

Contribution of maritime traffic to concentrations of airborne particulates

Delivered from the results of the project
ECOMOBILITY

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1. INTRODUCTION

The atmospheric impacts of shipping at regional scale, especially on coastal cities, need an increasing knowledge because of direct and indirect effects of harbour traffic and related logistic activities. This is particularly true in the Adriatic-Ionian area where an intense traffic of commercial, tourist and cruise ships affecting air quality in coastal is present (Muntean et al., 2019).

Most of the available studies characterise the impacts of shipping to air quality (gaseous and particulate pollutants) by means of large spatial scale chemical transport models, of local spatial scale dispersion modelling, of receptor models based on chemical composition (mainly for particulate matter), of statistical analysis approaches based on high temporal resolution concentration measurements. Furthermore, specific studies in port-cities of the Adriatic-Ionian area investigating the impact of shipping to particles of different sizes are extremely scarce (Merico et al., 2016). This is particularly true for the east Adriatic coast, with evaluation of maritime traffic on environment is at the very beginning, with hardly any data.

The project ECOMOBILITY tries to fill some of these gaps applying the methodologies (Merico et al., 2017) based on both chemical characterization of particulate matter and high-temporal resolution online measurements, used in the POSEIDON project (Pollution Monitoring of ship emissions: an Integrated approach for harbours in the Adriatic basin, Interreg MED program 2007-2013) that is capitalized in ECOMOBILITY, to characterize the impact of ship traffic to particulate matter of different size starting from nanoparticles and up to PM₁₀. The same approach and the same measurement instruments have been used in the two areas (Venice and Rijeka) to ensure comparability of results.

2. METHODOLOGY USED TO ELABORATE HIGH TEMPORAL RESOLUTION DATA

High temporal resolution data have been collected at 1 minute resolution using a Condensation Particle Counter (CPC Grimm 5.403), to measure the total number concentration of particles in the size range between 0.01 – 0.25 µm, and an Optical Particle Counter (Grimm 11-A) able to measure particle number size distributions in the size range 0.25-31 µm in 31 size channels. Additional meteorological measurements are also taken at the same temporal-resolution. A full description of the instruments used is reported in the deliverable “Analysis report in Venice”. The same instrumental setup was used in both areas (Venice and Rijeka). The dataset collected in each site allows to identify concentration peaks associated to the plumes of the ships (Fig. 2.1). This makes possible to statistically investigate the influence of these peaks to measured concentrations.

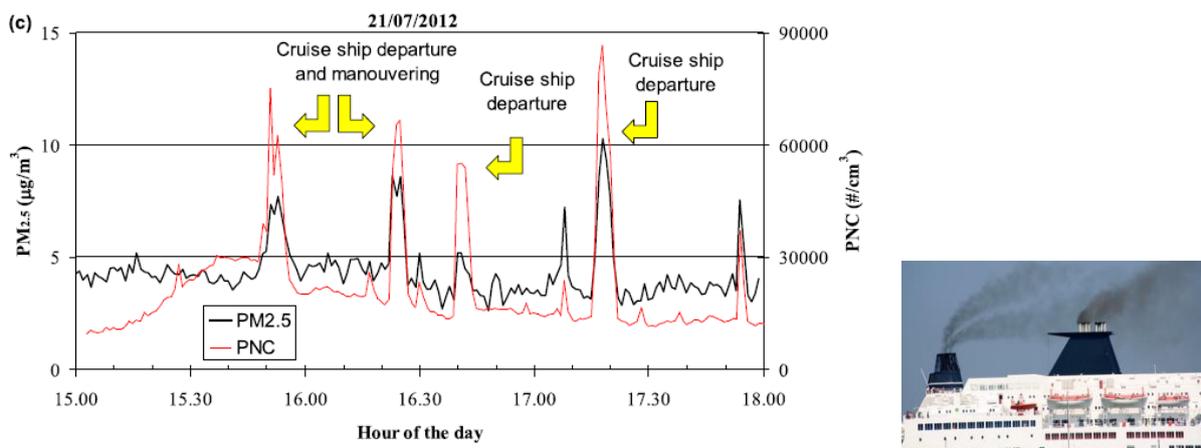


Figure 2.1. PM2.5 and total particle number concentration (PNC) at 1-min resolution with indication of ship arrival and departure. Obtained from Contini et al. (2015).

Data is post-processed on 30-minutes average and combined with the information relative to ship traffic to evaluate the relative contribution (ϵ) to particle concentrations in each size range. The evaluation of ϵ could be due using the formula initially developed in Contini et al. (2011):

$$\epsilon = \frac{(C_{DP} - C_{DSP})F_P}{C_D} = \frac{\Delta_P F_P}{C_D} \quad \text{Eq. 2.1}$$

where: C_{DP} is the average concentrations in the selected wind direction sector (in which the site is downwind of the emissions) considering periods potentially influenced by ship emissions; C_{DSP} is the average concentrations not significantly influenced by ship emissions; C_D is the average concentration in the specific wind direction sector and F_p is the fraction of cases influenced by ship emissions.

3. METHODOLOGY USED TO ELABORATE SIZE-SEGREGATED CHEMICAL DATA

Weekly samples have been collected in both areas (Venice and Rijeka) using a model 110 MOUDI cascade impactor that allowed to collect particulate matter in 12 different size ranges on quartz fiber filters. The filters have been chemically analysed as described in the deliverables “Analysis report in Venice” and “Analysis report in Rijeka” to determine the content of carbon, major water soluble ions and metals.

V and Ni have been used as tracers of ship emission, but this approach was abandoned since these metals are emitted from other sources, as well. Therefore, the contribution of ship emissions to primary PM was extracted considering the V as a marker for the combustion in ships’ engines as it was done in Zhao et al. (2013). The primary contribution PM_{ship} of ship emissions to the atmospheric PM was calculated using the formula:

$$PM_{ship} = R * \frac{V}{F_{V,HFO}} \quad (\text{Eq. 3.1})$$

where R equals 8205.8 (Agrawal et al., 2009), a value internationally applied for locations with HFO-burning ship emissions; V is the in-situ ambient concentration of Vanadium (ng/m^3); $F_{V,HFO}$ is the typical V content (ppm) in HFOs used by vessels; in the absence of chemical analyses of fuel, the value of 65 ± 25 ppm was used to cover the typical range of $F_{V,HFO}$ (Cesari et al., 2014). The obtained primary contribution was converted in percentage dividing for the PM corresponding concentration.

4. DATA PREVIOUSLY AVAILABLE IN VENICE AND RIJEKA

The available information on the impact of ship traffic in the two areas (Venice and Rijeka) were collected during POSEIDON project and other publications available in the scientific literature. The weight of shipping emissions compared to road traffic for year 2010 is reported in Fig. 4.1 (taken from Merico et al., 2017). It shows that shipping has a non-negligible contribution comparable to that of road traffic and even larger for SO₂.

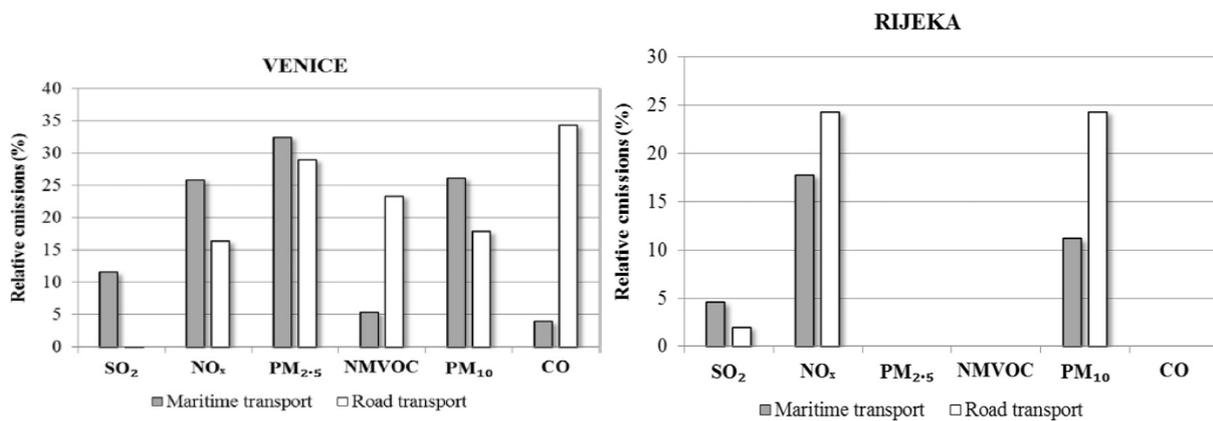


Figure 4.1. Maritime and road transport emissions for specific pollutants in Venice and Rijeka in 2010. Taken from Merico et al. (2017). Data are presented as percentage of total emission in the Municipality.

Other publications focused on impact of shipping to air quality in coastal towns of the Adriatic-Ionian macro-area and the results of the POSEIDON project, that is capitalized in ECOMOBILITY, are summarized in Fig. 4.2. These results indicate that impact to total particle number concentration (PNC) is significantly larger than that to mass concentrations (PM_{2.5}, and PM₁₀) and that there are not detailed information of the impact of shipping on size-segregated particles for this area.

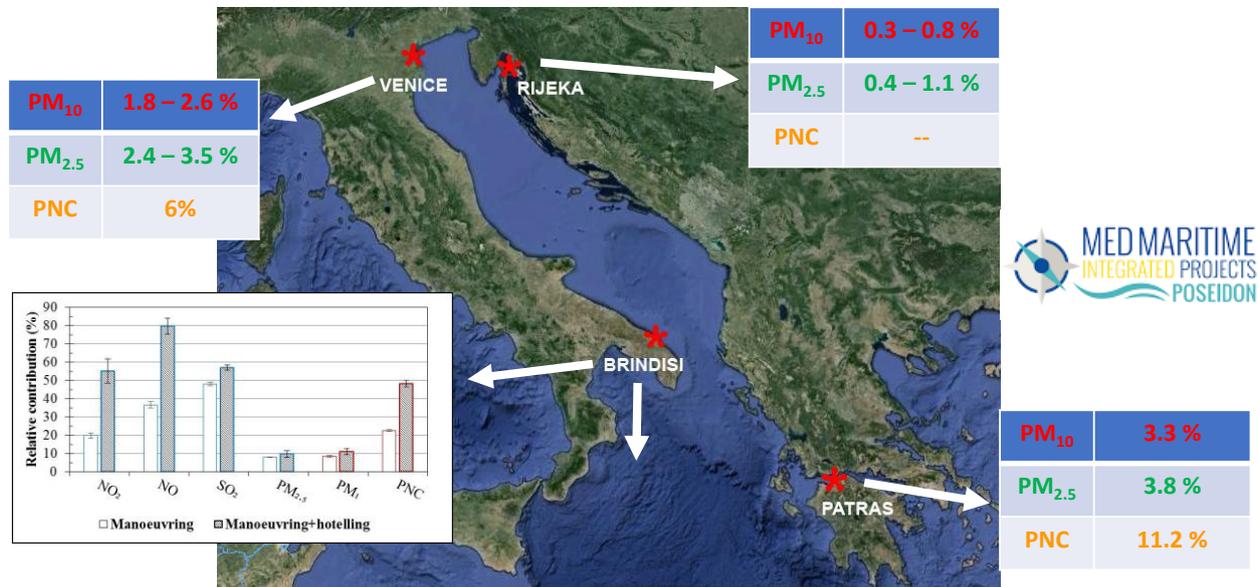


Figure 4.2. Average relative impact of shipping to atmospheric particle concentrations in different sites of the Adriatic-Ionian macroarea.

5. DISCUSSION OF RESULTS

This chapter will be divided in two sections according to the methods described in Chapter 3:

- High temporal resolution of particulate number (PNC) and mass concentrations data of airborne particulates
- Chemical analyses of vanadium in different airborne particulate fractions

5.1. Impact of high temporal resolution measurements.

The approach discussed in Chapter 3 has been applied to the data collected at high temporal resolution at the measurement site (located on the roof of the Health Teaching Institute of the University of Rijeka, 45°19'55.58''N – 14°25'32.84''E) in Rijeka between 28/03/2019 and 13/05/2019. The average size distributions in number and in mass obtained combining the measurements of the CPC and OPC are shown in Fig. 5.10. It is possible to identify the same three size ranges already identified in Venice for further analysis of number concentrations:

nanoparticles ($D < 0.25 \mu\text{m}$); fine particles ($0.25 < D < 1 \mu\text{m}$); coarse particles ($D > 1 \mu\text{m}$). The concentration in mass will be analysed in the standard size ranges PM_{10} , $\text{PM}_{2.5}$, and PM_{10} .

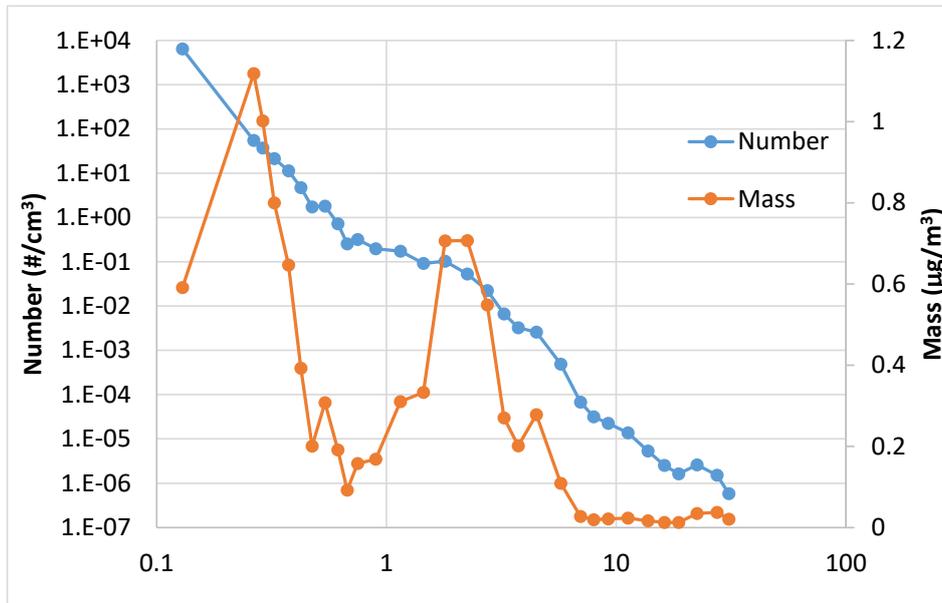


Figure 5.10. Average particle size distribution in number and in mass in Rijeka.

The data of ship traffic furnished by Rijeka Port Authority for the period of the campaign included 92 ships. The arrival and departure times were synchronised with concentration measurements using information obtained from the video-camera. Ship traffic does not have a clear daily pattern (Fig. 5.11). The measurement site (Fig. 5.12) is downwind of emissions for wind directions in the range 122.5° - 180° (hoteling of ships) and 122.5° - 247.5° during manoeuvring of ships. The daily pattern of the percentage of time in which the site is downwind of ship emissions is reported in Fig. 5.13. It indicates that the site is potentially influenced by ships during diurnal hours and this means by the majority of ship traffic.

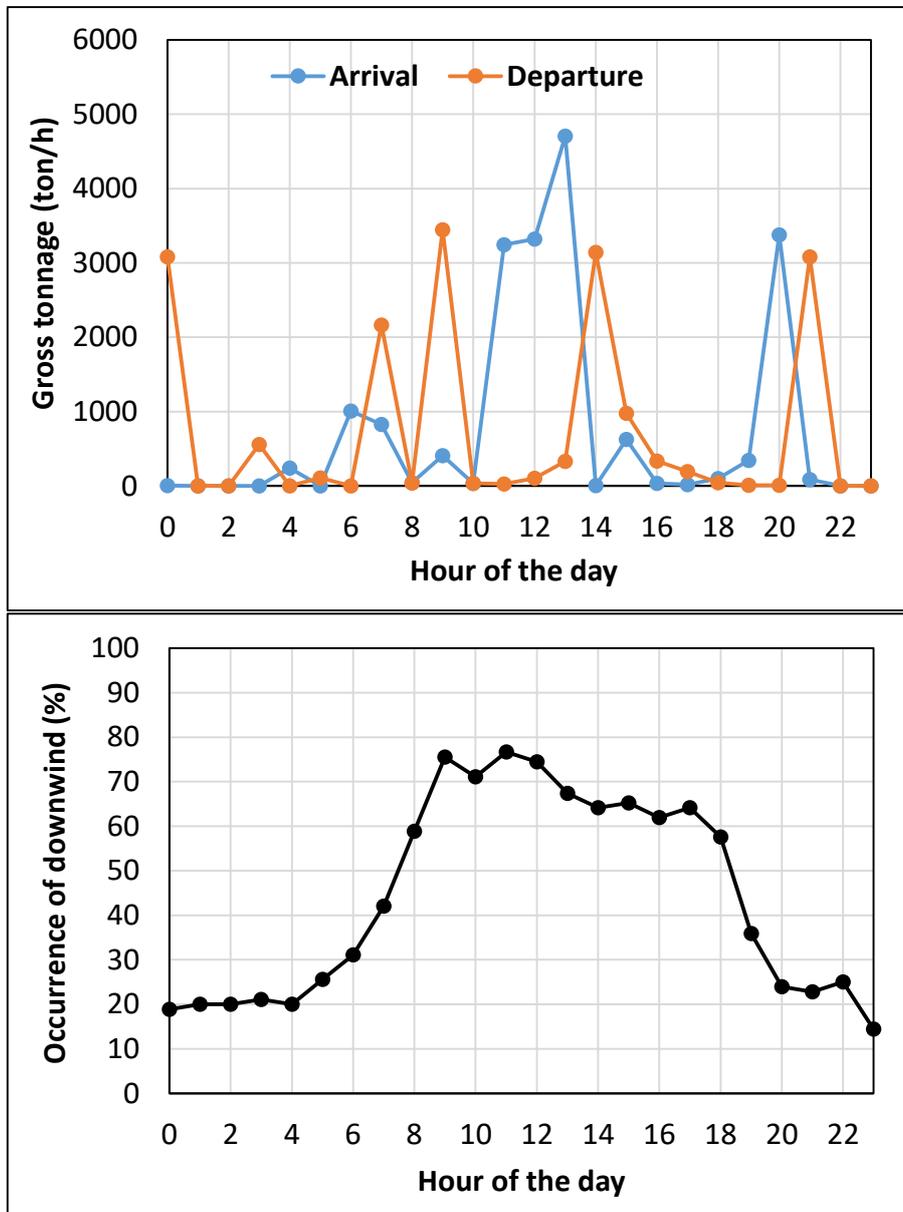


Figure 5.11) Daily trend of ship traffic (top) and daily trend of the percentage of time in which the site is downwind of the emissions located in the harbour area in Rijeka.



Figure 5.12: Measurement site in Rijeka with indication of the angles used in post-processing.

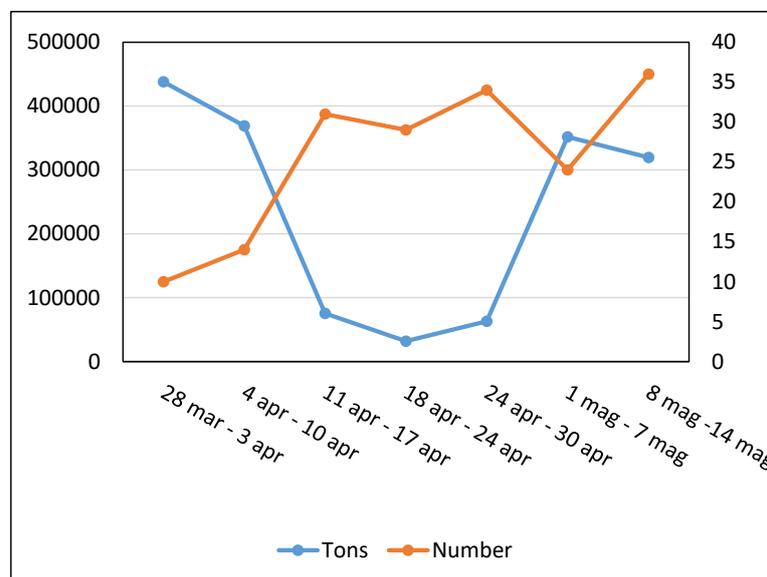


Figure 5.13: Weekly trend of number and gross tonnage of ships calling (sum of arrival and departures) at the harbour of Rijeka.

The average and standard deviation of particle concentrations measured for the different size ranges analysed in mass and in number are reported in Table 5.2 with their standard deviations. The concentration values are smaller than those found in Venice (Table 5.1) for all size ranges.

	PM ₁ (µg/m ³)	PM _{2.5} (µg/m ³)	PM ₁₀ (µg/m ³)	Nanoparticles (#/cm ³)	Accumulation (#/cm ³)	Coarse (#/cm ³)
Average	11.4	13.4	14.8	6552	134.1	0.45
STD	6.9	8.8	10.4	3738	90.4	0.89

Table 5.2: Averages and standard deviations of mass and number concentrations of particles for the different size ranges analysed in Rijeka.

The formulae discussed in Chapter 3 have been used to investigate the impact of shipping to measured concentrations. These have been applied on a subset of data in which it has been removed the period between 24 and 26 April 2019 because of an intense event of African dust advection observed. This event will change the balance between fine and coarse particles introducing a confounding effect on the determination of the impact of shipping.

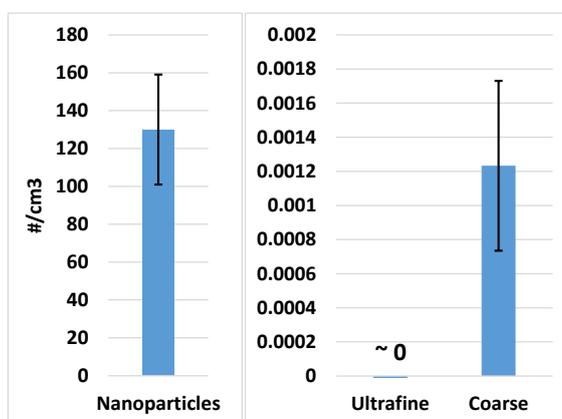


Figure 5.14: Absolute contributions of ships to measured concentrations (in number) in Rijeka. Contributions to PM₁, PM_{2.5}, and PM₁₀ are negligible and not shown.

Absolute contribution of ships to measured concentrations in the different size ranges is reported in Fig. 5.14. It is observed that contributions to mass concentrations are essentially negligible and not discernible among the different sources acting on the site studied. The same apply for ultrafine particles, instead, a non-negligible contribution is observed for coarse particles in number. Relative contribution to nanoparticles is about 1.8% (Fig. 5.15) and a measurable (about

0.5%) contribution to coarse particles was observed. In all the other cases, the relative contributions were negligible (< 0.2%).

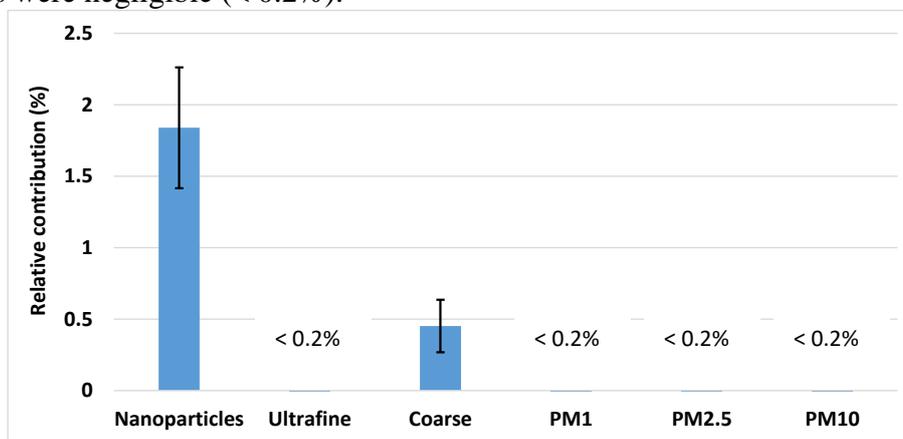


Figure 5.15: Relative contributions of ships to measured concentrations in Rijeka for the different size ranges.

5.2. Analysis of sources from chemical composition data in Rijeka

Particulate sampling was carried out at the monitoring station on the terrace of the Teaching Institute of Public Health Rijeka, Krešimirova 52a (N 45°19'54" E 14°25'32", 20 m.a.s.l) during two campaigns: autumn: from October 16th to December 10th 2018, and spring: from March 26th to May 21st 2019. Airborne particulate samples were collected with 10 stages cascade impactors plus inlet and back-up filter (stages >18 µm; 10-18 µm; 10-5.6 µm; 5.6-3.2 µm; 3.2-1.8 µm; 1.8-1.0 µm; 1.0-0,56 µm; 0.56-0.32 µm; 0.32-0.18 µm; 0.18-0.10 µm, 0.10-0.056 µm, < 0.056 µm).

The sampler allows to separate particles of different sizes. Particulate matter can be divided into:

- coarse (>1 µm)particles, represented by stages 1 to 6,
- fine particles (between 0.1 µm and 1 µm), represented by stages 7 to 10 and
- ultrafine particles (<0.1 µm), represented by stages 11 and 12.

Moreover particulate matter can be also divided in:

- PM₁₀, including particles with dimension below 10 µm (stages from 3 to 12),
- PM₁, corresponding to the sum of fine and ultrafine particles (stages from 7 to 12)
- nanoparticles, corresponding to ultrafine particles (stages 11 and 12).

Fractional distribution of collected weekly samples is given in fig. 5.16. Distribution of coarse, fine and ultrafine particles in collected weekly samples (indicates the average coarse contribution of 55% (range 30.5-80,4%), fine contribution 36,6% (range 14.4-50,1%) and ultrafine 8,4% (range 0.0-37,5%).

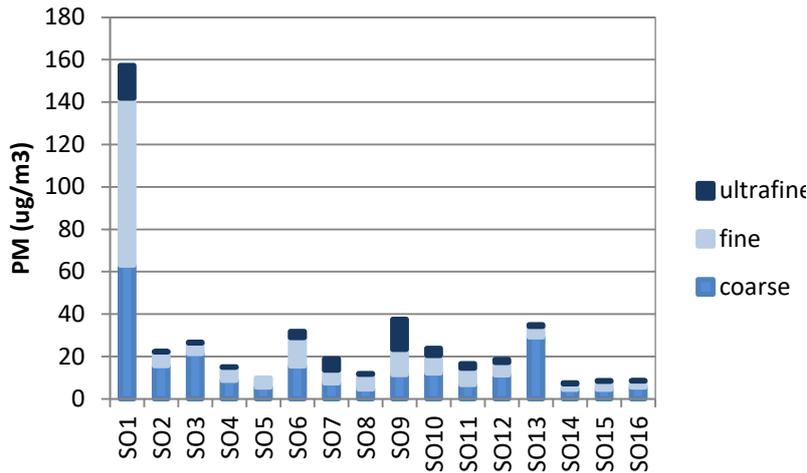


Figure 5.16. Distribution of coarse, fine and ultrafine particles in the weekly samples of particulate

The concentration of PM collected during the first week of monitoring was very high (157 $\mu\text{g}/\text{m}^3$) due to Saharan sand episode. Surprisingly, this sample contained high quantity of fine and nanoparticles, as seen from the size distribution profile (Fig. 5.17)

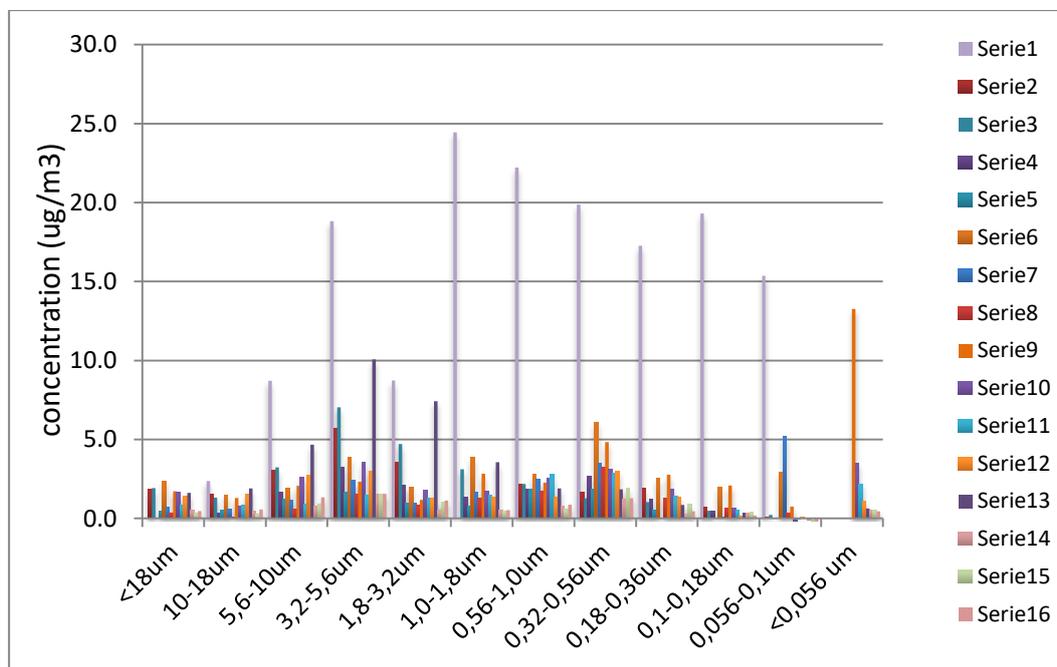


Fig 5.17.: Size distribution of collected particulate matter from both campaigns

Omitting this exceptional sample, the size distribution of airborne particulates show a bimodal curve, with two maxima: at $d=3,2-5,6 \text{ um}$ (S3) and $d=0,32-0,56 \text{ um}$ (S7). The sample with Saharan sand has somewhat different profile having additional maximum at $d=1,8-3,2 \text{ um}$ (S5). Such a trimodal curve is typical for particulate samples containing desert sand (Fig. 5.18). Regarding the fractional profile, the one in Venice is somewhat “red shifted” with maxima at $d=3,2-10 \text{ um}$ (S3-S2) and $d=0,56-1,0 \text{ um}$ (S6) with lower ultrafine particles contribution (0,3%) relative to Rijeka (8%). This might be due to different environmental conditions, of both, natural and anthropogenic sources.

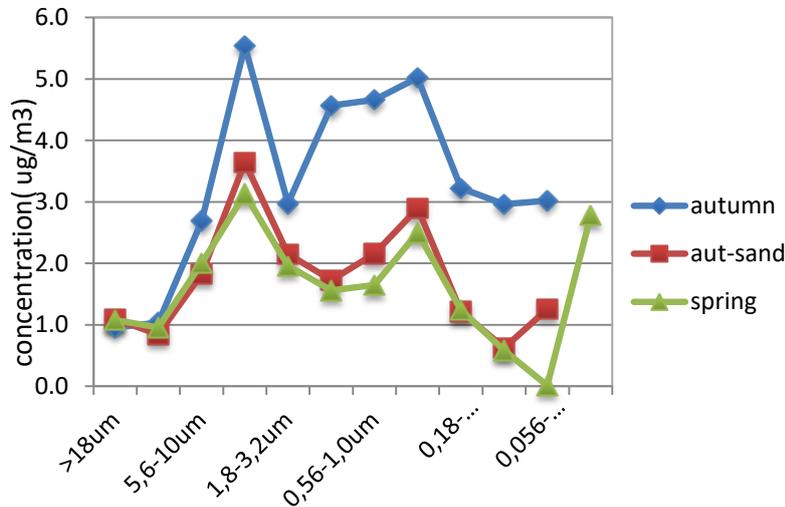


Figure 5.18: Average size distribution profile of airborne particulates in autumn and spring campaign. The profile is practically equal if sample with desert sand was omitted (autumn-sand).

Vanadium is a well known indicator of ship emission, even if other industrial sources could influence its concentration. The presence of vanadium in fine and ultrafine fractions (nanoparticles) indicate combustion as its source. The overall average of vanadium per fraction obtained during both campaign was $1,50 \pm 1,45 \text{ ng/m}^3$. The high standard deviation indicates high dissipation of the results. Thus the minimum obtained was $0,1 \text{ ng/m}^3$, while the maximum $4,8 \text{ ng/m}^3$. No vanadium is detected in 22 out of 178 fraction samples. The vanadium profile by fraction is given in Fig. 5.16. As seen from the figure, the obtained average vanadium in $\text{PM}_{2,5}$ and PM_{10} were $13,9$ and $16,7 \text{ ng/m}^3$, respectively, and are approx. three times higher than the concentrations obtained during the project POSEIDON (Deliverable 2.5, 2015).

Higher concentrations in vanadium will result in higher contribution to primary particulate emissions from the ships. One must have in mind that there are other sources of vanadium in the Rijeka area, as shipbuilding facilities within the city area that could contribute to vanadium in higher particulate fractions. This might be the reason that the highest concentration of vanadium

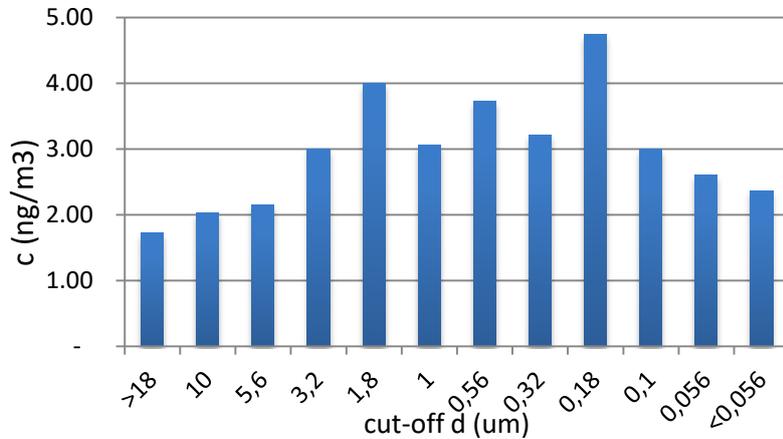


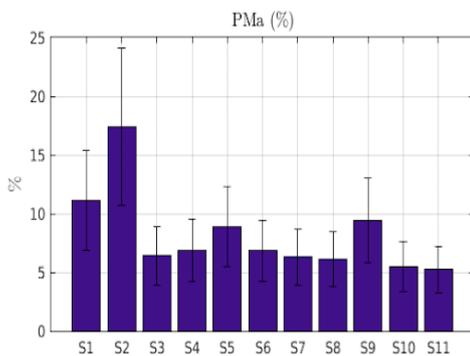
Figure 5.19: fractional profile of airborne vanadium mean concentrations

is found in the coarse fraction. On the other hand, due to the two petroleum refinery facilities within and next to the city, an elevated content of vanadium is found in soil (Prohić, 1994)

The average size distribution of the primary contribution of ship traffic to particulate matter is shown in Fig. 5.20 a (with corresponding error). The average contribution is in the range of 5% to 17%, with maxima in the coarse fractions. This profile could be the result of either mechanical ground particulates or aggregated primarily fine particulates from the combustion sources.

The primary contribution of ship traffic to coarse, fine and ultrafine particle fraction are similar, as seen from fig.5.20b

a.



b.

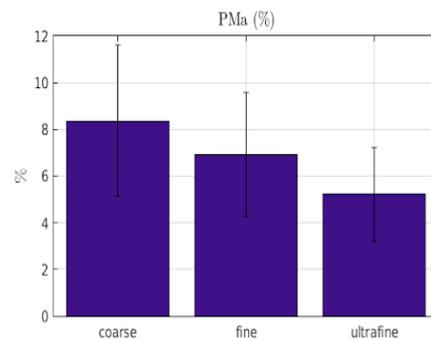


Figure 5.20: Primary contribution of maritime traffic in: a. different particle sizes (with corresponding error marks); b. different fractions)

The contribution estimated in 2019 was compared to that estimated during the period 2012-2014, within the POSEIDON project, and expressed as $\mu\text{g}/\text{m}^3$, as concentration of $\text{PM}_{2.5}$ are not available to calculate the relative contribution. The monitoring site in POSEIDON was located approx. 1200 m East from the Institute building and with elevation of 4 m asl (Deliverable 2.5, 2015). The contribution to $\text{PM}_{2.5}$ and PM_{10} calculated from 2019 data was considerably higher than the previously observed in 2013-14 (Fig. 5.21.). The estimation of contribution to PM_1 and $\text{PM}_{0.1}$ have been conducted for the first time in this area. It is interesting to note that although the contribution to primary particulate emissions to various PM fractions increased, the highest rise was observed within coarse particulates, unlike in Venice where contribution increased with diminishing the particulate dimensions.

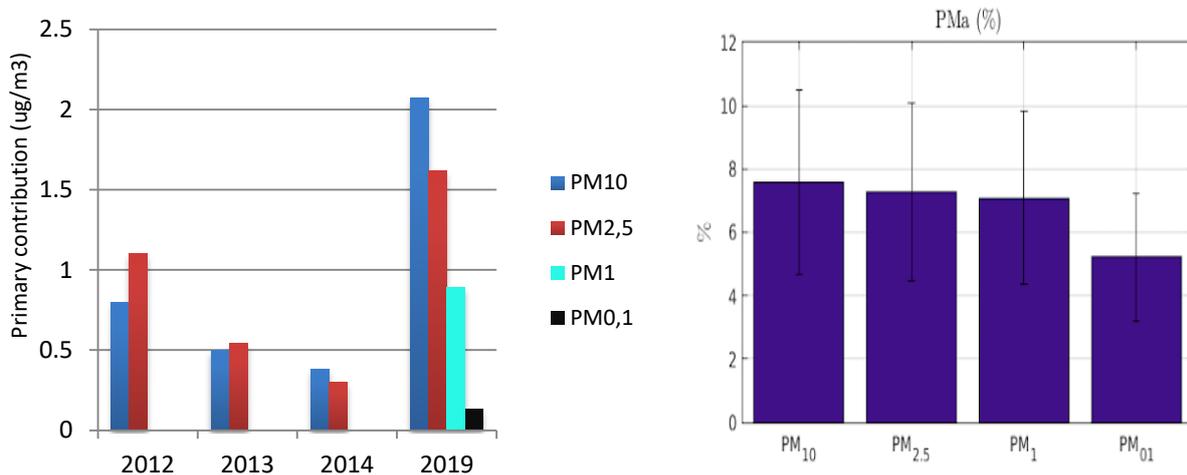


Figure 5.21: a. Comparison between the primary contribution of ship traffic to particulate matter, calculated using vanadium as tracer of shipping emissions, obtained within POSEIDON (2012-14) and ECOMOBILITY (2019) projects. b. relative primary contribution (%) to differently defined particulate fractions

The increase in primary contribution to airborne particulates is due to increased trade traffic in the port. Thus, after several years of constant decline, the port recuperated the activity as in the last pre- crisis year: 2008 (Port authority, 2019).

CONCLUSION

Regarding PM_{10} , $PM_{2.5}$ and $PM_{1.0}$, the concentration measured in Rijeka were approx. 50% of those obtained in Venice (though the monitoring time was different). The same is valid for fine particulate, while coarse are lower by approx. 30% and ultrafine by approx. 40%. Having in mind that the ship traffic is approx. 3 times lower in Rijeka relative to Venice, the current air pollution in Rijeka is obviously the result of other sources and factors.

Impact of other sources than ship traffic is seen in Fig. 5.20, where maximum contribution based on V content is found in the coarse fractions, that obviously cannot be attributed to maritime traffic. The other sources of vanadium in the Rijeka area higher V content is soil dust, (Prohić, 1994) due to long term combustion of HFO in petroleum refinery, as well as ship building and ship repair in two still active shipyards.

The calculated primary contribution of ship traffic to fine and ultrafine fractions in Rijeka are by 40 % lower than in Venice, but having in mind that the ship traffic is just one third of that in Venice, the “excess pollution” of the Rijeka bay must be due to other sources and complex orography that keeps the pollutants within the bay. The increase in primary contribution to airborne particulates is due to increased trade traffic in the port. Thus, after several years of constant decline, the port recuperated the activity as in the last pre- crisis year: 2008 (Port authority, 2019).

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