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An integrated strategy for air quality monitoring and management in industrial port areas

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1. Introduction

Port areas are under increasing social pressures to minimise their environmental impact and to demonstrate their environmental commitment, which is a major challenge especially in the case of industrial port areas close to urban areas (Langenus and Dooms, 2018). In this regard, the relevance of port areas is recognised as air pollution hotspots worldwide (Cesari et al., 2014; Contini et al., 2011; Feng et al., 2019; Genga et al., 2017; He et al., 2023; Kwon et al., 2023; Merico et al., 2016, 2017; Minguillón et al., 2008; Murena et al., 2018; Nguyen et al., 2022; Song et al., 2022; Tseng et al., 2021; Viana et al., 2009, 2014, 2020a; Winebrake et al., 2009). The same is true for the health impacts of shipping and port-sourced emissions, which include stack emissions as well as in-harbour transportation (on-road vehicles) and cargo handling (in industrial ports) (Broome et al., 2015; Corbett et al., 2007; Lee et al., 2019; Minguillón et al., 2008; Mueller et al., 2023; Shin and Cheong, 2011; Sofiev et al., 2018; US-EPA, 2009; Viana et al., 2020b; Winebrake et al., 2009; Yang et al., 2022). Due to globalisation, the contribution of seaborne transport to global GHG emissions is predicted to increase to 17% by 2050 if left unchecked (Schnurr and Walker, 2019), therefore carbon emission reduction in this activity has become a new challenge (Hong et al., 2023; Meng et al., 2022; Sou et al., 2022).

Acknowledging the urgent need to tackle this emission source, the EU Green Deal "Leading the transition to zero-emission maritime transport" calls to action for air quality management in port areas across Europe, among other targets. Strategies to minimise air quality impacts differ across ports, as a function to the key contributing sources: passenger ports frequently focus on stack emissions, which may be minimised through energy transition and shore-based power, while industrial ports are more highly impacted in relative terms by port emissions (on-road vehicles, handling of cargo materials) requiring tailored solutions (Lee et al., 2019; Minguillón et al., 2008), and shipyards are impacted by on-road transport and emissions from vessel refit operations (López et al., 2021).

Due to the variety of emission sources impacting port areas and their variability (mostly spatial but also temporal), air quality monitoring in ports requires dedicated strategies and instrumentation different to those typically implemented in urban areas. The use of real-time, state-of-the-art scientific instrumentation for particulate and gaseous pollutants is frequently not viable in ports due to its cost and the need for scientific expertise to process the data generated. As a result, recent studies focus on local-scale dispersion modelling tools and low-cost sensor technologies (Casazza et al., 2019; Ding et al., 2021; Isakov et al., 2017; Schalm et al., 2022; Tryner et al., 2021; Zhou et al., 2022), at times combined in an integrated air quality management tool (Merico et al., 2019). Passive dosimeters have also been used (WA Health, 2016). The limitations of sensor technologies in terms of data quality and the concept of "fit for purpose" have been abundantly reported in the literature (Amegah, 2018; Fung et al., 2019; Gerboles et al., 2017; Hofman et al., 2022; Jayaratne et al., 2018; Kang et al., 2021; Li et al., 2022; Malings et al., 2020; WMO, 2018). These tools are advantageous in terms of economic cost, low maintenance and user-friendliness of their interfaces, which are high added values for port authorities tasked with air quality management. Examples of commercial sensor networks can be found in harbours in Estonia (Muuga harbour), Australia (Port of Townsville, Freemantle Ports, Port Hedland), Canada (Ridley Coal Terminals) and Spain (Bilbao and Balearian ports).

In addition to conventional pollutants, air quality degradation in port areas come from specific emission sources which can only be traced using non-regulated parameters such as black carbon (BC), ultrafine particles (UFP) monitored in terms of particle number concentrations (N), volatile organic compounds (VOCs) or even chemical components in PM2.5 and PM10 aerosol fractions. Monitoring of these non-

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conventional air pollution metrics requires high-end instrumentation (e.g., aethalometers, particle counters), typically designed for short-term monitoring and able to provide high-quality real-time data at high temporal resolution.

In order to optimise routine air quality management in environmentally complex port areas, such as the industrial ones, the present work proposes that air quality monitoring should be based on the combination of scientific-grade and cost-effective tools, applied with different temporal strategies in order to provide this innovative strategy for monitoring the particular material in this kind of scenario. As described above, port areas at present frequently lack air quality monitoring strategies due to their excessive cost and/or lack of technical expertise. The novelty of this work is the proposal and testing of an integrated strategy which has the potential to positively contribute to improved air quality management in porta areas. The long-term deployment of cost-effective tools to monitor conventional pollutants, combined with short-term measurements of non-regulated metrics for hotspot identification, would facilitate the simultaneous characterisation of (a) the broad variety of emission sources and air pollutants emitted and (b) their spatio-temporal variability across port areas. The synergies sourcing from combining both approaches would maximise the potential for air quality improvement in port areas. In this regard, the aim of this work was to test the approach proposed in the fully operational industrial port of Castelló (Spain), in the Western Mediterranean basin.

2. Methods

2.1. Study area, monitoring locations and monitoring strategy

The study was carried out in Castelló port (Spain), a fully operational industrial port in the Western Mediterranean (Fig. 1). This port is a major provider of raw materials for the ceramic cluster located in the region, which is the first exporter of ceramic tiles in the EU and the second-largest exporter worldwide. During the study period (2021), the main activities in this port were transport of bulk solids (9.39 million tonnes, the main solid material being: feldspars, clays, kaolin, petroleum coke, clinker, ammonium sulphate and cereal) and bulk liquids (9.9 million tonnes, the main liquid materials being: crude oil, fuel oil, gasoline, diesel and biodiesel oil) (Port Castelló, 2021). The detailed data of the handled solid material is provided in Table S1 (Supplementary Material). From the environmental point of view, the bulk solid handling was of more concern, as it was mostly performed in unconfined facilities, with subsequent impacts of fugitive particulate matter emissions on air quality, while liquids were managed in a fully confined manner. It should be noted that some of the best practices in the handling of solid bulk (Puertos del Estado, 2015) to prevent these emissions were already in place in this port in the study period, such as unloading of materials with higher dustiness in hoppers instead of in piles (Table S1, Supplementary Material), load covering with tarpaulins on trucks or speed limit control and wheel washing systems. In addition to these activities, other potential emission sources in the studied industrial port area were those of the surrounding industrial activities, which mainly comprised a biodiesel manufacturing plant, a petroleum coke storage facility, a clinker milling plant, a gas-fired power station, a refinery, and a chemical plant producing caprolactam and ammonium sulphate.

The study aimed to cover the full port area, which included two main docks (Fig. 2). In total, 8 monitoring locations were selected to characterise the main activities carried out in the port, specifically: powdered materials handling (locations 2,3 and 6), traffic locations (locations 1 and 7), containers and liquid terminals (location 4 and 8, respectively) and a background point (location 5). General information on the main activities typically carried out and potential air pollution sources in the vicinity of each location was provided by the port managers (Table 1). Location 5 was referred to as background because was chosen to determine the background aerosol chemical composition, with the precaution of sampling for this purpose only on weekends. This procedure was established because this site is the furthest away from the bulk loading and unloading areas, and it was mainly influenced by sea breeze and the emissions from ships entering and leaving the port, which were minimal during the weekend studied. Meteorological data were obtained from the local air quality network (Generalitat Valenciana, 2022).

Access to the port area was granted for the present study over a 2-week period (from 12 to 23 July 2021). Based on this time window, the monitoring strategy was defined with two main goals: air pollutant mapping and hotspot characterisation. Air pollutant mapping across the port area was carried out using online monitors (low-cost sensors, see point 2.2) deployed at fixed locations over the full 2-week period, and passive samplers (NO_2) (see point 2.3) deployed at fixed locations over 2 sampling periods (5 workdays/week). In addition, hotspot characterisation was based on the results from high-end instrumentation (see point 2.4), which was deployed subsequently at different locations during 45–60 min periods on 2 different sampling days, following a peripatetic approach (Gillespie et al., 2017; Lin et al., 2017). Peripatetic measurements allow the collection of observations through a monitoring network over a period of time and over relatively large areas with limited equipment, an approach that has been successfully used to monitor air pollution at sequential locations in studies in Canada, Germany, the Netherlands, Spain, Switzerland and the USA (Gillespie et al., 2017). This approach was selected due to the technical complexity of the high-end instrumentation (e.g., inability to be deployed outdoors and be left unattended).

2.2. Low-cost sensors

Low-cost sensors were used to map ambient air $PM_{2.5}$ concentrations across the port area (Fig. 3a). One sensor unit was deployed at each of the 8 monitoring locations (Fig. 2), and they operated continuously for 2 weeks reporting $PM_{2.5}$ concentrations with a 2-min time resolution. The selected sensors operate with PMS-5003 sensing units at a 0.1 l/min air flow driven by an internal fan. While the limitations of sensor technologies are well-known (Jayaratne et al., 2018; WMO, 2018) and were considered in the present work, the performance of this type of sensor has been assessed in previous works (Barkjohn et al., 2020, 2022; Tryner et al., 2020; Wallace et al., 2021) and it was deemed adequate for the purpose of the present study. The particle size targeted was 0.3–100 μm and the concentration range is 0–500 $\mu g/m^3$. Even though the sensors provide readings for PM_{10}, $PM_{2.5}$ and PM_1, in the present study only the $PM_{2.5}$ size fraction was used due to its higher precision when compared to other size fractions (Barkjohn et al., 2020, 2022). The sensing units also include relative humidity, temperature and atmospheric pressure sensors.

The sensors include two PMS-5003 sensing units (A and B) in parallel for quality assurance. Data quality was ensured following the strategy validated in previous works (Malings et al., 2020; Wallace et al., 2021), based on the criterion that $PM_{2.5}$ estimates from the two sensor units within each monitor were required to agree within 30% of each other, corresponding to a precision [abs $(A - B)/(A + B)$] of 0.130 (Wallace et al., 2021). Datapoints with differences >30% between both sensor readings in a node were removed from the dataset (12% of the data were filtered out due to insufficient data quality).

Prior to deployment in the Castelló port area the sensors were intercompared with high-end monitoring instrumentation (GRIMM180 laser spectrometer) by co-location in a reference air quality monitoring station in Barcelona (Palau Reial) during 1.5 months. Subsequently, in order to validate sensor performance when challenged with a similar air pollution mix than in the studied area, the sensors were intercompared once again in an urban background location in Castelló city (at the Institute for Ceramic Technology, ITC) using a Grimm Mini-LAS spectrometer for 48 h. The results of the intercomparisons are provided in Supplementary Material (Figs. S1 and S2).

Fig. 1. Location of the studied area.

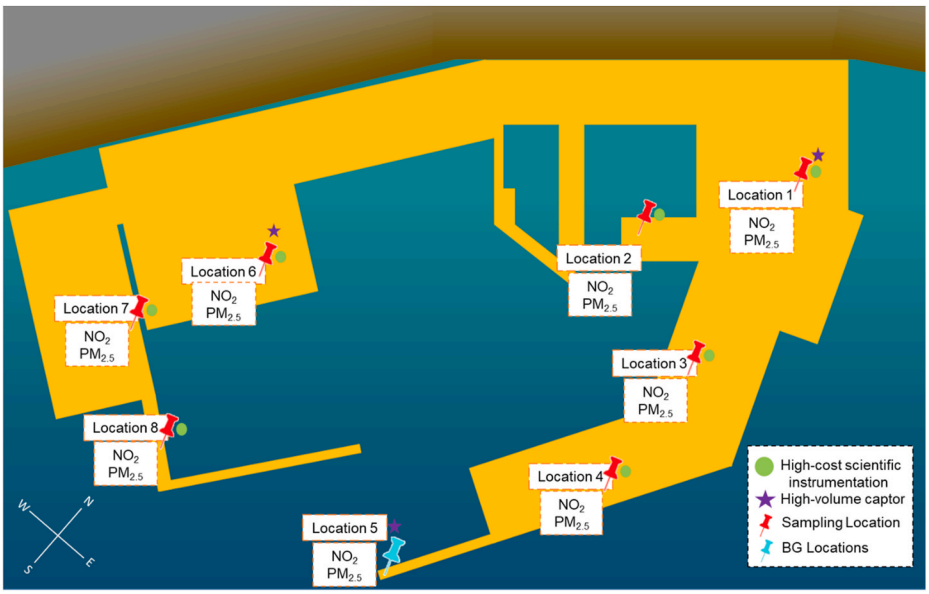

Fig. 2. Map of the Castelló port area (Spain), indicating the monitoring/sampling locations and the pollutants monitored in each of them.

Table 1
Main characteristics of each study location and main activities carried out in their vicinity.

Location	Main activities	Location	Main activities
1	Main entrance/exit to the port	5	Background, major influence from sea spray
2	Bulk solid terminal-North	6	Bulk solid terminal-South
3	Bulk solid terminal-North	7	Transport area nearby the biodiesel facility
4	Container terminal	8	Bulk liquid terminal

2.3. Passive dosimeters for NO_2

A total of 16 dosimeters for NO_2 (Fig. 3a) was deployed at locations 1 to 8 during workdays (Monday to Friday) in two consecutive weeks (8 dosimeters per week). Dosimeters trap NO_2 on a tri-ethanolamine impregnated filter, converting NO_2 into NO_2^-. After exposure, NO_2^- is quantified by ion chromatography (IC). Passive dosimeters have proven to have high accuracy when deployed in urban and/or industrial environments (Lorenzo-Sáez et al., 2021; Yu et al., 2008).

2.4. High-end instrumentation

The following high-end instruments (Fig. 3b) were deployed over 45–60 min periods using a peripatetic approach at each of the 8 monitoring locations, on 2 sampling days (1 in each study week):

- TSI NanoScan (SMPS Model 3910), monitoring fine and ultrafine (UFP) particle number (N) and size distributions from 10 to 420 nm in 13 channels with a 1-min time resolution. This instrument monitors ultrafine particle number concentrations segregated across 13 particle size bins (11.5, 15.4, 20.5, 27.4, 48.7, 64.9, 86.6, 115.5, 154, 205.4, 273.8, 365.2, 420 nm). The data are subsequently processed to produce mean aerosol size distributions.
- Grimm Mini-LAS laser aerosol spectrometer (Mini-LAS 11-R), monitoring total and size-segregated particle mass concentrations between 0.25 and 32 μm (monitoring inhalable (total airborne particles which is inhaled through the nose and mouth), thoracic (PM10) and respirable particles (PM4), PM_{10}, $PM_{2.5}$ and PM_1 concentrations), in 31 channels with a 6-s time resolution.

The NanoScan SMPS and Grimm laser spectrometers were deployed simultaneously at each monitoring location. The main limitations were that the instruments were deployed when access to the port area premises was granted, to avoid interfering with activities in the port, which influenced the time of day when monitoring was carried out, and not enough high-end devices were available to monitor all 8 points simultaneously. As a result, meteorological and port operational conditions were not fully comparable across measurement locations.

Finally, two high-volume samplers (MCV, Spain) operating at 30 m^3/h were deployed over 24-h periods at monitoring location 1 (representative of truck traffic at the entrance of the port) and location 6 (representative point of truck traffic in a loading and unloading zone of the port) during the first and second week, respectively (Fig. 3c). PM_{10} and $PM_{2.5}$ samples were collected at each location, on 15 cm diameter

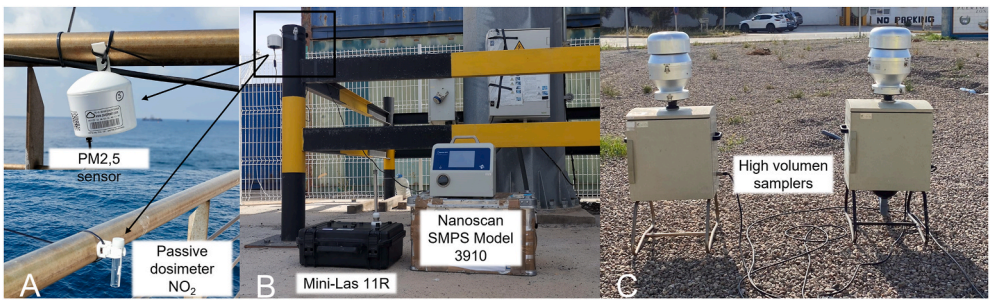

Fig. 3. Sampling devices: A) PM2.5 low cost sensors and NO_2 passive samplers, B) High-end devices, C) High volume samplers.

quartz fibre filters. In total, 4 PM_{10} and 4 $PM_{2.5}$ samples were collected at each location. During the weekend, and in order to characterise background aerosols, the samplers were moved to location 5 (Fig. 2), where 1 PM_{10} and 1 $PM_{2.5}$ 48-h samples were collected. PM mass concentrations were determined by gravimetry under standard conditions (Mettler Toledo AX205). Subsequently, aerosol chemical composition was determined offline in the laboratory by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma atomic emission (ICP-AES) after acid digestion to determine trace and major elements (Querol et al., 2001). The organic and elemental carbon content (OC/EC) was determined using a Sunset OCEC Analyzer.

3. Results and discussion

3.1. Sensor calibration and validation for mapping purposes

Data quality validation and sensor re-calibration were carried out for the 8 individual sensors by comparison with high-end instrumentation (laser spectrometers Grimm180 in Barcelona and MiniLAS in Castelló). Results are shown in Fig. 4. Given that the main aim of monitoring with sensors was to obtain $PM_{2.5}$ concentration maps of the port area, the focus of the comparisons was placed on intra-unit variability. After initial filtering of the data following the data quality checks in the literature (Malings et al., 2020; Wallace et al., 2021), intra-unit comparability was shown to be high with a standard deviation between sensors of <2 μg/m^3 (over a mean concentration of 6.6 μg/m^3) at the Barcelona site (1.5-month duration). During the Castelló (at ITC) intercomparison, with a shorter duration (48 h), the standard deviation between sensor units was <1 μg/m^3 (over a mean concentration of 11.6 μg/m^3). As a result, the comparability between sensor units was validated for the purpose of mapping $PM_{2.5}$ concentrations in the Castelló port area. The individual correlation coefficients are reported in Fig. 4 and Figs. S1 and S1 in Supplementary Material.

3.2. Mapping of $PM_{2.5}$ concentrations

Mean ambient $PM_{2.5}$ concentrations were monitored with sensors at the 8 locations in Fig. 2. In order to highlight the similarities and differences in $PM_{2.5}$ across the port area, concentrations are presented normalised with regard to the average concentrations over the two monitoring weeks (Fig. 5, showing the ratio between the average $PM_{2.5}$ concentrations at each location and the bi-weekly average for all locations, for week 1 and week 2, respectively). The absolute concentrations are reported in Table 2.

The assessment of the normalised values facilitated the identification of location 6 as the most significant $PM_{2.5}$ hotspot, as it consistently showed higher than average concentrations during both monitoring weeks (1.7 on week 1 and 1.2 on week 2). This location is the busiest truck traffic area in the port area, as well as the main loading and unloading area for powder materials. A temporary hotspot was identified during week 1 in location 2, with $PM_{2.5}$ concentrations 1.3 higher than the average while concentrations were lower than average (0.8) during week 2. The berthing information (Table S1) evidenced that the tons of bulk solid material handled were comparable during both weeks (17,000 *vs.* 23,000 tons), and therefore the differences in $PM_{2.5}$ concentrations could be due to differences in meteorological conditions (wind speed and direction; Figs. S3 and S4 in Supplementary Material) and/or other port emissions. The variability across the remaining

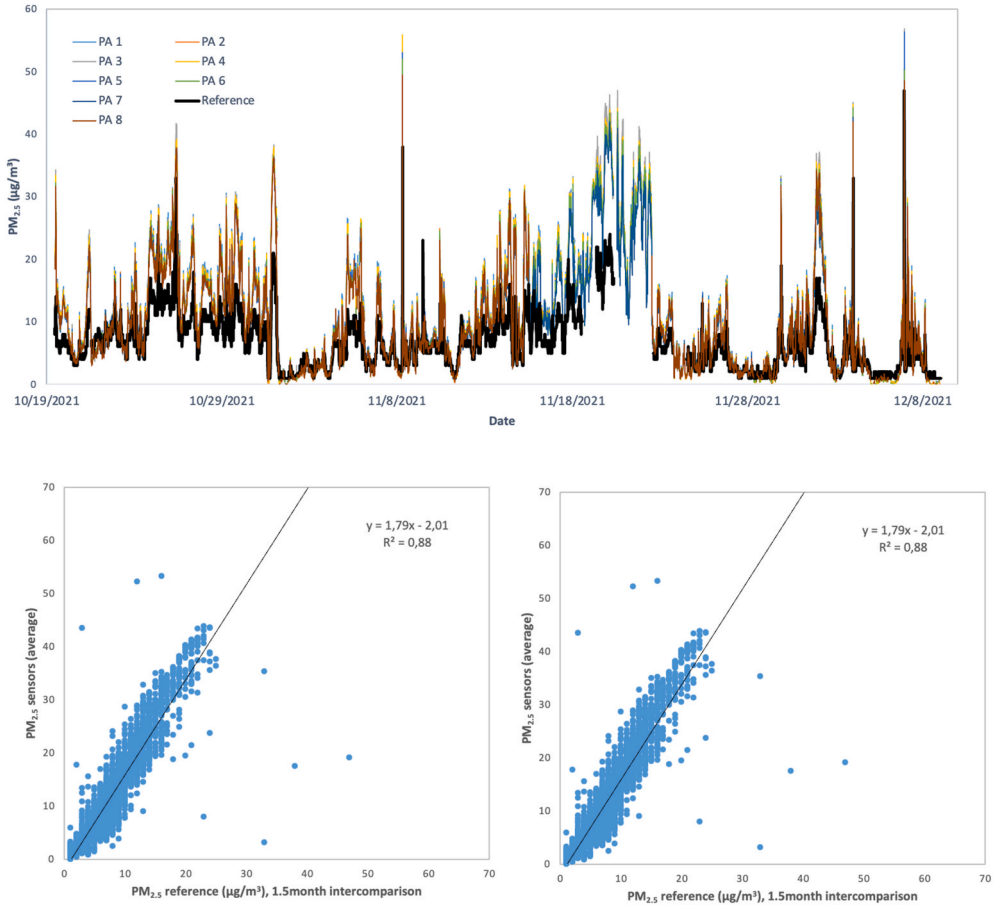

Fig. 4. Intercomparison of the 8 individual sensors with high-end instrumentation (a GRIMM180 laser spectrometer previously calibrated against EU-reference gravimetric data), by co-location in the Palau Reial monitoring station in Barcelona (Spain).

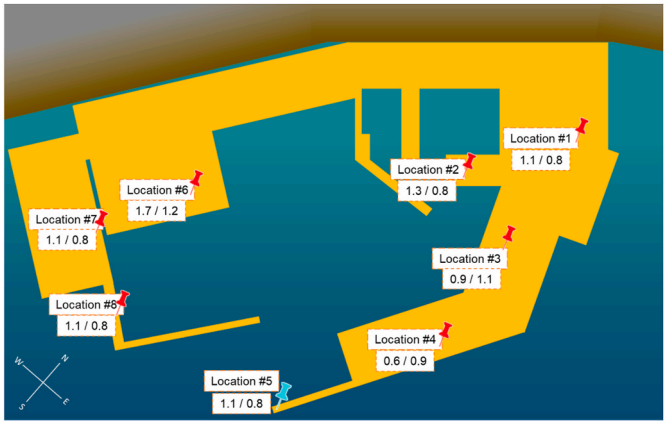

Fig. 5. Map of $PM_{2.5}$ concentrations (recorded with sensors) at the different locations, reported as the ratio between monitored concentrations and average concentrations across locations and over the full period.

locations was relatively low, suggesting that $PM_{2.5}$ concentrations were impacted similarly by the overall background concentrations across the port area. Thus, $PM_{2.5}$ concentrations were not especially sensitive to emissions from handling of bulk solid materials (due to their coarser size distribution), which is one of the main activities in this and many industrial port areas.

3.3. Mapping of NO_2 concentrations

The same approach was applied to NO_2. Results (Fig. 6 and Table 3) evidenced a clear relationship between central area in the port area, with more intense activity in terms of truck traffic, and NO_2 concentrations. The highest ratios were recorded in locations 6 (1.3/1.4 for weeks 1 and 2, respectively), 3 (1.3/1.6), and 2 (1.2 for week 1). Specifically, locations 6 and 3 recorded the highest mean concentrations during both sampling weeks. Location 7 (1.1 during week 1), in the vicinity of the central area, showed slightly higher concentrations than average, whereas locations 4, 5 and 8 recorded lower than average concentrations as they were less influenced by vehicular traffic emissions (furthest from truck traffic and loading and unloading operations). In absolute values, mean 2-weekly NO_2 concentrations ranged between 15 and 46 μg/m^3. The lowest concentration values are slightly higher than the mean annual values registered in the surrounding towns in 2022 (12.5–13.4 μg/m^3, (Borriana and Grau, respectively) (Generalitat Valenciana, 2022), while the highest concentration values may be considered comparable to mean concentration on a high traffic road in an urban area (45.5 μg/m^3 (Amato et al., 2019)). As stated previously, this behaviour can be explained by the intense truck traffic inside the port area, comparable in some locations to traffic in cities larger than the towns surrounding the study area.

3.4. Hotspot characterisation

A further in-depth but short-term analysis was implemented using high-end scientific instruments. The purpose of this analysis was to characterise the particle emissions detected with the aim to identify their potential emission sources, in view of the implementation of targeted mitigation strategies.

3.4.1. Ultrafine particles

Ultrafine particle (UFP) concentrations showed major differences between the measurements collected on both weeks. The highest concentrations were recorded consistently at location 7, with 80,273/cm^3 (mean diameter, Dp = 24 nm) on week 1 and 13,278/cm^3 (Dp = 32 nm) on week 2. While these concentrations may be considered not especially high when compared to other occupational settings (e.g., >10^5/cm^3 in indoor environments in industrial areas; (López et al., 2023; Viitanen et al., 2017), it should be noted that they were monitored in ambient air

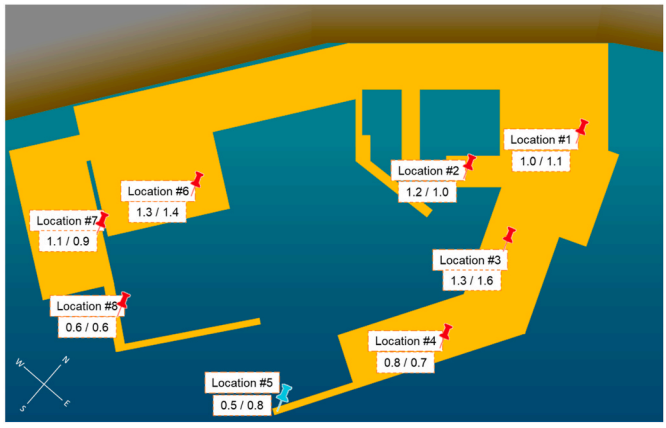

Fig. 6. Map of NO_2 concentrations (measured with dosimeters) at the different locations, reported as the ratio between measured concentrations and average concentrations across locations and over the full period.

Table 2
Mean daily $PM_{2.5}$ concentrations recorded with PA-II-SD sensors at each location, during both weeks, and the ratio between each location and the average for all locations for the full period. StDev.: standard deviation.

	$PM_{2.5}$: Week 1					$PM_{2.5}$: Week 2				
	Average (μg/m^3)	Max. (μg/m^3)	Min. (μg/m^3)	StDev.	$PM_{2.5}$/bi-weekly mean	Average (μg/m^3)	Max. (μg/m^3)	Min. (μg/m^3)	StDev.	$PM_{2.5}$/bi-weekly mean
Location 1	21.3	86.9	3.3	14.5	1.1	15.7	95.3	4.5	11.3	0.8
Location 2	25.9	81.1	3.5	18	1.3	16.3	60.7	4.3	10.6	0.8
Location 3	16.7	64.6	3	10.2	0.9	20.9	60.7	7.7	9.5	1.1
Location 4	12.2	39.5	3	7.9	0.6	16.5	126.7	4.3	12.3	0.9
Location 5	21.9	88	3.4	16.8	1.1	15.1	76.8	4	10.2	0.8
Location 6	32.2	92.5	3	19.9	1.7	23	90.5	4.6	11.2	1.2
Location 7	21.9	72.7	5	14.7	1.1	15.5	59.6	6.2	9.2	0.8
Location 8	20.6	77.9	3.9	14.4	1.1	14.6	73.8	4.6	9.3	0.8

Table 3
Mean weekly NO_2 concentrations measured using passive dosimetry at each location, and the ratio between each location and the average for all locations for the full period.

	NO_2: Week 1		NO_2: Week 2	
	Average (μg/m^3)	NO_2/bi-weekly mean	Average (μg/m^3)	NO_2/bi-weekly mean
Location 1	30.9	1.0	34.7	1.1
Location 2	37.9	1.2	30.2	1.0
Location 3	41.2	1.3	50.4	1.6
Location 4	23.6	0.8	23.5	0.7
Location 5	14.7	0.5	25.9	0.8
Location 6	40.9	1.3	42.9	1.4
Location 7	33.9	1.1	29.5	0.9
Location 8	19.5	0.6	18.2	0.6

in the port area. The concentrations monitored are, in contrast, much higher than the mean annual concentrations in typical urban locations under the influence of traffic emissions worldwide (8.0×10^3/cm^3 to 19.5×10^3/cm^3; (de Jesus et al., 2019). The particle size distribution for this location (Fig. 7) evidenced high concentrations of <60 nm aerosols during the full monitoring periods and in both weeks, in addition to slightly coarser aerosols (100–300 nm) during the first week. This suggested the presence of a continuous emission source of UFP in the area, which could be at least partly related a biodiesel production plant located in the port area (Brahma et al., 2022; Caruso et al., 2015; Motevali et al., 2023) although other contributions (Section 2.1) cannot be ruled out. Volatile organic compound (VOC) emissions and their subsequent nucleation were identified as a possible mechanism of UFP generation in the area.

In addition to location 7, three other monitoring location showed markedly high UFP levels during week 2: locations 1, 2 and 6, all of them reporting UFP concentrations >10,000/cm^3 (Table 4). In all 3 cases (Figs. 5 and S5 in Supplementary Material), the aerosol size distribution showed the prevalence of particles <100 nm on average, and <60 nm during most of the monitoring period, suggesting vehicular emissions as a probable source. While location 6 had been previously identified through the $PM_{2.5}$ and NO_2 mapping as being strongly impacted by truck traffic (for loading and unloading operations), location 1 was the main entrance/exit point to the port and was thus also under the influence of traffic emissions. As shown in Fig. 2, location 2 was found in the vicinity of this exit and was also influenced by vehicular traffic inside the port area (with high $PM_{2.5}$ and NO_2 concentrations, Figs. 3 and 4). Figs. 5 and S6 highlight the clearly different aerosol size distributions monitored in the different locations (e.g., locations 1 and 8) during both periods, showing that the variability in UFP emissions across different days was quite significant and, consequently, that there is a need for monitoring this type of emissions with high-end instrumentation to better understand the emission patterns.

3.4.2. PM10 aerosols

A further analysis was carried out aiming to understand the air quality impacts of handling of powdered materials as one of the key activities in the studied port area. Results from targeted PM_{10} monitoring (Fig. 8), shown as the ratio for each location with regard to the mean, provided a much clearer view of the coarse aerosol hotspot areas across the industrial port. These were locations 3, 6 and 2, where concentrations were 3.6, 1.5 and 1.2 times higher than the port average, respectively (Fig. 8 and Table 5). Fig. 8 evidences the large potential of the targeted measurements collected during this work, as they provided valuable information for action by the port authorities despite the high-cost and technical complexity of the high-end instrumentation used. In terms of the metrics used, PM_{10} seemed to be a more sensitive parameter to characterise the emissions from port activities such as handling of bulk solid materials, in contrast to $PM_{2.5}$.

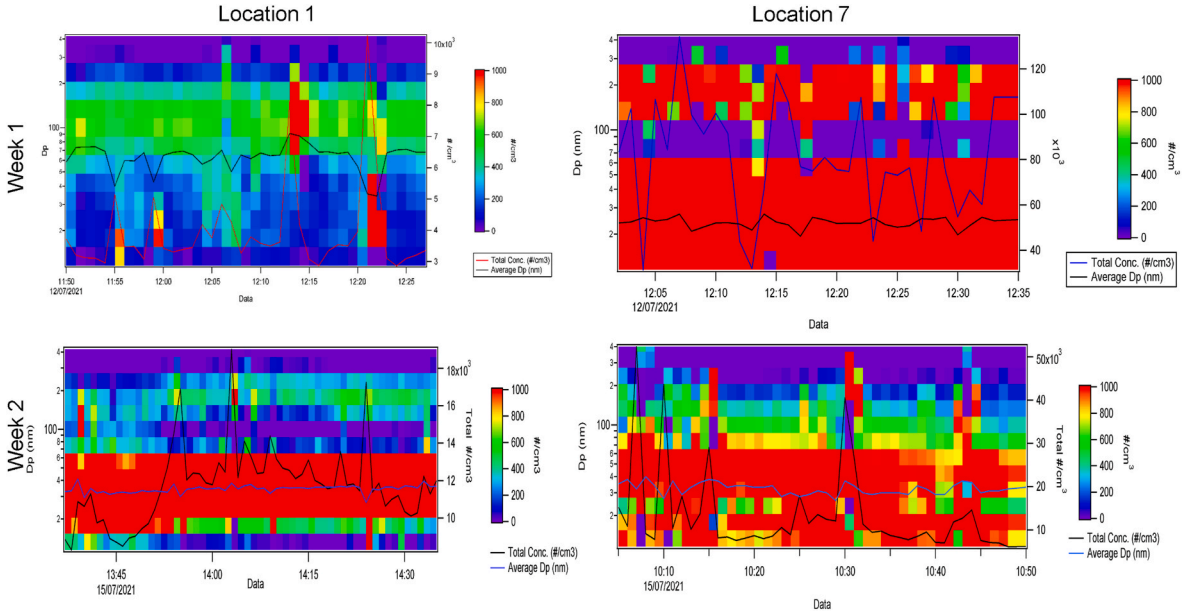

Fig. 7. Particle size distribution (range 20–420 nm) monitored during 45–60 min at locations 1 and 7 in weeks 1 and 2.

Table 4
Mean and maximum ultrafine particle (UFP) concentrations (particle number concentration, PN) and mean diameter (Dp) monitored using a TSI Nanoscan-SMPS during 45–60 min periods, at different locations. StDev.: standard deviation.

	UFP: Week 1						UFP: Week 2					
	Average		Max.		StDev		Average		Max.		StDev	
	PN (#/cm^3)	Dp (nm)	PN (#/cm^3)	Dp (nm)	PN (#/cm^3)	Dp (nm)	PN (#/cm^3)	Dp (nm)	PN (#/cm^3)	Dp (nm)	PN (#/cm^3)	Dp (nm)
Location 1	3924	64	10,216	91	1443	12	11,750	34	19,015	41	2054	2
Location 2	3297	69	4997	91	610	12	10,860	37	33,398	49	6333	4
Location 3	4604	71	11,136	93	1898	10	7029	37	15,063	44	2941	4
Location 4	4967	63	8800	71	1210	5	6229	32	8263	46	1056	5
Location 5	7354	55	9757	62	1059	4	4226	29	5178	34	636	3
Location 6	3880	70	7057	76	650	6	11,320	39	28,956	87	5290	10
Location 7	80,273	24	134,134	27	25,041	2	13,278	32	52,334	40	10,056	3
Location 8	3517	71	3963	74	192	2	4920	31	6732	35	685	2

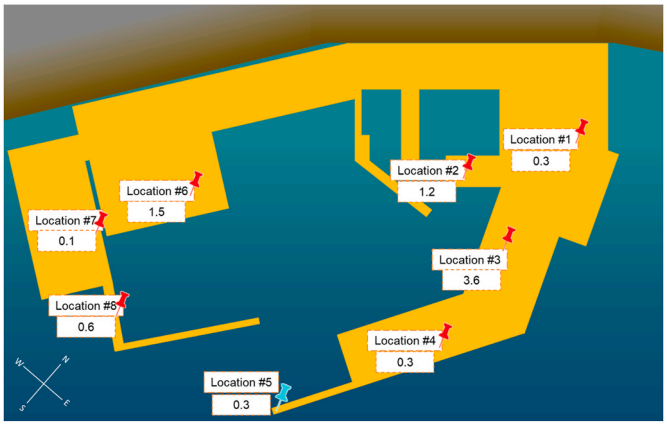

Fig. 8. Map of PM_{10} concentrations (measured by laser spectrometry) at the different locations, reported as the ratio between monitored concentrations and average concentrations across locations and over the full period.

Table 5
Mean and maximum PM_{10} concentrations monitored using a Grimm Mini-LAS spectrometer during 45–60 min periods, at different locations. StDev.: standard deviation.

	PM_{10} (μg/m^3): 12-07-2021			
	Average	Max.	Min.	StDev.
Location 1	37.1	85.0	16.4	16.0
Location 2	131.6	368.1	44.4	75.8
Location 3	391.8	1194.1	120.8	254.9
Location 4	28.4	135.4	13.0	21.5
Location 5	30.4	272.9	11.9	43.2
Location 6	158.7	828.2	11.5	145.0
Location 7	14.1	25.7	10.3	3.8
Location 8	67.0	446.1	15.2	95.2

3.4.3. Chemical tracers

Finally, filter samples of PM_{10} and $PM_{2.5}$ aerosols were selectively collected in the port area: locations 1 and 6 representing in-port emissions during workdays, and location 5 representing background aerosols during the weekend. High-volume samplers were only available at a limited number of locations due to instrument availability and logistical complexity regarding instrument setup. Because the number of samples was certainly limited (2 × 24-h samples for PM_{10} and 2 × 24-h samples for $PM_{2.5}$ in locations 1 and 6, respectively, and 1 × 48-h sample for PM_{10} and 1 for $PM_{2.5}$, for location 5), the results presented in this section should be considered only indicative.

The mean relative (%) chemical composition of aerosols sampled at the 3 locations is summarised in Fig. 9, for major (top) and trace (bottom) components. In terms of major inorganic components, results evidenced relative similarities between locations 1 and 6 which clearly differed from the results obtained for background aerosols (BG, location 5) which were strongly dominated by Na as tracer of marine aerosol (Na; >50% of major components in PM_{10} and $PM_{2.5}$, in comparison to <30% in locations 1 and 6). Conversely, major contributors to coarse and fine particles in areas impacted by emissions form handling of bulk solids (locations 1 and 6) were Al (27–44%) and Ca (19–28%), probably resulting from solid bulk materials rich in Al (mainly clays and feldspars) and Ca (mainly clinker and anhydrite) respectively, which were handled in the port during the study period (Table S1 and Table S2 in Supplementary Material).

Trace elements, while still showing major differences between in-port emissions (locations 1 and 6) and background aerosols (location 5), presented a larger variability than major components (Fig. 9, Fig. S7 in Supplementary Material). Ti (25–57% and 70–72% of trace element contributions in PM_{10} and $PM_{2.5}$ in locations 1 and 6, respectively) was highlighted as a key tracer of this port loading and unloading operations, which is consistent with the type of materials (ceramic raw materials; Tables S1 and S2) handled in the studied port (in clays the Ti according to Celades (2013), Minguillón et al. (2013)) is the highest trace element) and the fact that the concentrations in location 6, more influenced by material handling, were higher than in location 1 (more influenced by traffic).

Present in lower proportions, but also known tracers of road traffic (Zn, Cu and Ba) (Johansson et al., 2009; Querol et al., 2004; Viana et al., 2008), and to some extent of ceramic raw materials (Table S2) (Celades et al., 2022; Minguillón et al., 2013) were Zn (4–30% of trace components), Ba (5–13%), Sr (2–7%) and Cu (4–11%), which showed higher levels in $PM_{2.5}$ in location 1, more influenced by traffic. This suggests that the in-port traffic of trucks and shovels was the main source of these elements (Isakson et al., 2001).

Finally, similar results were obtained for carbonaceous aerosols (Table S3 in Supplementary Material): organic (OC) and elemental carbon (EC) concentrations were always higher in-port when compared to background aerosols, for both size fractions (ranging between 1.65 and 4.28 μgOC/m^3 and 0.48–1.11 μgEC/m^3 in $PM_{2.5}$ in-port vs. 1.24 μgOC/m^3 and 0.17 μgEC/m^3 in $PM_{2.5}$ in the background). In addition, the higher OC concentrations recorded in location 6 in fine and coarse aerosols (e.g., 4.26 μgOC/m^3 in $PM_{2.5}$) in comparison to location 1 (1.65 μgOC/m^3) were consistent with handling of a corn shipment in the former docking area (Table S1). As a result, in spite of the limited number of samples available, the data obtained on organic aerosols contributed to the identification of a specific shipment (in this case, of corn) as a key contributor to higher UFP levels. Therefore, this example clearly shows how this methodology can be used to characterise the impact of specific operations, as well as to propose and evaluate preventive measures.

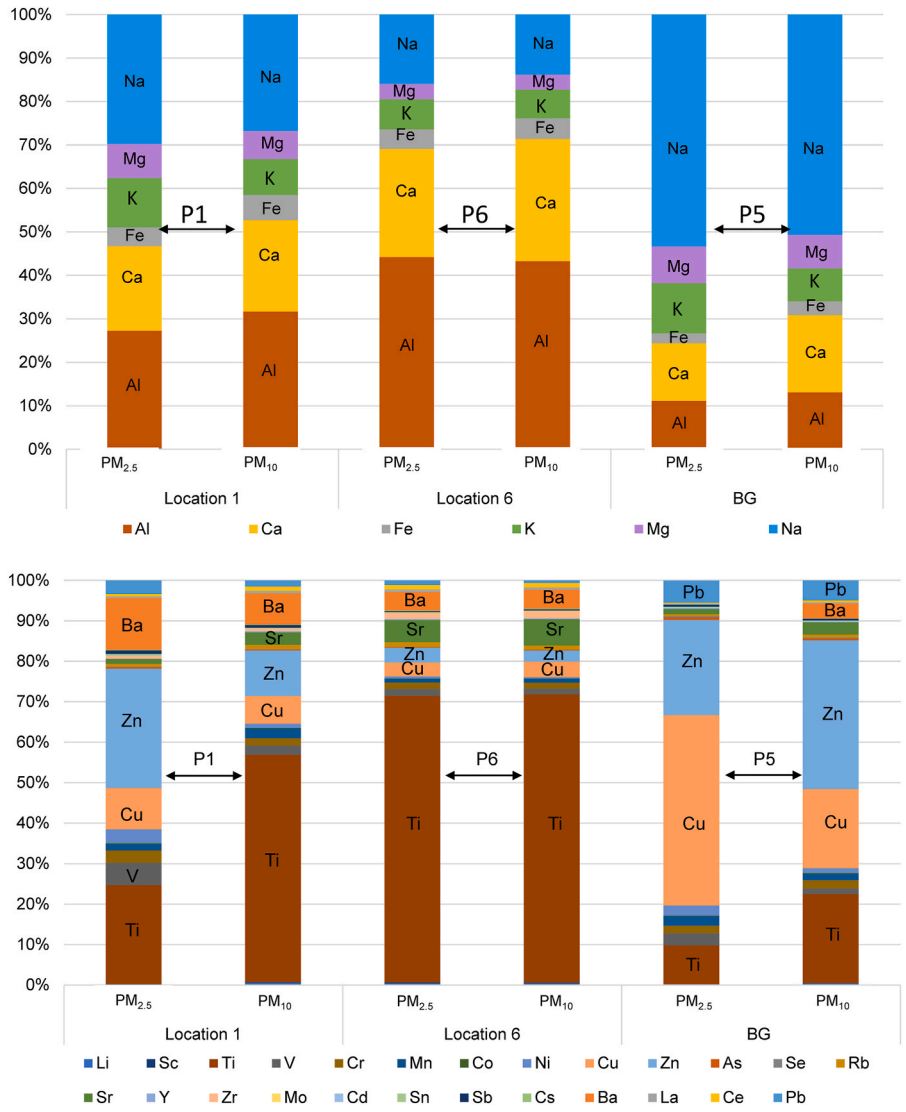

Fig. 9. Mean relative (%) chemical composition of PM_{10} and $PM_{2.5}$ aerosols sampled at locations 1, 6 and 5: major (top) and trace (bottom) components.

3.5. Discussion

This work proposed an integrated strategy for routine air quality monitoring in ports, which was tested in a fully operational industrial port area in Spain with the following outcomes:

- The combination of low-cost air quality sensors and passive dosimeters with the analyses of the activities performed in the port area provided valuable information to generate air pollution maps across the port area, which facilitated identification of hotspots (e.g., diesel combustion emissions from docked ships, cranes, trucks and shovels) in terms of $PM_{2.5}$ (location 6, and location 2 during week 1) and NO_2 (locations 3 and 6, and 2 during week 1). These maps, previously unavailable for the Castelló port managers, may be used to design mitigation strategies to minimise air quality impacts of these activities, for example by implementing automatization of these operations (for instance the use of conveyor belts instead of trucks), machinery electrification, optimising truck traffic across different routes or times of the day to avoid accumulation in locations 2, 3 and 6. Similar mapping approaches have been reported in the literature (Beloconi et al., 2016; Chu et al., 2020).

The high-end scientific instrumentation, subsequently deployed for short-term monitoring at target locations, facilitated source identification. For example, the analysis of particle size distributions highlighted a nearby biodiesel facility as a possible source of elevated particle number concentrations (up to 80,000/cm^3) recorded at location 7. The particle number concentrations and size distribution (D_p = 24 nm) registered suggested the influence of nucleation processes from VOC emissions from the plant, and/or of combustion aerosols. In addition, high OC concentrations at location 6 suggested the impact of a specific shipment (e.g., corn) on air quality in the area. Once again, dedicated actions can only be undertaken once specific emission sources are identified, as was the case with the high-end instrumentation in this work.

- PM_{10} concentrations were a more sensitive marker of bulk solid emissions, in comparison to $PM_{2.5}$, as it was shown in location 3. The comparison between PM_{10} aerosols across the studied area provided a clearer indication of the emission hotspots associated to powdered materials handling than $PM_{2.5}$. As a result, PM_{10} could be recommended as a relevant metric for monitoring to the port authorities, either using long-term or short-term monitoring approaches. Previous studies (Ribalta et al., 2018, 2019) also identified PM10 as a more adequate tracer of bulk emissions than PM2.5, in the case of indoor industrial environments (specifically, during handling of dusty raw materials).

- Trace element analysis of PM_{10} and $PM_{2.5}$ aerosols provided valuable information for source identification, although the longer time necessary to obtain results (due to the need for analysis in laboratory) renders this metric less valuable for port authorities, in the short-term. However, although it is not considered necessary for routine monitoring, periodic characterisation of chemical tracers is recommended, at least those included in air quality legislation, or when substantial changes are implemented in port activity (materials, fuels, etc.).

As a result, the approach proposed presented a number of actionable outputs and advantages: i) access of port managers to high-quality scientific data, ii) cost reduction, iii) emission hotspot and source identification (on-road traffic, unloading of corn shipment, VOCs from biodiesel facility), and iv) identification of a dedicated metric (PM_{10}) sensitive to the air quality impacts in this specific port. These outcomes may be considered sufficiently specific in terms of emission sources and their locations for the port managers to design and implement effective mitigation strategies. However, the approach proposed also suffers from a number of limitations which should be balanced against the advantages discussed: i) low data quality of sensor technologies, which should always undergo the necessary QA/QC procedures (e.g., local calibration and assessment of drifts over time), ii) short duration of the peripatetic monitoring with high-end instruments, which is an intrinsic limitation of this approach, and iii) lack of annual representativity of the data, given the short-term measurements proposed (2 weeks), which can only be overcome by repeating the monitoring if a seasonal variation of emission sources is expected. In sum, current trends on air quality monitoring in complex port areas seem to follow mainly two different approaches: they are either research-oriented (focusing on scientific instrumentation and monitoring strategies; Genga et al., 2017; He et al., 2023; Kwon et al., 2023; Song et al., 2022, among many others), or application-driven (with a focus on the use of results by port and city air quality managers; e.g. among others, Casazza et al., 2019; Merico et al., 2019). To the authors' knowledge, the results presented evidence that the strategy proposed innovates by building on the advantages of each of these approaches and improving the capacity of port managers to mitigate air quality impacts from port activities.

4. Conclusions

An integrated monitoring strategy is proposed to contribute to routine air quality management in industrial port areas. The strategy is based on the combination of scientific-grade instrumentation, deployed following a peripatetic approach, and cost-effective tools deployed longer-term. These different temporal approaches take advantage of the strengths of each of these types of instrumentation: while large-scale data can be obtained from cost-effective tools for mapping purposes, the high-end instrumentation provides robust air pollutant metrics for hotspot characterisation and subsequent management.

Results evidenced that the strategy was able to provide actionable results for air quality management by port authorities, while (a) reducing its cost through cost-effective instrumentation, and (b) generating high-quality scientific results using a streamlined, short-term approach. Specifically, for the port of Castelló, this approach was able to report mean PM2.5 concentrations up to 32.2 $\mu g/m^3$ in the main hotspot (location 6) while in the other locations the concentrations ranged between 12.2 and 25.9 $\mu g/m^3$. Similarly, ultrafine particle number concentrations were highest (80,273 $\#/cm^3$) at the location 7 impacted by nearby VOC emissions, whereas other locations concentrations ranged between 3297 and 11,320 $\#/cm^3$. Key tracers identified for raw materials handled in the port were Ti, Cu, Zn, Al, Ca.

While this approach builds on the advantages of high-end and cost-effective instrumentation, the limitations of both types of monitors should also be acknowledged: these are (a) low data quality of low-cost sensors, if not adequately calibrated, and (b) high cost and technical complexity of high-end instruments including challenging deployment in industrial scenarios. The latter resulted in short time series from the high-end monitors, which should be considered a limitation of this work. The synergies sourcing from combining both approaches could maximise the potential for air quality improvement in complex port areas.

The proposed strategy was tested and validated in an industrial port area in Spain, which includes an industrial bulk port, and an industrial estate with different chemical industries, but can be useful for other environmentally complex port areas, such as other industrial ports or large terminals of any type, such as passenger or cargo.

CRediT authorship contribution statement

María López: Investigation, Methodology, Writing – original draft. **Clara Giner-Cifre:** Data curation, Formal analysis, Methodology. **Ana López-Lilao:** Data curation, Formal analysis, Writing – review & editing. **Vicenta Sanfélix:** Data curation, Methodology, Writing – original draft. **Eliseo Monfort:** Conceptualization, Funding acquisition, Project administration, Supervision, Validation, Writing – original draft, Writing – review & editing. **Mar Viana:** Conceptualization, Funding acquisition, Project administration, Supervision, Validation, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors 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

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