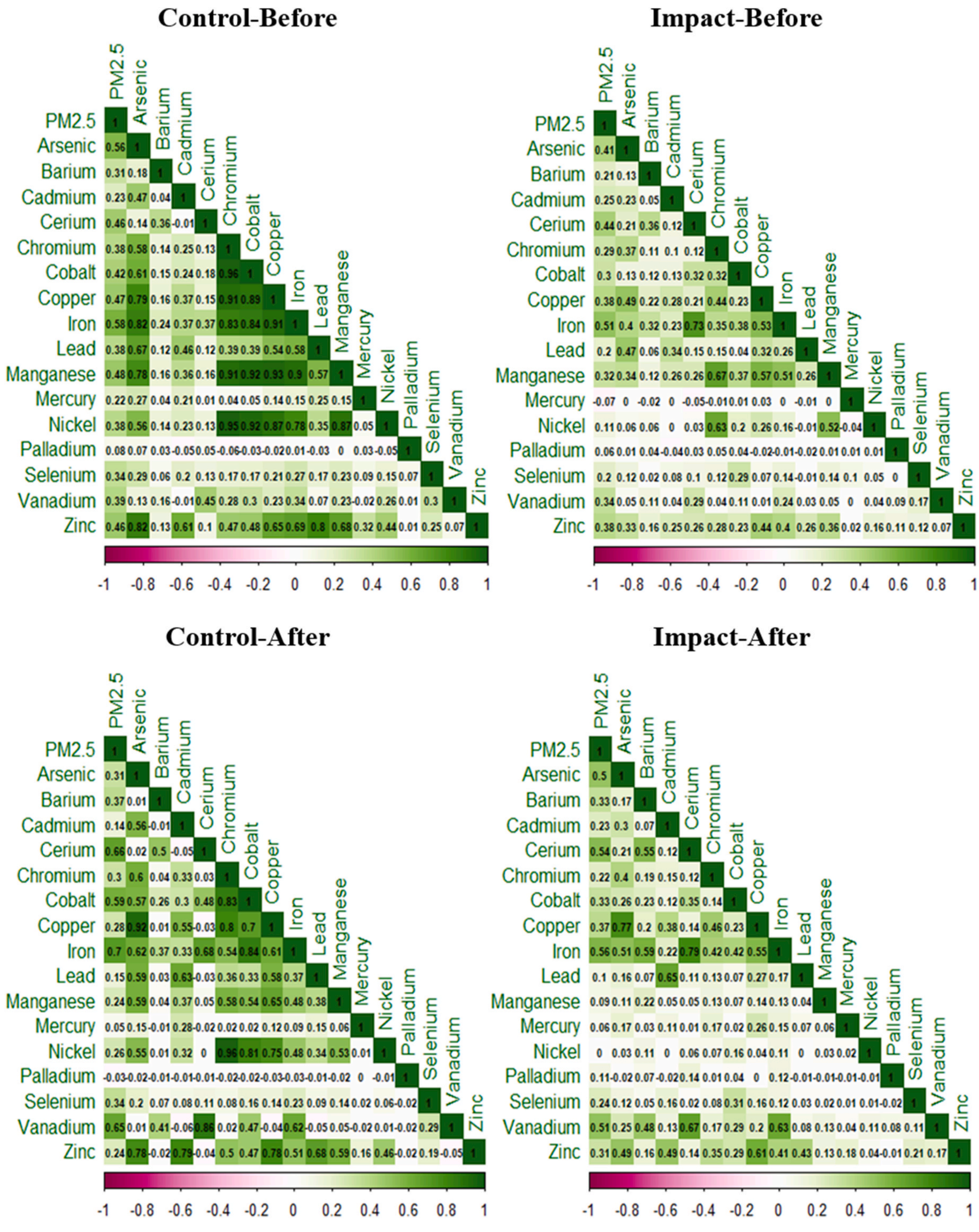

Fig. 2. Correlation plots between $PM_{2.5}$ and trace elements for each combination of zone and periods.

quality in the vicinity of energy production facilities (Aboh et al., 2007; Pan et al., 2021). However, most studies on incinerators and air quality focus on PM_{10} particle levels. To facilitate comparison, we converted our $PM_{2.5}$ results to PM_{10} by using the $PM_{2.5}/PM_{10}$ ratio for each sampling point. For example, in 2021, the average annual $PM_{2.5}$ concentration in IZ was 8.9 μg m^{-3}, corresponding to 15.32 μg m^{-3} for PM_{10}. In CZ, the average $PM_{2.5}$ concentration was 9.38 μg m^{-3}, equating to 15.73 μg m^{-3} for PM_{10}. These levels are similar to or lower than those reported in other studies measuring PM_{10} near urban waste incinerators in Italy (Conca et al., 2020; Barrera et al., 2015) and England (Douglas et al., 2017).

Chromium, cobalt, copper, iron, lead, manganese, nickel, and zinc exhibit similar patterns, indicating a common industrial origin. By the end of 2020, levels of these pollutants in the CZ began to decline due to an industrial crisis and intervention and control measures implemented by the Environmental Management Directorate of the Basque Government. In 2016, it was found that nickel levels in CZ1 exceeded legal air quality standards throughout 2015. In response, the Environmental Management Directorate set up a working group to address this problem. Nickel concentrations were lowest on weekends and holidays,

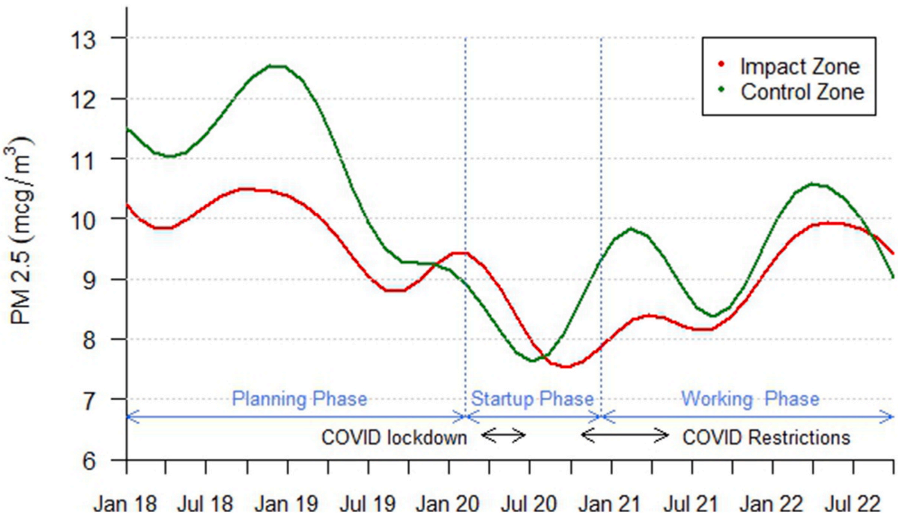

Fig. 3. Temporal changes in mean levels of $PM_{2.5}$ in air in each zone. The study period covers three stages (Planning Phase, Startup Phase and Working Phase), but the data for the Startup Phase have no role in the BACI analysis itself and are shown here only for completeness and descriptive purposes.

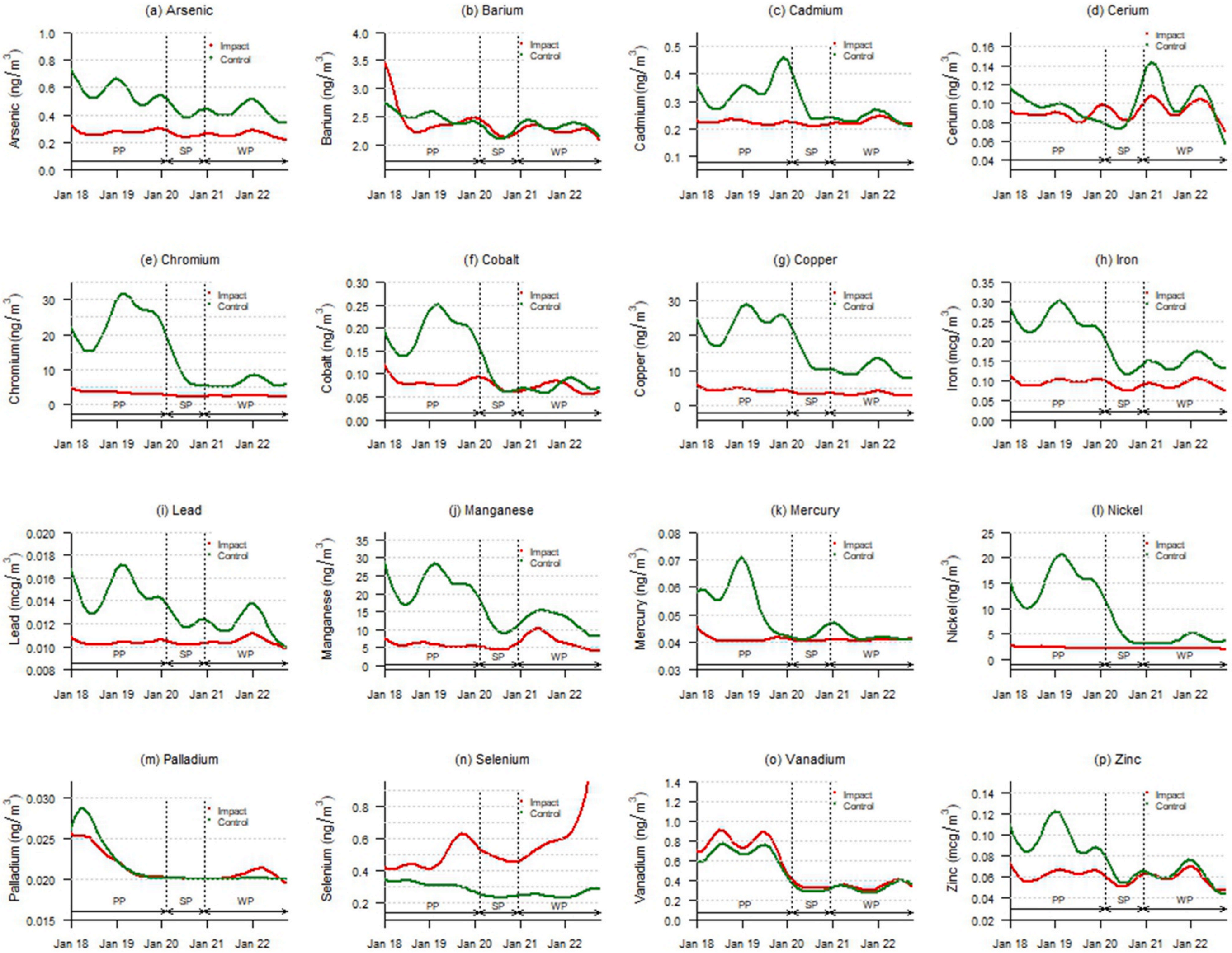

Fig. 4. Temporal changes in mean levels of trace elements in air in each zone. The study period covers three stages (Planning Phase, Startup Phase and Working Phase), but the data for the Startup Phase have no role in the BACI analysis itself and are shown here only for completeness and descriptive purposes.

Table 3
BACI effects for PM$_{2.5}$ and all trace elements.

Variable	Point estimate	95% Lower bound	95% Upper bound	df	F	p-value
PM$_{2.5}$ (µg m^{-3})	−0.4	−0.4	−0.3	(1, 3192.6)	6.9	0.00848
Arsenic (ng m^{-3})	−0.0	−0.0	−0.0	(1, 3269.5)	15.4	<0.0001
Barium (ng m^{-3})	−0.1	−0.1	−0.1	(1, 3320.6)	7.5	0.00599
Cadmium (ng m^{-3})	−0.0	−0.0	−0.0	(1, 3409.4)	41.9	<0.0001
Cerium (ng m^{-3})	−0.0	−0.0	−0.0	(1, 3222.9)	19.0	<0.0001
Chromium (ng m^{-3})	−2.9	−6.3	−1.3	(1, 3365.7)	106.7	<0.0001
Cobalt (ng m^{-3})	−0.0	−0.0	−0.0	(1, 3256.8)	64.1	<0.0001
Copper (ng m^{-3})	−3.0	−7.9	−1.1	(1, 3256.5)	54.8	<0.0001
Iron (µg m^{-3})	−0.0	−0.0	−0.0	(1, 3206.1)	140.5	<0.0001
Lead (µg m^{-3})	0.0	0.0	0.0	(1, 3394.3)	26.6	<0.0001
Manganese (ng m^{-3})	−2.5	−5.4	−1.1	(1, 3327.2)	60.4	<0.0001
Mercury (ng m^{-3})	0.0	0.0	0.0	(1, 2802.3)	27.6	<0.0001
Nickel (ng m^{-3})	−2.0	−4.0	−1.1	(1, 4416.1)	166.5	<0.0001
Palladium (ng m^{-3})	0.0	0.0	0.0	(1, 4419.3)	57.2	<0.0001
Selenium (ng m^{-3})	−0.0	−0.0	−0.0	(1, 3342.9)	36.9	<0.0001
Vanadium (ng m^{-3})	0.0	0.0	0.0	(1, 3107.84)	0.1	0.74006
Zinc (µg m^{-3})	−0.0	−0.0	0.0	(1, 3253.7)	8.4	0.00368

Df: degrees of freedom.

while peak levels coincided with periods of low wind speeds, pointing to a nearby industrial source near the air sampler. An industry located 300 m from the sampler was identified as a potential nickel emitter. Despite corrective measures undertaken by the company, nickel levels remained elevated by the end of 2019. Subsequently, a comprehensive study was conducted involving specific data collection from 32 companies and the installation of 5 air samplers to geographically pinpoint the problem. One of the air samplers detected unusually high nickel levels. Inspections of nearby companies identified a second source of nickel emissions in December 2019. Both interventions included enhancements to ventilation systems, installation of additional filtration systems, and cessation of used oil recycling in stamping processes at these factories. As a result, there was a notable 92% reduction in nickel levels (Gobierno Vasco, 2021). Other pollutants such as iron, copper, manganese, and chromium also decreased significantly, ranging between 73% and 91% before the startup phase began. Other pollutants (iron, copper, manganese, and chromium) decreased between 73% and 91% before the start-up phase.

However, the patterns for arsenic, palladium and vanadium seem to be element-specific. Vanadium behaves the same in both zones and selenium is the only trace element increasing in the impact zone, but not in the control zone, since the study started. Selenium is an oligoelement element crucial for the enzymes synthesis. Nevertheless, excessive exposure can result in neurological effects, respiratory tract irritation, bronchitis, and stomachache (Agency for Toxic Substances and Disease Registry, 2003). Prior to the start-up of the ERP, variations in selenium levels were observed, with occasional increases in concentrations. Although these increases have been significant, both maximum and average concentration are four orders of magnitude lower than the daily environmental exposure limit value of 0.1 mg m^{-3} established for the exposed working population. These rises have increased after the operation of the ERP; therefore, it is necessary to keep monitoring this element in order to identify potential sources.

The point estimates of the BACI effects for PM$_{2.5}$ and each trace elements are predominantly negative suggesting dissimilar pollutant trends between periods and areas within the impact zone compared to the control zone (Smith, 2002). This discrepancy may arise from the fact that the difference between control and impact sites becomes more pronounced after the intervention, relative to before. Vanadium is the only trace element with a positive point estimate, although not statistically significant, and it exhibits consistent behavior across both zones.

One challenge in accurately interpreting our findings stems from emergency interventions carried out in two industries that exceeded regulatory nickel emission levels. Consequently, two interventions occurred during the study period—the “nickel intervention” in the CZ and the startup of the ERP in the IZ—making it challenging to attribute our estimates solely to the ERP. To which intervention can we attribute the observed patterns? Two hypotheses or scenarios were considered to elucidate the time series of nickel, copper, iron, cobalt, manganese, and potentially other pollutants with similar patterns. A first hypothesis assumes that the intervention carried out to reduce nickel levels only affected the CZ and not the IZ, as it has been previously mentioned. Consequently, the significant decrease observed for nickel, copper, iron, cobalt, manganese, etc. levels, should be ascribed to the intervention in the two companies situated in the CZ. This intervention does not appear to have affected the air quality in the IZ regarding these pollutants for several reasons. In one hand, the contaminants trend remains consistent in both periods. On the other hand, the nickel levels measured in an air sampler located 2 km from these two companies did not reveal high nickel levels before and during the interventions. Hence, it is unlikely that the intervention could have affected the IZ, which is situated 26 km away.

The second hypothesis assumes that if the intervention affected both zones, it would suggest that ERP activity could have also contributed to the rise in nickel, copper, iron, cobalt, manganese, etc., in both areas. In such a scenario, the negative BACI estimation in both zones should be attributed to the combined impact of the two interventions, with nickel showing a more pronounced decrease in the CZ. However, since the CZ was selected as an area unaffected by the ERP and the decrease in nickel was not observed in the IZ, the first scenario is considered more plausible. The distinct behaviors of the two zones concerning exposure to air pollutants are evident for all compounds except vanadium.

The study’s strengths are rooted in the extensive dataset used and the extended study duration, encompassing periods before, during, and after the intervention, providing a comprehensive perspective. Furthermore, the applied methodology stands out as a significant positive aspect, as it enables continuous measurement of pollutants. The design type represents a major strength, as other studies on air quality concerning ERPs have typically focused on mean and median comparisons, lacked control sites, and/or lacked pre-installation data. The BACI design uniquely combines temporal and spatial variability, offering a more thorough understanding of the ERP’s effects in the study area. An apparent, but not effective, limitation of this study is the absence of a direct examination of the influence of weather conditions on PM$_{2.5}$ and other pollutants. Certainly, weather factors such as general conditions, temperature, and wind direction and strength can either enhance or deteriorate air quality (Chen et al., 2020; Liu et al., 2020). Understanding these environmental variables could provide insights into how temperature, humidity, and wind direction specifically contribute to air quality. However, the fact that we did not model the effect of weather conditions on air pollutants—such as temperature or wind measures—does not undermine our conclusions for two main reasons. First, we utilized comparable periods and entire seasons for both the “before” and “after” phases in both the CZ and IZ. Second, we incorporated season, month, and date (with date nested within month, and month within season) as random factors to account for the significant components

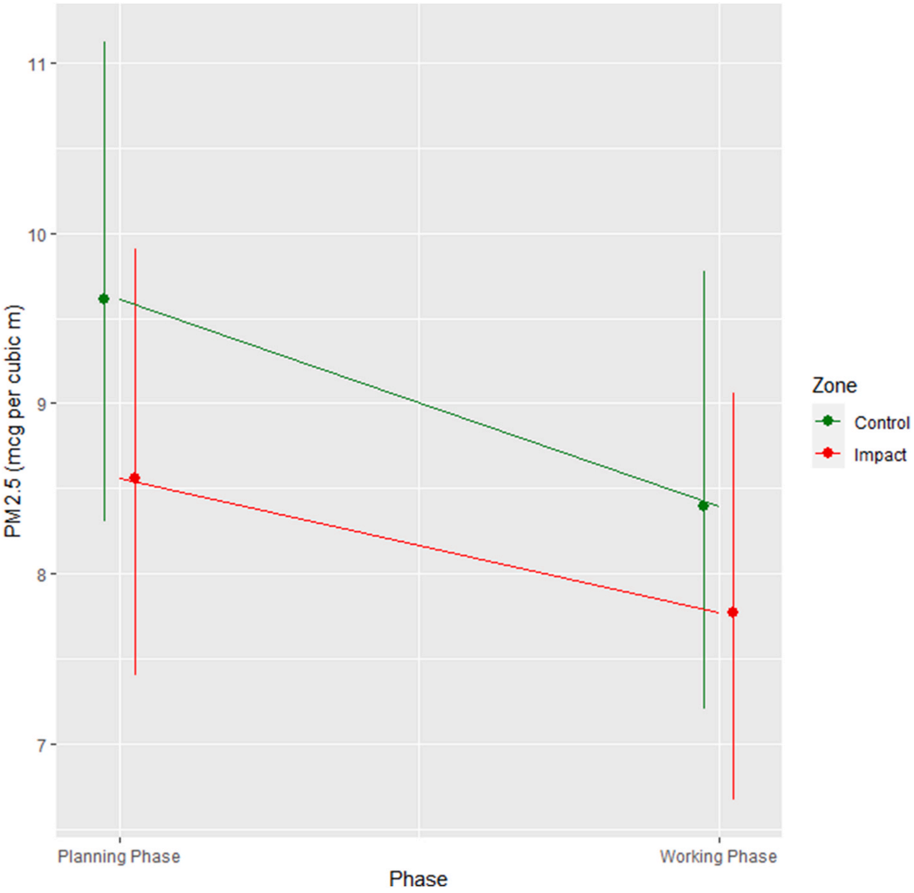

Fig. 5. BACI analysis for $PM_{2.5}$. Notice that the BACI analysis itself implies the comparison of the Planning Phase ("Before" period) to the Working Phase ("After" period) and that the data for the Startup Phase have no role in this assessment.

(seasonality and residual variation) of time series data. Moreover, throughout the study period, various industries in the region may have closed, reduced, or increased their industrial activities. Nonetheless, it is improbable that these fluctuations could have differentially impacted air pollutants in the IZ and CZ.

5. Conclusions

This is the first study where the BACI methodology has been used to assess the impact of an incinerator on air quality. We observed a clear decrease in pollutants (particles and trace elements) in both study zones, with the decrease being less pronounced in the IZ. It is important to note that the industrial activity in the study area is highly specialized in the metal sector. Similar variations in some metals are attributed to steel production processes, while other specific variations are linked to different industrial processes. The only element that shows a significant increase after the implementation of the ERP is Selenium; however, it is worth noting that it already showed an upward trend before the ERP was implemented. In this scenario, there is no evidence that the operation of the incinerator has influenced the levels of the pollutants studied. Nevertheless, continuous monitoring of pollutant trends in both zones is imperative to assess the long-term impact of the ERP on air quality.

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CRediT authorship contribution statement

Nuria Errasti: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Aitana Lertxundi:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization. **Ziortza Barroeta:** Writing – review & editing. **Jon Iñaki Alvarez:** Formal analysis. **Jesús Ibarluzea:** Writing – review & editing, Methodology, Funding acquisition, Conceptualization. **Amaia Irizar:** Writing – review & editing, Funding acquisition, Conceptualization. **Loreto Santa-Marina:** Writing – review & editing, Methodology, Funding acquisition, Conceptualization. **Nerea Urbieta:** Investigation, Data curation. **Gonzalo García-Baquero:** Writing – review & editing, Supervision, Methodology, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jesus Ibarluzea reports financial support was provided by Gipuzkoa Provincial Council. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2024.142809.

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