Interreg Italy - Croatia ECOMOBILITY

EUROPEAN UNION

ECOlogical supporting for traffic Management in cOastal areas By using an InteLlIgenT sYstem

AXIS 4: Maritime Transport
Objective 4.1: Improve the quality, safety and environmental sustainability of marine and coastal transport services and nodes by promoting multimodality in the Programme area

Analysis report in Rijeka

University of Rijeka-Faculty of Medicine -Teaching Institute of Public Health

Institute for Atmospheric Science and Climate of the National Research Council (ISAC-CNR)

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WP3: Assessment of the effect of ship traffic to particulate matter
ACT 3.2: Experimental activities in Rijeka

CONTRIBUTED TO THIS WORK

Ana Alebić-Juretić, University of Rijeka- Faculty of Medicine, Environmental Health Department

Boris Mifka, Departmant of Physics, University of Rijeka

Velimir Zubak, Environmental Health Department, Teaching Institute of Public Health

Dario Kontošić, Environmental Health Department, Teaching Institute of Public Health

Paula Žurga, Environmental Health Department, Teaching Institute of Public Health

Marjana Mezlar, Environmental Health Department, Teaching Institute of Public Health

Dajana Odorčić, Environmental Health Department, Teaching Institute of Public Health

Eva Merico, Institute for Atmospheric Science and Climate of the National Research Council (ISAC-CNR)
Fabio M. Grasso, Institute for Atmospheric Science and Climate of the National Research Council (ISAC-CNR)
Daniela Cesari, Institute for Atmospheric Science and Climate of the National Research Council (ISAC-CNR)
Marianna Conte, Institute for Atmospheric Science and Climate of the National Research Council (ISAC-CNR)
Daniele Contini, Institute for Atmospheric Science and Climate of the National Research Council (ISAC-CNR)

INTRODUCTION

This report deals with experimental activities carried out in Rijeka within activity 3.2 of the ECOMOBILITY project. The activity includes sampling of particulate matter by fractions of different sizes, chemical analyses, high-resolution measurements and measurement of ancillary data. This report does not give the results about the sources of pollution, since this is the subject of the another deliverable "Assessment of the impact of shipping to particulate matter in Venice and Rijeka" These two deliverables are strictly connected, since the results of one are the input to the other.

This report is composed of two parts:

- the first part is dedicated to the sampling of particulate matter in different size and chemical characterization;
- the second part is dedicated to the high-resolution measurements of particulate matter and related parameters.

Every chapter contains the procedures carried out and the main results reported.

SUMMARY

PART I
Sampling and chemical analysis

1. SAMPLING

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,
- spring: from March 26th to May 21st 2019,

During each campaign 8 week samples were collected using 110 MOUDI cascade impactor with quartz fibre filters (d=47mm, Whatman) as a collecting substrates.
The sampler allows to separate particles of different sizes (Table 1.1). 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) and
- nanoparticles, corresponding to ultrafine particles (stages 11 and 12).

Table 1.1. Dimension of collected particles

	Stage Size
Stage 1 -inlet	>18 µm
Stage 2	18-10 µm
Stage 3	10-5.6 µm
Stage 4	5.6-3.2 µm
Stage 5	3.2-1.8 µm
Stage 6	1.8-1.0 µm
Stage 7	1-0.56 µm
Stage 8	0.56-0.32 µm
Stage 9	0.32-0.18 µm
Stage 10	0.18-0.10 µm
Stage 11	0.10-0.056 µm
Stage 12 back-up	<0.056 µm

A total of 16 weekly samples were collected, eight in each campaign with corresponding dates and volume given in table 1.2. The volume was controlled by an orifice flow-meter (Oriflow, Sven Leckel) . Weekly samples were collected on quartz fiber filters (47 mm diameter). Dates and sampled air volumes are reported in Table 1.2. For every set of sample filters, a blank filter was taken.

Table 1.2: Sampling periods with corresponding volumes

Sample No	Period	V (m3)
	Autumn 2018	
Sample 1	16.10.-23.10	220,67
Sample 2	23.10.-29.10.	235,13
Sample 3	29.10.-05.11.	257,96
Sample 4	05.11.-12.11.	274,59
Sample 5	12.11.-19.11.	273,90
Sample 6	19.11.-26.11.	275,55
Sample 7	26.11.-03.12.	236,50
Sample 8	03.12.-10.12.	277,75
	Spring 2019	
Sample 9	26.03.-02.04.	255,74
Sample 10	02.04.-09.04.	259,07
Sample 11	09.04.-16.04.	254,59
Sample 12	16.04.-23.04.	258,18
Sample 13	23.04.-30.04.	257,36
Sample 14	30.04.-07.05.	259,84
Sample 15	07.05.-13.05.	184,90
Sample 16	13.05.-21.05	246,21

Samples and blanks collected in autumn 2018 and spring 2019 were used to evaluate the size distribution of:

- particulate matter concentration;
- concentration of ions associated to particulate matter;
- concentration of metals associated to particulate matter;
- concentration of carbon species associated to particulate matter.

For this purpose, after weighting the collected mass on the filters, filters were first punched with a punch tool to extract a 1 cm^2 cut and the filter, and subsequently cut in two halves.

One half of the filter was used for metal analyses; the 1 cm^2 punch was analyzed for elemental and organic carbon (EC and OC), while the punched half of the filter was used for subsequent analyses of water soluble organic compounds (WCOC) and ions from the same water extract.

2. PARTICULATE MATTER CONCENTRATION

2.1. Material and methods

Blanks and sampled filters collected were weighed according to the standard HRN EN 12341:2014 in the appropriate weighing room with controlled humidity (50 ± 5% RH) and temperature (20±5 °C).

The weights of the filters before the sampling were subtracted from the weights of the corresponding filters after the sampling, to obtain the quantity of the deposited material. The quantity of the blanks were subtracted to the weights for each sample in the week set, in order to correct a potential contribution due to contamination during transport, conservation and manipulation of samples.

Concentration values below the detection limit, calculated as the blank average plus three times the standard deviation of the blanks, were rejected.

2.2. Results and discussion

The concentration of particulate matter in different size is reported in the Annex (Table A1). The term "ND" indicates not detected values (values below the detection limit).

Weekly values of total particulate matter, calculated as the sum of all stages, ranged from 9,7 µg/m3 to 157 µg/m3 during the autumn campaign, and from 8,6 to 37,4 µg/m3, during the spring campaign.

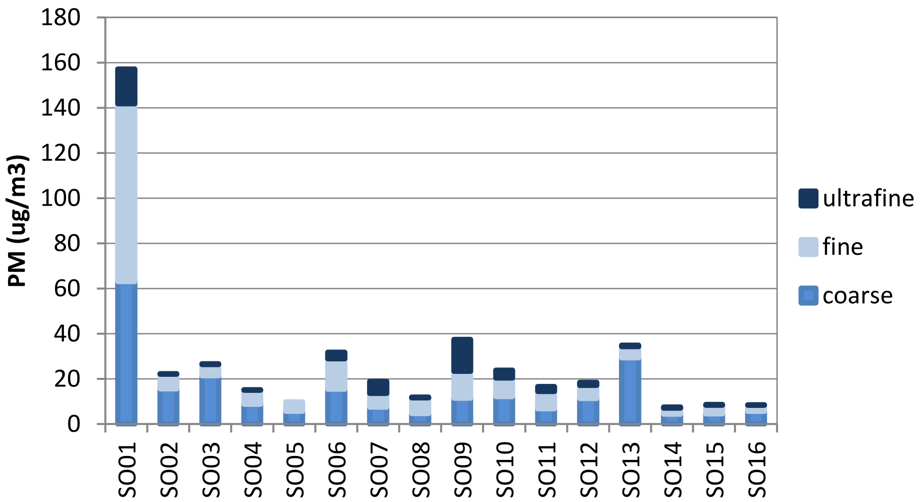

Figure 2.1. Distribution of coarse, fine and ultrafine particles in the weekly samples of particulate matter

Unexpectedly high average concentration of particulates was obtained in the first week of the autumn campaign. Meteorological situation and chemical analyses of the weekly sample confirmed that it was due to the rather strong Saharan sand episode, as seen from the satellite picture (Fig 2.2.). The second, minor desert sand episode occurred in April 2019.

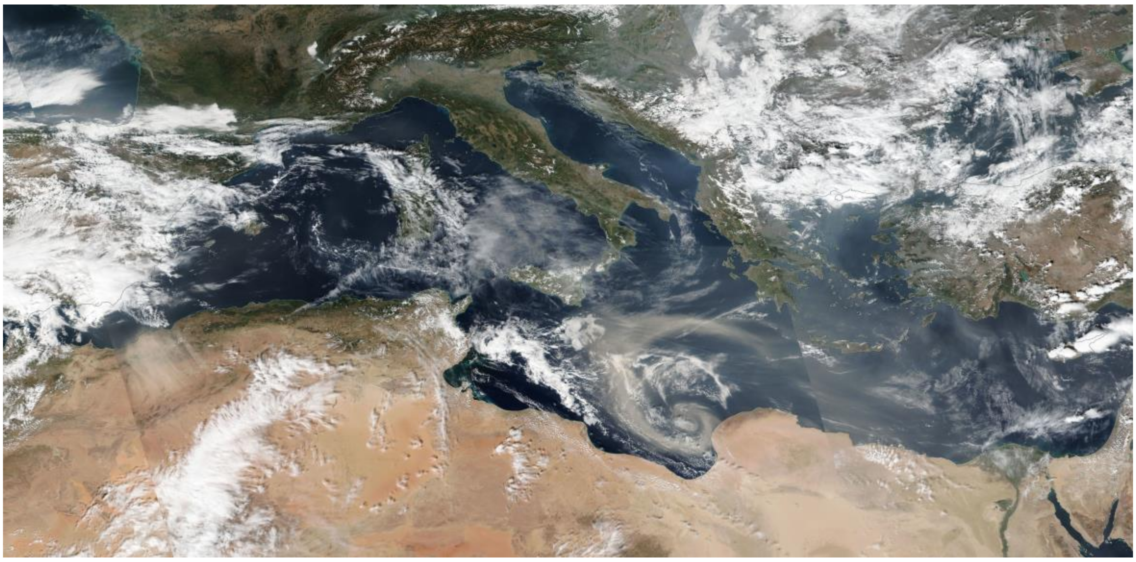

Figure 2.2.: Terra/Aqua satellite, VIS data: corrected reflectance (https://worldview.earthdata.nasa.gov)

This unusually high concentration of particulates is sometimes excluded from trend analyses. Distribution of coarse, fine and ultrafine particles in collected weekly samples (Fig.2.3) 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%). Different percentage of particular fraction indicates different sources of airborne particulates. Thus, the contribution of ultrafine particles show an increase with the

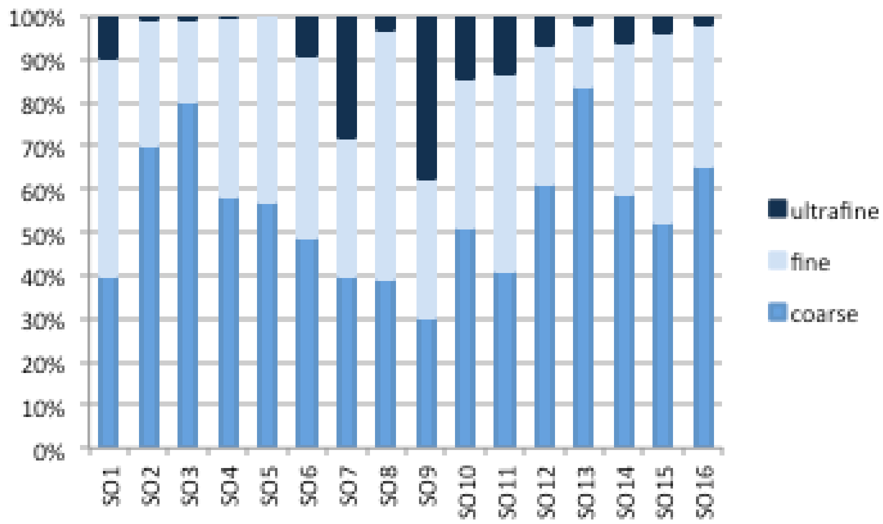

Figure 2.3.: Percentage distribution of coarse, fine and ultrafine particles in collected weekly samples

beginning of heating season (mid-November) indicating domestic heating to be important, if not major sources of. In addition, autumn meteorological conditions with lower PBL relative to warmer seasons might also contribute to the rise of ultrafine particle concentrations.

Surprisingly, the sample with Saharan sand contained also high content of fine (50,1%) and ultrafine particles (9,8 %), indicating aging process of lifted sand in the atmosphere to be responsible for. This is contrary to recent studies undertaken on desert dust episodes in Turkey[1] where no fine particulates were found in such a sample.

The size distribution of particulate matter (Fig. 2.4) shows a random pattern at the first sight due to multiple higher concentrations observed in the particulate samples containing desert

[1] Uzunpinar E.S. et al: Proceeding of Abstracts, 18th World Clean Air Congress 2019, Sept 23rd- Sept 27th 2019, Istanbul, Turkey, pp.111

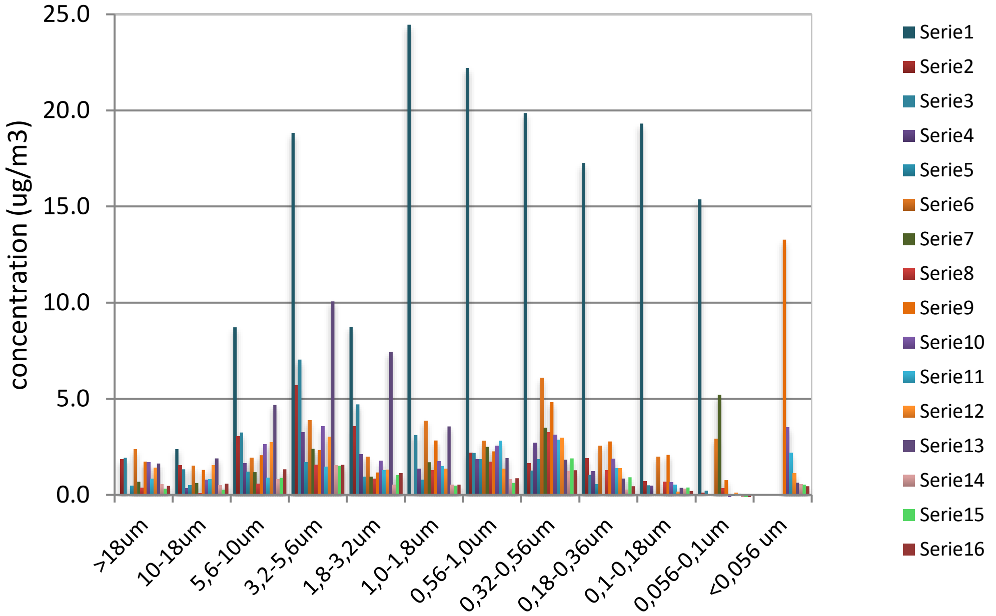

Figure 2.4.: Size distribution of collected particulate matter from both campaigns

sand. Omitting this exceptional sample, the size distribution of airborne particulates show a bimodal curve, with two maxima: at d=3,2-5,6 um (S3) and d=0,32-0,56 um (S7). The sample with Saharan sand has somewhat different profile having additional maximum at d= 1,8-3,2 um (S5). Such a trimodal curve is typical for particulate samples containing desert sand (Fig. 2.5). Regarding the fractional profile, the one in Venice is somewhat "red shifted" with maxima at d=3,2-10 um (S3-S2) and d=0,56-1,0 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.

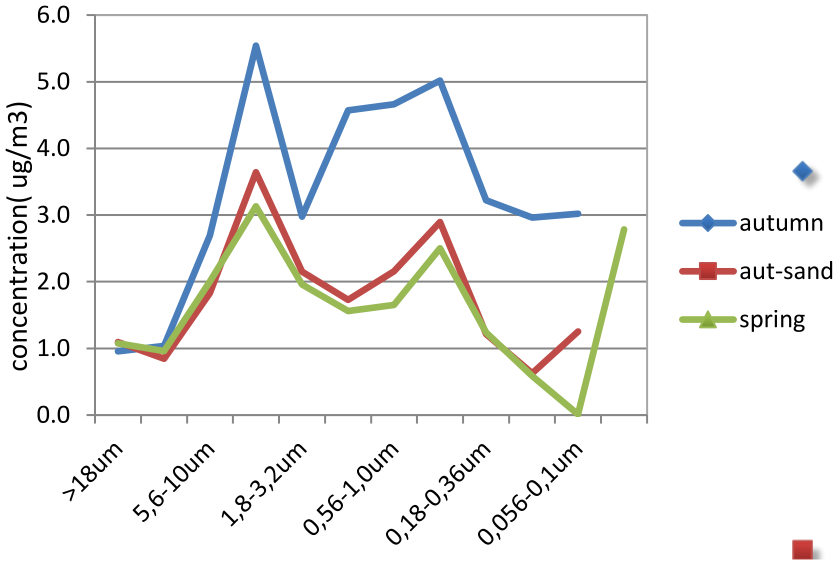

Figure 2.5: Average size distribution profile of airborne particulates in autumn and spring campaign. The profile is practically equal if sample with desert sand is omitted (autumn-sand).

3. ANALYSIS OF IONS

The ionic contribution of collected particulates were determined by analysing basic species:

- Anions: chloride (Cl^-), nitrate (NO_3^-), sulphate (SO_4^{2-}) and phosphate (PO_4^{3-}). The phosphate was analysed due to the fact that phosphate fertilizer is reloading in the port area just opposite to the Institute building.
- Cations: sodium (Na^+), ammonium (NH_4^+), potassium (K^+), calcium (Ca^{2+}) and magnesium (Mg^{2+})

3.1. Material and methods

For ion analyses the punched half of the filter was extracted in 30 ml of demineralized water in ultrasonic bath (marka) for 15 min. The water extract was filtered through the filter with pore size 0,2 µm (Whatnman).

The analyses of ions was carried on 940 Professional IC Vario (Methton) supplied with conductivity detector. The Metrosep A Sup 5-250 column with corresponding A Sup 5 guard column was used for determination of the anions (Cl^-, SO_4^{2-}, $NO_3^{-,}$, PO_4^{3-}). Bicarbonate/carbonate (HCO_3^-/CO_3) buffer solution was used as mobile phase. Cation determination (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}) was carried on Metrosep C4 150 column with C4-guard column and tartaric/dipicolinic acid as mobile phase.

Both methods for determination of anions (HRN ISO 10304-1:2009) and cations (HRN EN ISO 14911:2001) by ion chromatography are accredited according to EN ISO 17025:2006.

Only concentrations above the quantification levels (instrumental blanc + 10 standard deviations of the blanks) were took in consideration. The values between lowest detection level (instrumental blanks + 3 standard deviation of this blanc) were considered detected, but unreliable.

3.2. Results and discussion

Concentrations of all ion species analysed are given in the Annex (Table A2). Method detection limits (MDL) for each ionic species with corresponding quantification limits (MQL) are given in Table A3 of the Annex.

Although some ions do not appear in all stages, due to their source and atmospheric chemistry fate and its importance, all are included in the discussion.

Distribution of ions in particulate matter

Concentrations of ions are summarized as median, 1st quartile and 3rd quartile in Table 3.1. Concentration of ionic species are referred to the total suspended particulate, obtained by summing-up by all collected size segregated fractions.

Table 3.1: Distribution of ions in total suspended particulates (ng/m3)

ion	Median	1st quartile	3rd quartile
PO43-	9,2	2	18,1
Cl-	22,2	4,4	43,8
NO3-	20,1	6,7	39,8
SO42-	41,9	16,5	106,2
Na+	39,4	15,3	80,8
NH4+	0	0	9,8
K+	2,3	0	10,9
Ca2+	80,2	28,4	182,2
Mg2+	3,3	0	13,9

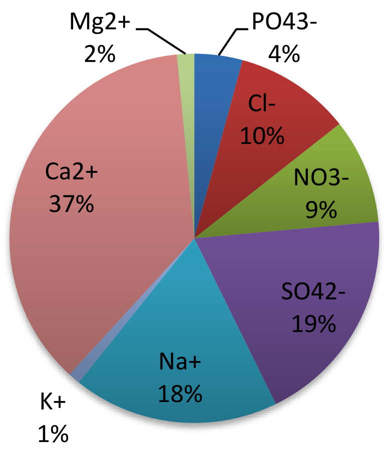

Figure 3.2. Percentage distribution of median concentrations of major ions

As visible from the Table 3.1 and Figure 3.2., some species have zero (0) as median (NH_4^+) or the first quartile (NH_4^+, K^+ and Mg^{2+}). This is not the result of impossibility for detection, but chemical fate of this species in the atmosphere. Thus, NH_4^+ as secondary inorganic aerosol (SIA) appears only in some of the finer fractions (less than 50%), and consequently the median and 1st quartile are zeros. Similar behavior is found for K and Mg^{2+} in the lower quartile.

The must abundant ionic species in total particulates are calcium (37%), sulphates (19%) and sodium (18%). High content of Ca and Na indicates natural sources, (carst) soil and sea salt. Although we cannot determine carbonates with our IC system, having such a high content of calcium in mind, this ion should be recalculated from other chemical species.

Comparing these results with those obtained in Venice, it is clear that our ion concentration levels are by an order of magnitude lower then in Venice. The percentage contribution of sulphates are lower by a third, nitrate by two thirds and chloride by approx. 50% in Rijeka comparative to the respective values in Venice[2]. This is not surprising since Venice population is double compared to Rijeka, with enormous number of tourist visits, active industry area and maritime traffic 3 times higher than Rijeka[3].

Size distribution of ions

The distribution parameters of ions (median, 1st and 4th quartile) among various dimensional fraction are given in Table A4 of the Annex.

The aerodynamic diameter of the collected particulates (Figure 3.3.) can suggest their source, as well as the fate and the lifetime in the atmosphere.

The most abundant ionic species was calcium (37%), originating mostly from the carst soil and thus distributed in the coarse fraction, with maximum at 5,6-10 µm. Saharan dust episode observed at the beginning of the autumn campaign also contributed to the high content of calcium, but omitting that values would not change much the content. Ca^{2+} is also found in the fine and ultrafine fractions. Except wood combustion, one of the possible source is reload of calcium phosphate fertilizer on the pier opposite to the Institute building. Phosphate fractional profile also has significant contribution of fine and ultrafine particles, though its percentage content to TSP is much lower (only 4%).

The second most abundant ions are SO_4^{2-} (19%) and Na^+ (18%). Sodium is tracer of the sea- salt aerosols and is usually found in the coarse fraction. Na^+ is also present in the fine and ultrafine

[2] Ecomobility: Analysis report in Venice, University Ca' Foscari, Venice, 2019
[3] E. Merico et al. , Transportation research, Part D, 50, 2017, 431-445

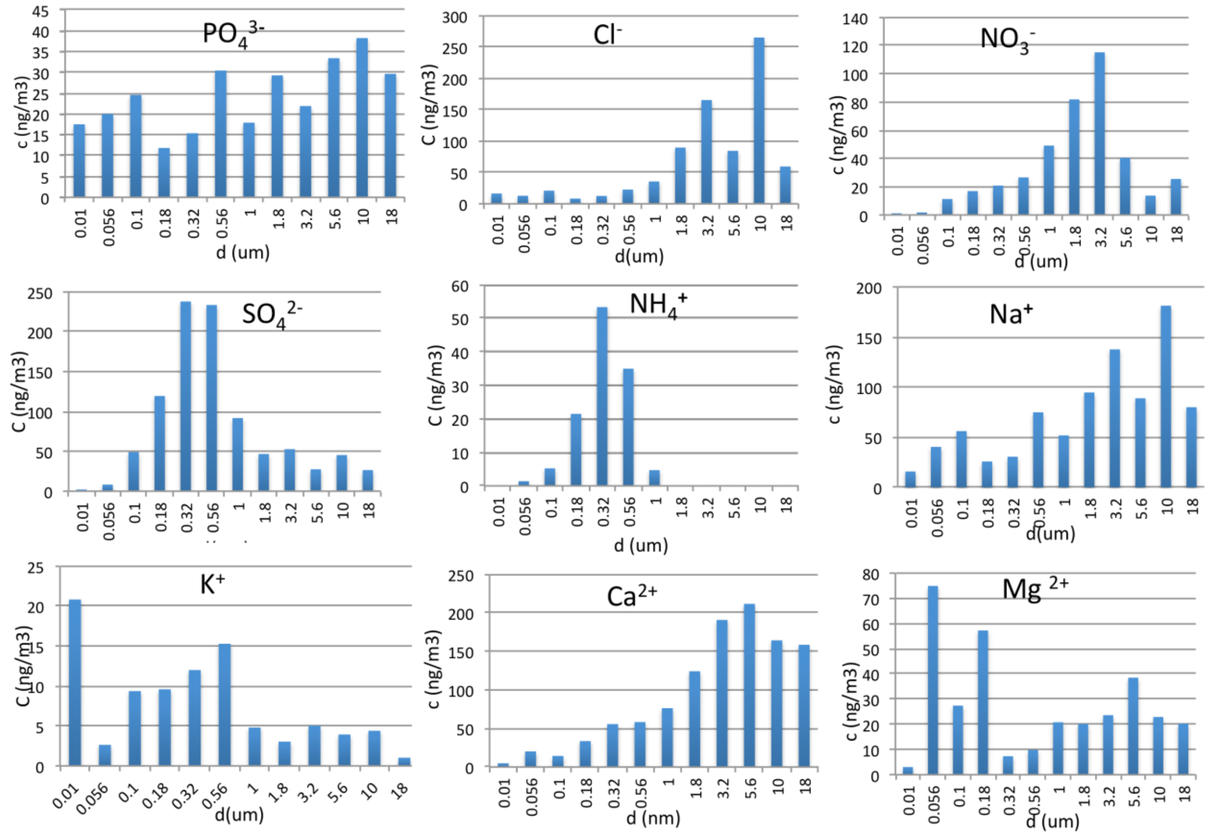

Figure 3.3.: Average size distribution of ions according to cut-off sizes d (µm)

fractions. One of the possible sources might be the reload of soda ($NaCO_3$) on the pier close to the Institute building. Similar profile is found with chlorine, mostly of marine origin and distributed in the coarse fraction within maximum at d=5,6-10 µm, but some minor quantities in fine and ultrafine fractions.

Magnesium, another marker of sea-salt, has also significant contribution in fine and ultrafine fractions, possibly indicating biogenic source, similarly to potassium whose presence in fine and ultrafine fractions are attributed to biomass burning.

Second most abundant ion SO_4^{2-} (19%), is found mostly in fine and less in ultrafine fractions, reflecting its atmospheric chemistry origin as secondary inorganic aerosol (SIA) or non sea-salt sulphate (nss-SO_4^{2-}). Minor quantity is found in coarse fraction, probably of marine origin (sea-salt sulphate ss- SO_4^{2-}). Other representative of SIA, nitrate is mostly found in coarse fraction, as the result of intrusion of nitric acid in sea-salt (releasing HCl), but minor quantity is found also in the fine fractions, as pure SIA. The only ion having a clear cut between different fraction is ammonium. As a result of atmospheric chemistry, it is exclusively distributed in fine fractions, as SIA.

It is interesting to note that samples with Saharan sand has the highest concentration of secondary pollutants SIA and WSOC, consequently found in the fine and ultrafine fraction (Fig. 3.4). This finding is suggesting the adsorption/absorption of gaseous pollutants to dust surface and their transformation in secondary pollutants on the way of transport from North Africa to Northern Adriatic area.

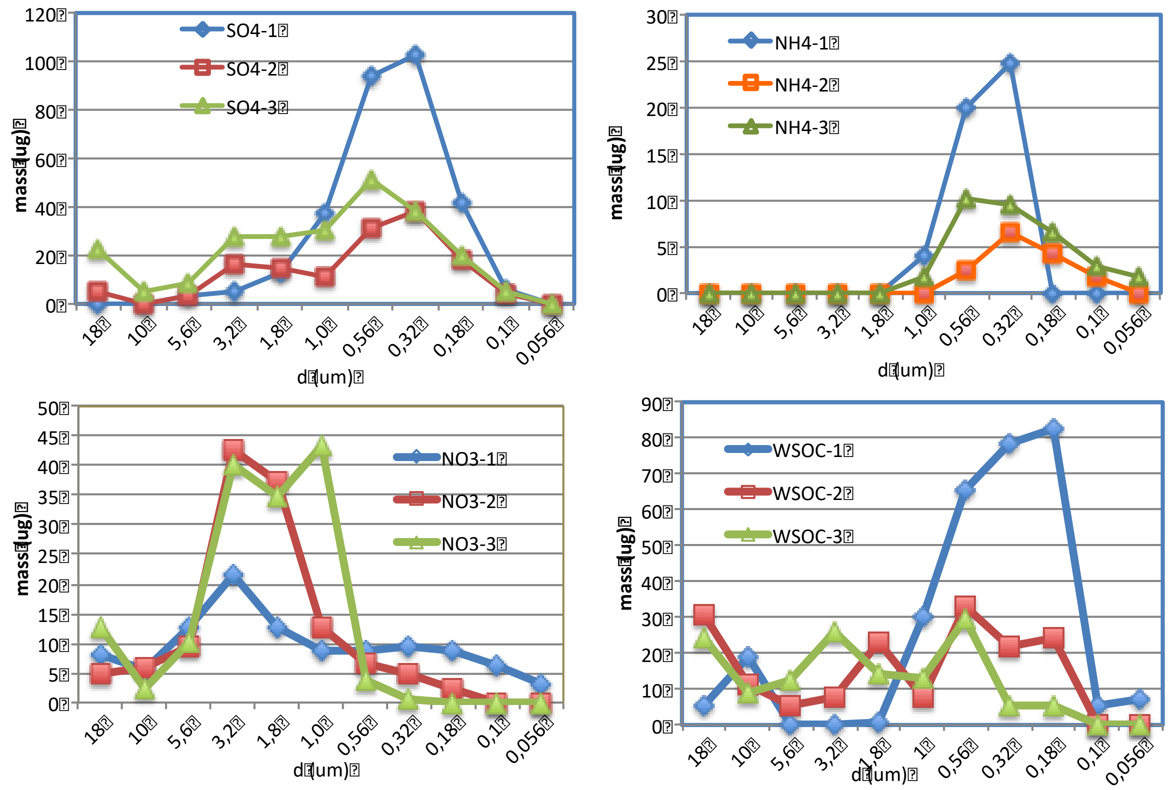

Figure 3.4.: Masses of secondary pollutants in sample (No. 1) containing Saharan sand relative to subsequent samples (No. 2 1nd 3). Only NO3- has lower values relative to other two samples, proofing less impact of sea salt.

4. ANALYSIS OF METALS

Samples of particulates were analysed on content of followimg metals: chromium (Cr), manganese (Mn), iron (Fe), cadmium (Cd), nickel (Ni), copper (Cu), zinc (Zn), lead (Pb) aluminium (Al), antimony (Sb) and vanadium(V).

4.1. Material and methods

Analyses of metals in particulate matter was carried out by mineralization of the half of quartz filter and subsequent instrumental analysis with ICP-MS (Inductively Coupled Plasma – Mass Spectrometry, NexION 300X, PerkinElmer). The mineralization of quartz filter was carried out in a microwave oven (Ethos Easy, MIlestone) and a mixture of the ultrapure reagents: nitric acid, hydrochloric acid and hydrogen peroxide. The temperature program for microwave mineralization consists of a heating up to 200^{o}C, and maintenance of this temperature for 20 min . The solution obtained in the microwave was diluted to 15 ml with fully deionized ultrapure water (18 MΩ/cm resistivity) produced by Ultra Clear System (Siemens). The ICP-MS was calibrated using the multielemental standard solution (Initial calibration verification standard, Perkin Elmer), with concentration varying between 3-200 µ, depending on their abundancy in the real samples. The calibration lines had R= 0.9999 for Cr, Cd, NI, Cu, Pb and V, R= 0,9998 for Mn and Sb and R= 0,9986 for Fe. A standard reference material (Urban particulate matter, CRM 1648a, NIST) was used at known concentration to evaluate recoveries that ranged between 93 % and 115%, except for Cr, Cd, Ni and V with higher values of 130%. The concentration of the elements were found as those values subtracted from the respective in blanks. Concentration values were considered only if they were above the detection limit, calculated as average instrumental blanc plus three times standard deviation of the blanks.

4.2. Results and discussion

The concentration of each species in each sample are given in the Annex (Table A5). The term ND indicates not detected values (below the average blanc). Detection limits for metals are given in Table A6.

Table 4.1.: Distribution of metals (median, 1st and 3rd quartiles) in total suspended particulates (ng/m^3), number of weekly samples with concentration values bellow the blanks

Metal	Median	1st quartile	3rd quartile	n<blanc
Cr	7,56	6,85	9,06	0/16
Mn	14,11	7,83	15,72	0/16
Fe	1174,96	712,81	1612,49	0/16
Cd	0,92	0,51	1,17	0/16
Ni	11,72	7,58	15,1	0/16
Cu	21,06	16,83	30,91	0/16
Zn	22,86	13,79	30,46	0/16
Pb	5,15	3,16	7,95	0/16
Al	328,5	216,84	375,23	0/16
Sb	0,92	0,80	1,55	0/16
V	16,35	8,26	26,05	0/16

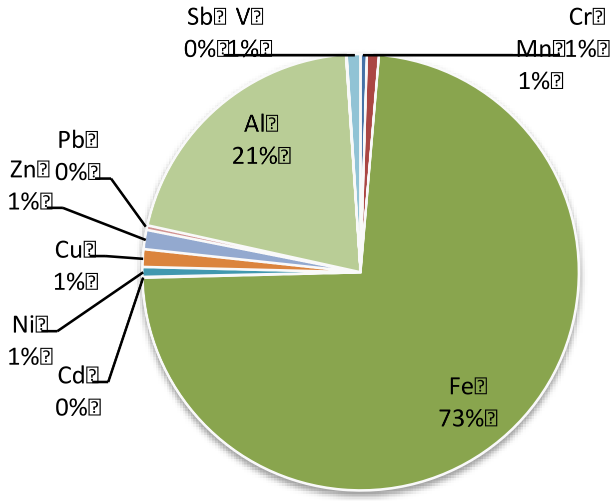

Figure 4.1. : Median distribution of metals in total suspended particulates

From the above table 4.1. and Fig. 4.1 it is evident that the most abundant metals are iron (73%) and aluminum (21%), mostly of natural origin, those of pure anthropogenic origins are ≤1%

Size distribution of metals

The distribution parameters (median, 1st and 3rd quartile) of metals among the various dimensional classes are reported in Table A7 of the Annex.

Size distribution of each metal depends on their sources and chemical fate in the atmosphere resulting in odd distribution in some fractions.

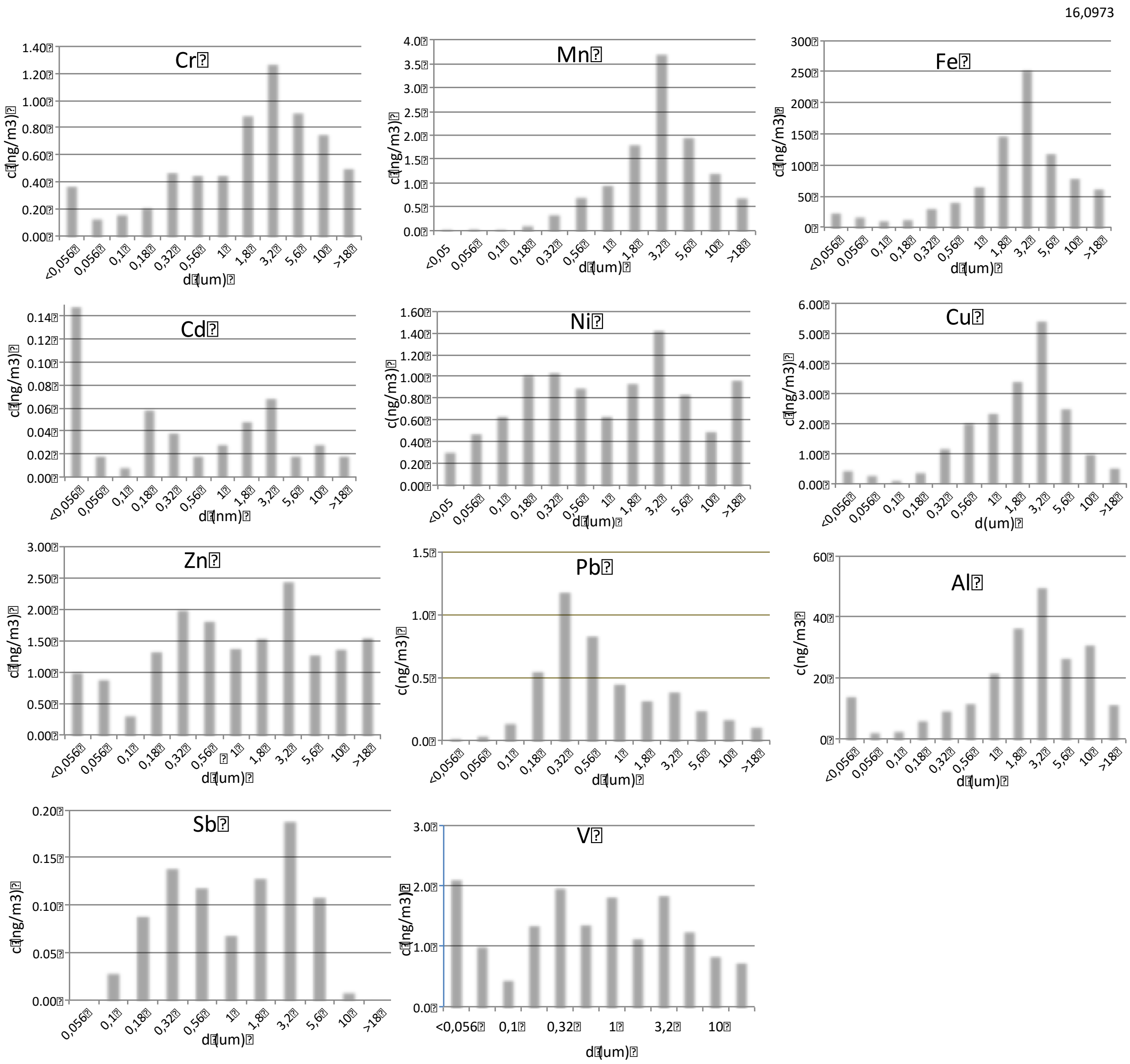

Figure 4.2 : Median size distribution vs, cut-off diameter (µm) of metals (ng/m3)

Thus metals with predominant natural sources or mechanical grinding have maxima in coarse fractions at 3,2-5,6 µm (Fe, Al, Cu, Zn and Cr), though having some contribution in fine and ultrafine fractions indicating anthropogenic origin, possibly from ship-building/repair industry (galvanization). Similar behaviour is also found with Mn, Ni and V.

Cadmium is often found with zinc in nature with maximum in coarse fraction (1,8-3,2 µm). Appearance in fine (0,18-0,32 µm) and ultrafine fractions (< 0,1 µm) might result from human activity: galvanization and oil combustion, the letter possible cause of irregular appearance in particulate samples.

Surprisingly, although leaded gasoline was abandoned in Croatia in 2006, lead had its maximum in fine fraction (cutoff d= 0,32 µm) suggesting anthropogenic sources.

Antimony, nickel and vanadium show a bimodal curve, with common maximum in fine fractions at cut-off diameter d= 0,32µm. All three metals are emitted during burning of oil fuels. Maxima in coarse fractions are observed in different size fractions.

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5. ANALYSES OF CARBON

The following carboceneous species have been analysed. Elemental carbon (EC), organic carbon (OC) and total carbon (TC) as their sum. In addition, water soluble organic carbon (WSOC) has been determined instead of the initially planned polynuclear aromatic hydrocarbons (PAHs), that could not be determined above detection limits.

5.1. Material and methods

EC, OC, and TC

Determination of OC and EC were obtained applying the thermo-optical transmittance method (TOT) for charring carbon correction using a Sunset laboratory carbon analyzer (Sunset Laboratory Inc,OR, USA) with temperature offset correction. Punches of 1 cm2 were cut from the quartz fiber filter and analysed according to the EUSAAR2 protocol, designed as the European standard procedure in the European Supersites for Atmospheric Aerosol Research.

Multipoint calibration allowed the accuracy of EC/OC analysis, by using a sucrose solution as external standard (2,198 gC/L in water, CPAchem Ltd). Analyses of blanc filters were carried out to correct concentrations of measured ambient samples (from possible contamination of EC and OC).

WSOC

WSOC was determined from the water extract obtained by sonication of the punched half of the filter in 30 ml demineralized water. The extract was filtered through a teflon filter (0,45 µm pore size), and WSOC was determined using a TOC analyzer (Shimatzu TOC-V_{CPH} with NDIR detector).

5.2. Result and discussion

The concentrations of each species in each sample is reported in Annex (Table A8). Total carbon (TC) is calculated as the sum of EC and OC. ΔEC, ΔOC and ΔTC represent the error associated to EC, OC and TC, respectively. The term "NQ" indicates values that cannot be determined.

Distribution of carbon in particulate matter

The concentration of carbonaceous matter (EC,OC,TC, WSOC) is summarized reporting the median, first and the third quartiles of the distribution. Concentration in this stage is calculated to an integrated sample, i.e. summing up all of the twelve size classes.

Table 5.1. Distribution parameters (median, 1st and 3rd quartiles) of the concentration of carbonaceous fractions in total suspended particulates (µg/m³)

	Median	1st quartile	3rd quartile
OC	0,15	0,03	0,19
EC	0,08	0,02	0,11
TC	0,28	0,04	0,33
WSOC	0,05	0,00	0,10

Size distribution of carbon species – OC, EC, TC

The distribution parameters (median, 1st quartile, 3rd quartile) of carbon species among the various dimensional classes are reported in Table A9 of the Annex.

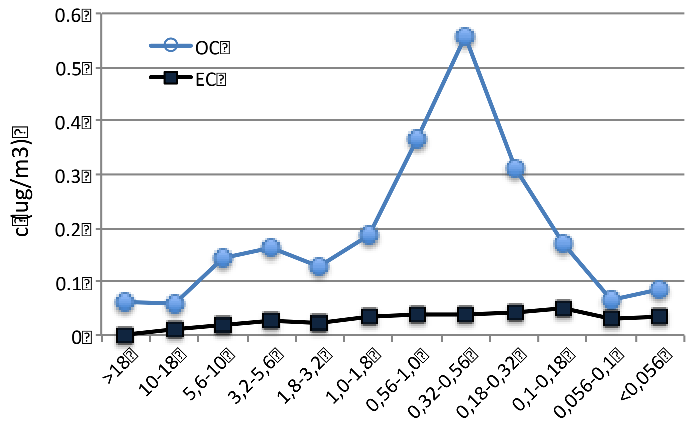


Figure 5.1.: Median size distribution of organic (OC) and (EC) carbon

The average concentrations of all integral weekly samples OC and EC are 2,47 µg/m³ (range 1,4-3,53 µg/m³) and 0,37 µg/m³ (range 0,24-0,56 µg/m³), respectively. The highest concentrations of OC and EC were observed in the sample No1 containing the Saharan sand (Fig. 5.2), proving the assumption that these air pollutants were soaked by the sand on the transport path. The higher concentrations of EC and OC were also observed in samples 4 and 7, due to beginning of domestic heating and samples 9 and 10, that could be the results of increased ship traffic in the harbor. The OC accounted in average to 86% of TC (range 84,3 -90,2 %). The correlation OCvs.EC is quite good with r=0,600, indicating some common sources.

The average ratio OC/EC is 6,68 (range 4,73-9,23 μg/m^3). The highest ratios OC/EC >9 is obtained at the end of the autumn campaign, indicating significant impact of domestic heating. All these values are lower relative to the results obtained in Venice, as the Rijeka area is less inhabited with lower industrial activity and ship traffic relative to Venice

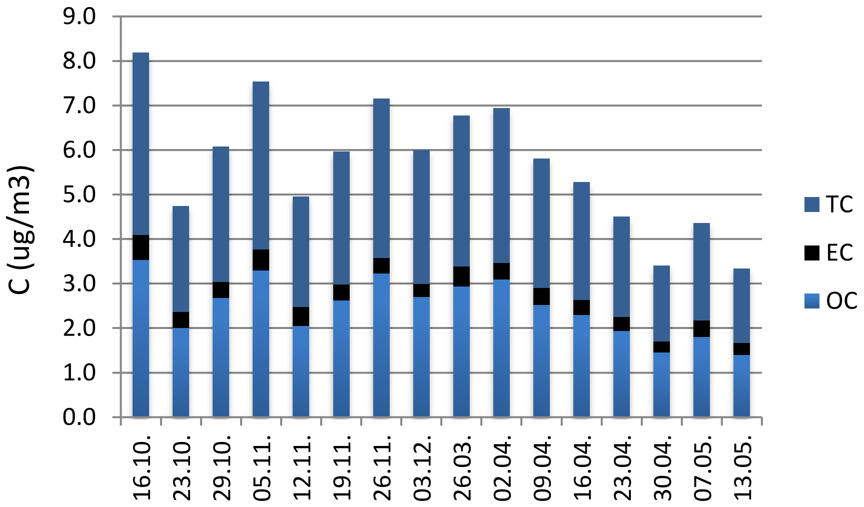


Figure 5.2.: Sample concentrations of OC, EC and TC

The size distribution of OC (Figure 5.1.) shows a bimodal curve, with first maximum at fine fraction (d=0,32-0,56 μm), and second at the coarse (d=3,2-5,6 μm).

The size distribution of EC does not show a clear profile. There is a steady increase towards smaller fractions, with maximum at st.10 (d=0,1-0,18 μm). This is the result of different nature of EC source, being primarily combustion processes, compared to OC, that could be both, primary and secondary pollutant.

WSOC

As a replacement for analysis of polynuclear aromatic hydrocarbons (PAH) that could not be performed due to small amount of particulates collected at each stage, we carried out the analyses of water soluble organic compounds (WSOC) as a proxy to secondary organic pollutants (SOA). WSOC represent an oxygenated organic compounds bound to particles and soluble in water. They could be either of natural origin (pollen and vegetation crumbs) collected at coarse fraction stages or the results of chemical transformation of gaseous pollutants in the atmosphere that fill the fine and ultrafine stages. The presence of WSOC in fine and ultrafine fractions indicates the formation of SOA, i,e. aging od airborne particulates during their persistence in the atmosphere.

The fractional profile of WSOC is given in Fig. 5.3, in combination with other carbonaceous compounds EC and OC, as WSOC represents the polar part of OC.

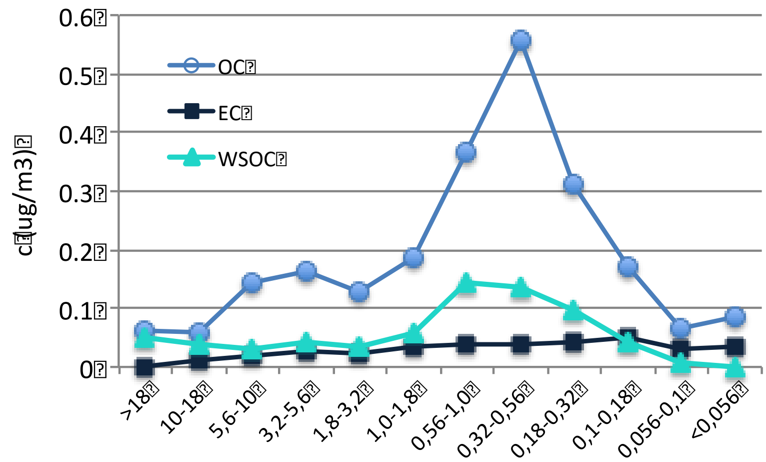


Figure 5.3.: Fractional median distribution of WSOC together with EC and OC

From the above figure is visible that WSOC are mostly found in the fine fractions (< 1 µm), with maximal contribution in St. 7 (d=0,56-1,0 µm), indicating atmospheric reaction as their source. Correlation of OC-WSOC is good (r=0,8770, p<95%) indicating their same source. Subtracting

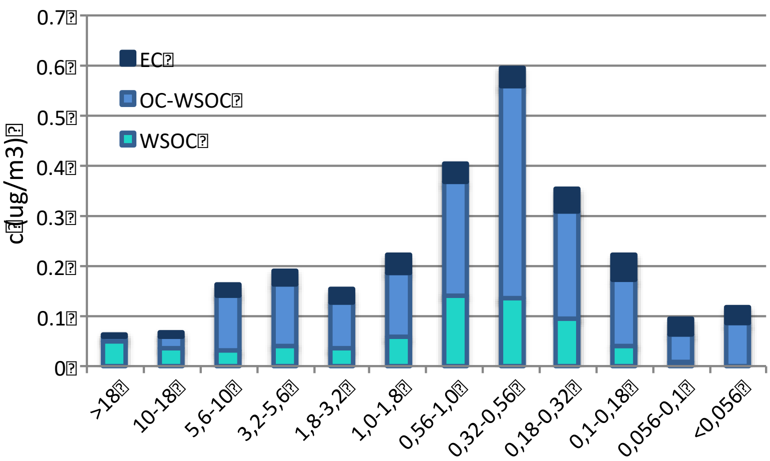


Figure 5.4.: Fractional distribution of EC, OC-WSOC and WSOC

WSOC from OC a non polar fraction of OC is obtained, thus giving better fractional profile (Fig. 5.4.).

Maxima of both organic fractions are found in stages 8 and 7 (d=0,32-1,0 µm. Although there are some WSOC in coarse fraction <10 µm, WSOC are not present in the ultrafine fraction. The contribution of WSOC to OC varies from 10% in lower fine fractions, to 38% at maximal level in stage 7 , while this contribution is up to 80% in coarse fraction < 10 µm.
Correlation of OC vs. (OC- WSOC) is also very good ($r=0,9837$, $p<0,05$), as well as EC vs. (OC- WSOC) being $r = 0,5867$, $p<0,05$), indicating similar fate in the atmosphere.

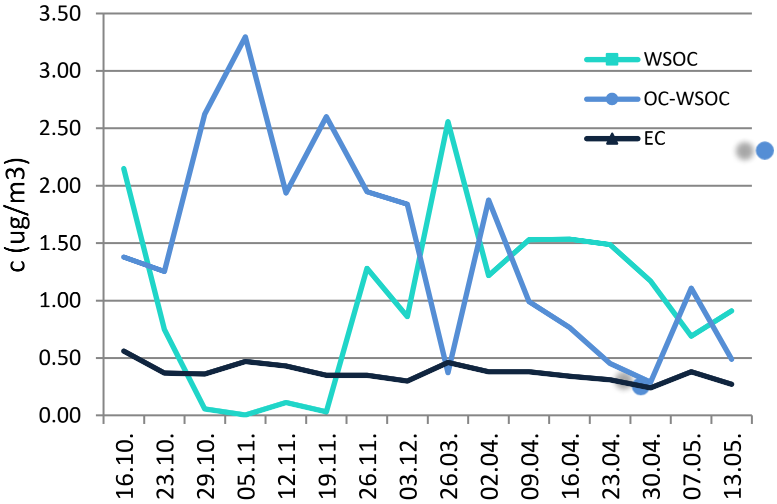


Figure.5.5. Distribution of EC, WSOC and nonpolar organic fraction (OC-WSOC) in weekly samples collected

The distribution of carbonaceous species (Fig. 5.5.) in composite samples is quite interesting: in spite of different meteorological situation in autumn and spring, with different PBL, as well as start of the heating season in mid November that lasted until end of March, the EC has practically no trend, with higher value recorded in the first week of study, and is attributed to Saharan sand contribution. The rise of nonpolar organics (OC-WSOC) coincides with the beginning of the heating season at the end of October, and is constantly declining since until the first week of May (Sample 15), when a second rise is observed. Such a situation is partially due to meteorological conditions with a lot of wind and rain. The profile of WSOC is the most interesting. After initially high values detected in samples with Saharan sand, with the beginning of heating season, their concentrations drops down to practically zero, and subsequently rising at the end of the autumn campaign. The start of spring campaign is characterized by a high concentration of WSOC (and inversely very low OC-WSOC), that subsequently drops down to almost constant levels for the following four weeks, following the further drop in the last three weeks of measurements.

PART II

High temporal resolution measurements

6. MEASUREMENT CAMPAIGN

The site chosen in Rijeka was on the roof of the Public Health building (45°19’56’’ N, 14°25’33” E, 34 m a.s.l.) in front of the entrance of the harbour (at about 500 m from the main routes and at about 1 km from the closest quay, in a straight line), as shown in Fig. 6.1. It was a background urban site, divided from the port commercial area with an intense cranes activity, by a busy seaside street (named Kresimirova).

Figure 6.1. Location of the measurement site in Rijeka.

Figure 6.2. Details of the outdoor cabinet used for high-temporal resolution measurements.

The measurement period was 28/03/2019-13/05/2019, collecting real-time measurements of meteorological parameters (wind velocity and direction, temperature, and relative humidity) and concentration of particles of different size ranging from 0.01 to 31 µm.

At same locations (Fig. 6.2) aerosol size-segregated sampling with a 10-stages micro-orifice impactor (MOUDI) for chemical analysis was done.

6.1. Material and methods

The equipment was arranged in an outdoor two-modular air-conditioned cabinet, as shown in (Fig. 6.3). Instruments were remotely controlled via PC. Specifically:

- an ultrasonic anemometer (Gill R3 at 100 Hz) coupled with a thermo-hygrometer (Rotronic MP100A, Campbell Scientific) were placed near the cabinet, measuring wind velocity, wind direction, temperature, and relative humidity at 1-min resolution;
- a CPC (Grimm 5.403) able to measure the total number of sub-micrometric particles, with 1-minute resolution, in the size range between 0.01 – 0.25 µm. Aerosol was sampled through a 70 cm-long sampling inlet and a portion of the main flow was injected into the CPC through a 50 cm-long conductive silicon tube and a diffusion dryer (silica gel cartridges) to reduce water vapour concentration before the CPC measurement (Merico et al., 2016);
- an OPC (Grimm 11-A) able to measure particle number size distributions in the size range 0.25-31 µm in 31 size channels, operating at controlled flow of 1.2 liter/minute. It used the same inlet as the CPC and it operated with 1-min time resolution. The internal software was also able to reconstruct mass size distributions as well as PM1, PM2.5, and PM10 mass concentration;
- a videocamera recording two frames per minute, detected ship movements synchronising them with concentration particle measurements. These data were compared/integrated with traffic schedule information provided by port authority.

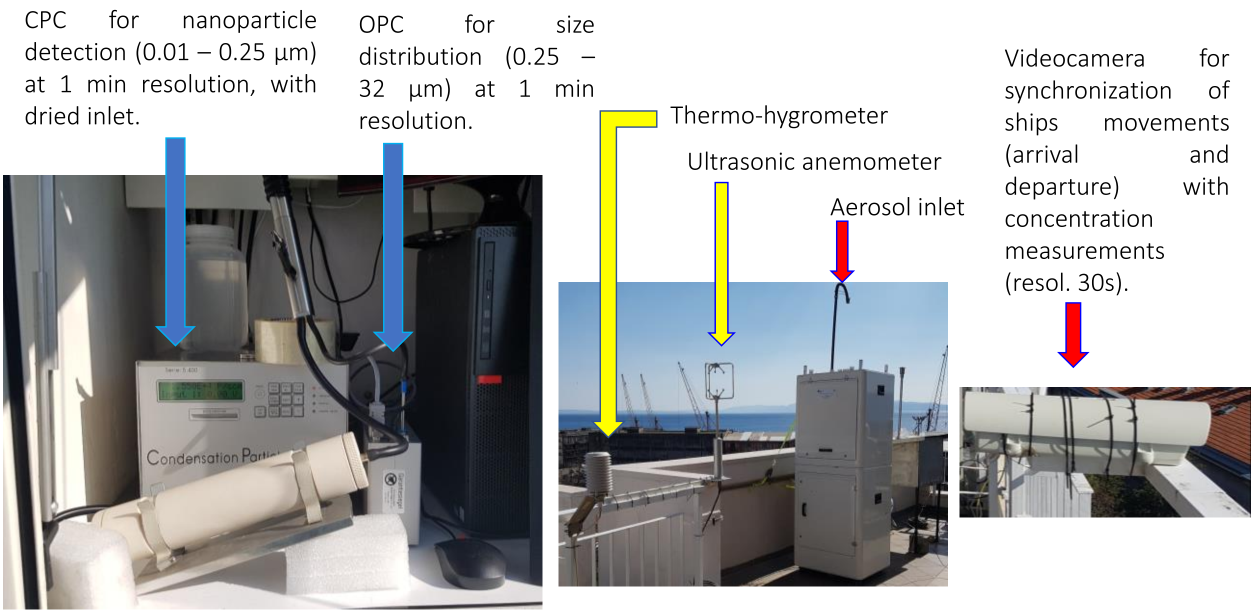


Figure 6.3. Instruments installed in (and nearby) the outdoor cabinet.

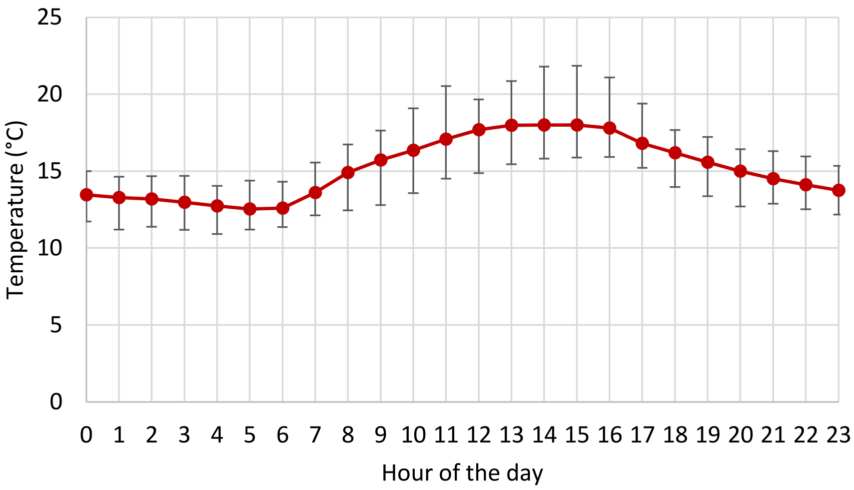
6.2. Result and discussion

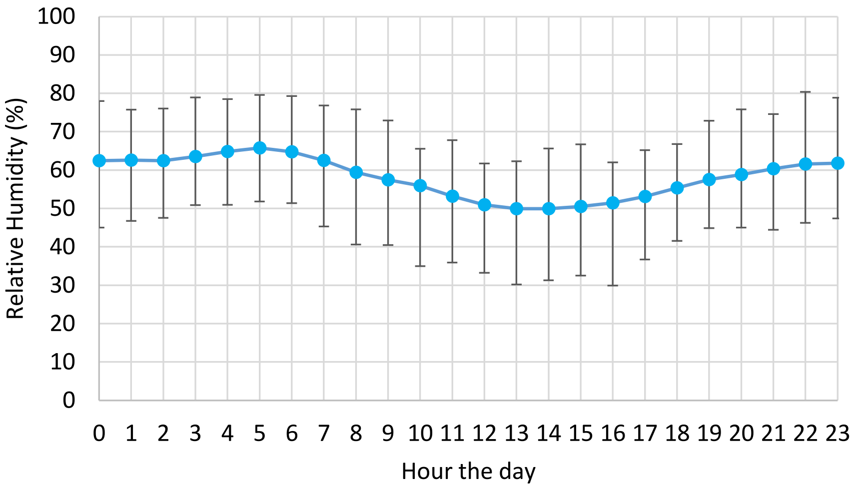
Collected data have been post-processed at 5, 30, 60 minutes averages, as well as daily averages, to summarize the variability in meteorological variables and particle concentration of different sizes.

Meteorology during the campaign

Meteorological data were acquired in addition to concentration measurements obtaining daily patterns of temperature, relative humidity, and wind velocity (Fig. 6.4). in particular, mean temperature of about 15°C was measured during the whole campaign and relative humidity was relatively low being between about 50% and 66%. Wind velocity was quite low, with an average value of 0.5 m/s, and maximum values during the day, between 8 a.m. and 7 p.m.

Looking at the wind rose of the whole campaign (Fig. 6.5), there was a dominant wind direction from ESE and a second wind direction sector from S to NE with slightly stronger winds from E-ENE. This indicated that the site was influenced mainly by Sirocco during the field campaign.


Temperature (°C)
25
20
15
10
5
0
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
Hour of the day


Relative Humidity (%)
100
90
80
70
60
50
40
30
20
10
0
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
Hour the day

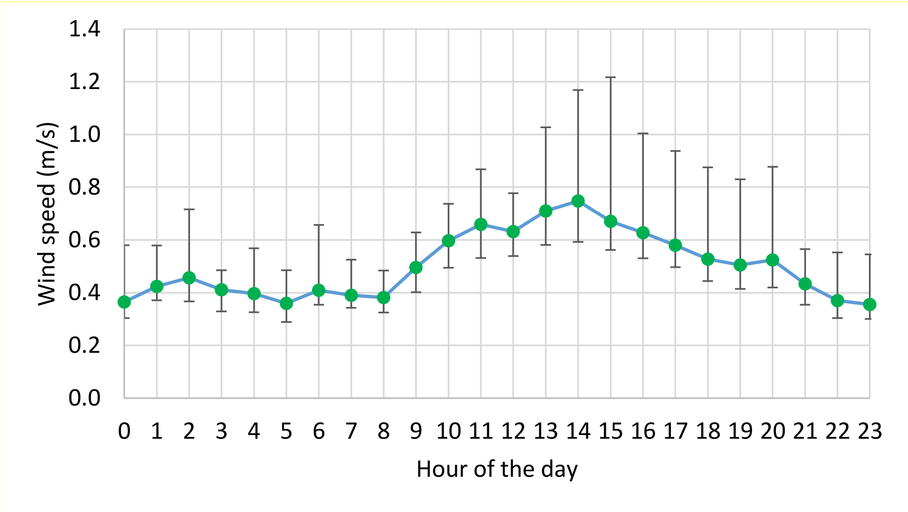


Figure 6.4. Daily pattern of the main meteorological parameters (with interquartile ranges indicated by the error bars).

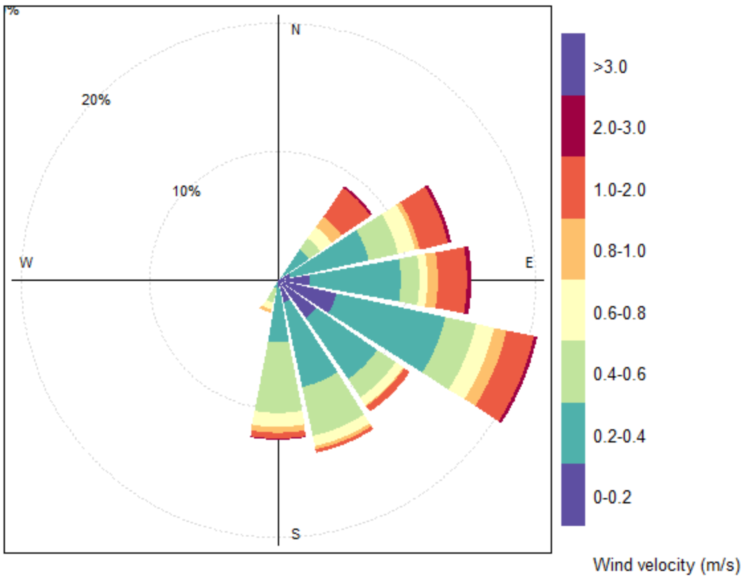


Figure 6.5. Wind rose of the Rijeka campaign.

From 24 April to 26 April 2019, an intense event of Saharan dust occurred on a large scale interesting also the studied area. This transport was confirmed by the back-trajectories of air masses calculated by Hysplit model (Fig. 6.6a) and the simulations of the Dust REgional Atmospheric Model (BSC-DREAM8b) (Fig. 6.6b). The event lead to a significant increase in the number of coarse particles, while a limited contribution on the concentration of nanoparticles was observed (Fig. 6.6c). Corresponding data were excluded by the analysis in order to avoid their influence on average concentrations.

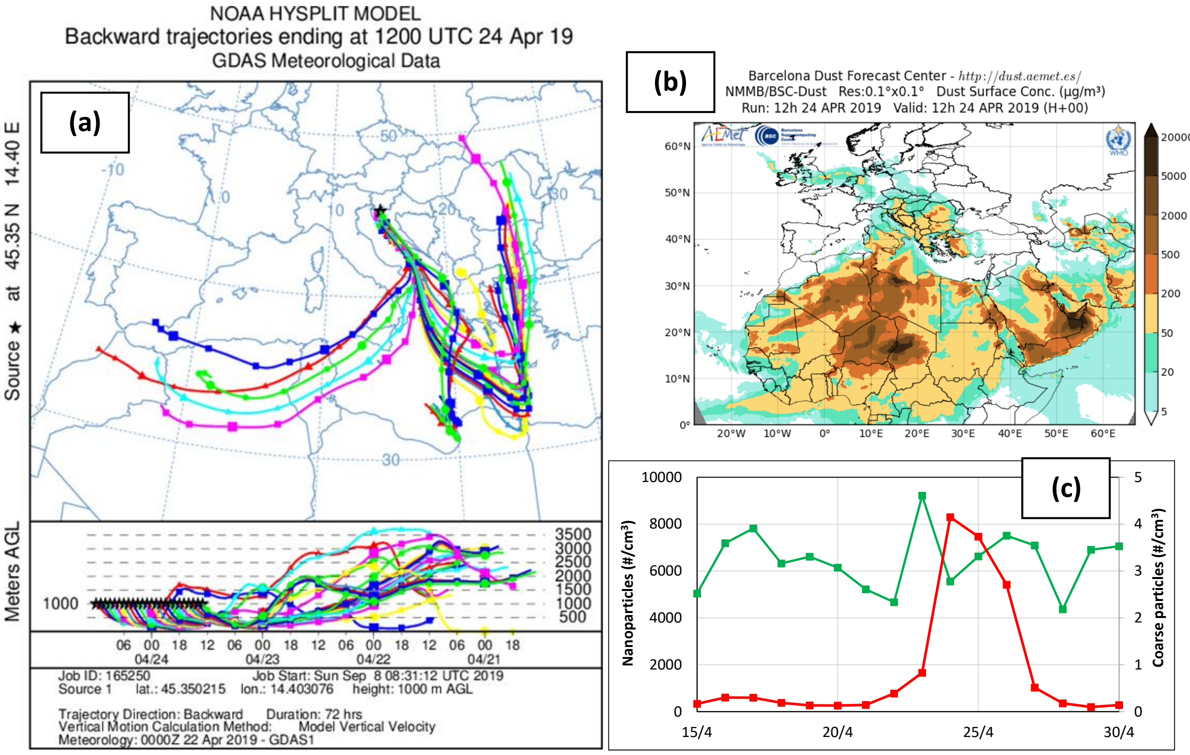


Figure 6.6. (a) Back-trajectories (http://arl.noaa.gov/ready/) and (b) NMMB/BSC images (https://ess.bsc.es/bsc-dust-daily-forecast); daily number concentration of coarse particles and nanoparticles for the Saharan dust event.

Particle number concentrations and size distributions

Comparison between daily PM_{10} concentrations measured by the OPC and those from air quality local network stations in Rijeka is reported. It should be noted that Mariscina is the closest station to the measurement site but at street level, therefore, the most influenced by vehicular traffic. On the other hand, Kresimirova 38 is the highest and the most remote station (Fig. 6.7a).

In general, there is a good agreement between time series, with some slightly higher values only for Mariscina and Kresimirova 38 station, and marked differences for the three days of the Saharan advection, reaching maximum values of about 87 µg/m^3 (Fig. 6.7b).

From OPC data, on daily basis for all campaign period, the average PM_{10} observed during the campaign was 14.4 µg/m^3, the average $PM_{2.5}$ was 13.1 µg/m^3, and the average PM_1 was 11.2 µg/m^3.

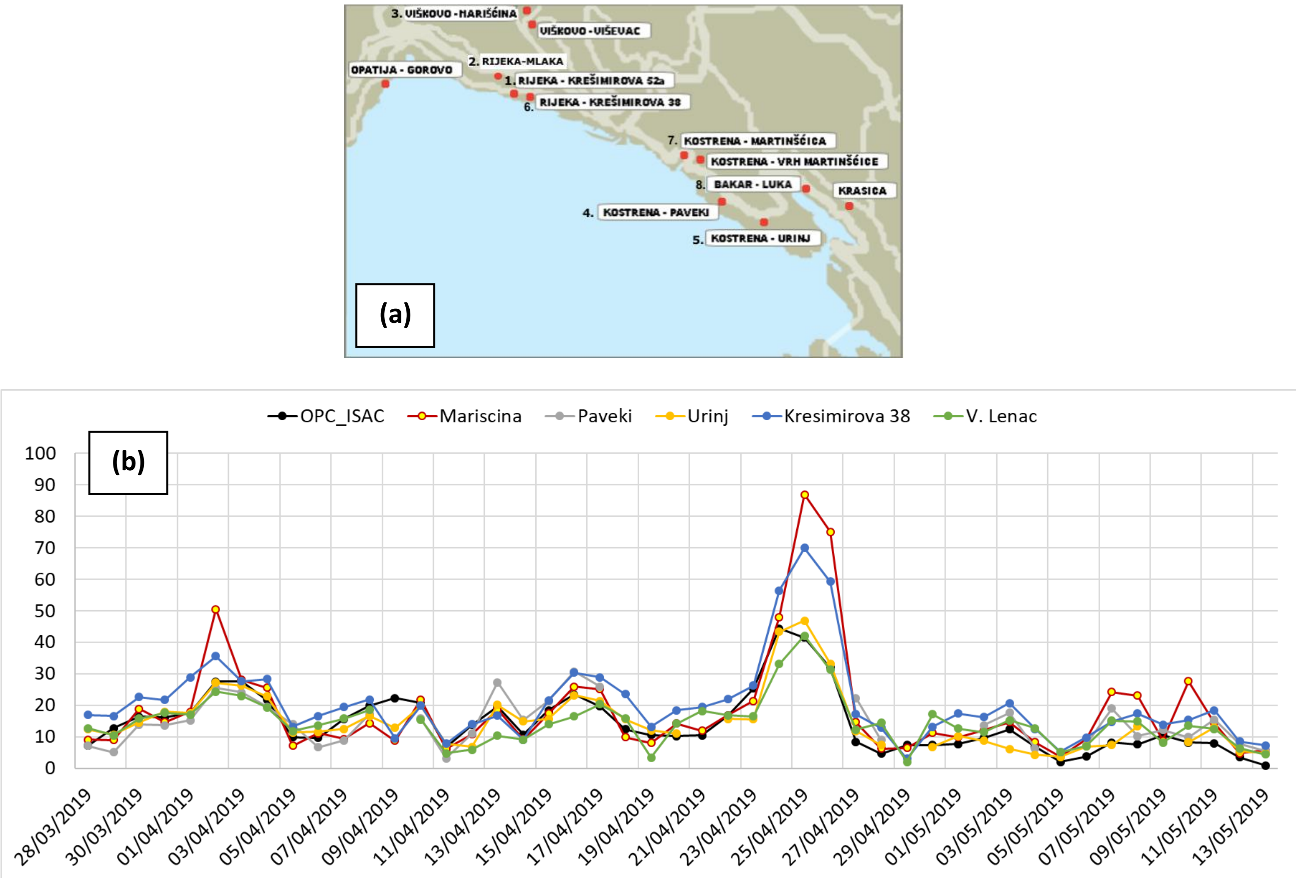


Figure 6.7. a) Air quality monitoring stations of local network and (b) comparison of daily PM_{10} measured by the OPC with results of some Rijeka stations.

Combining the CPC data with those of the OPC, average size distribution in number and in mass was obtained (Fig. 6.8). Three size ranges likely influenced by different sources and processes were identified:

- Nanoparticles (D< 0.25 µm)
- Fine particles (0.25<D<1 µm)
- Coarse particles (D>1 µm)

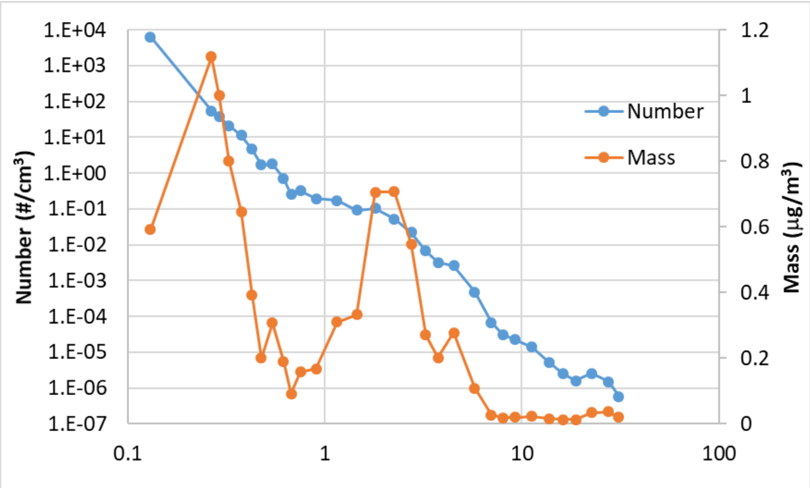


Figure 6.8. Average particle size distribution in number and in mass.

Daily pattern of nanoparticles and larger particles (D>0.25 µm) as concentration in number were compared (Fig. 6.9).

Nanoparticles revealed a daily trend with two peaks, however, the first one was the most significant accounting almost up to 12000 particles/cm^3 between 7 a.m. and 8 a.m. The second peak was less evident, in the evening hours (between 8 p.m. and 9 p.m.) with about 8000 particles/cm^3, followed by a low decrease in the night. Larger particles exhibited a comparable trend but maximum concentration (187 particles/cm^3) were detected starting from late afternoon reaching mean values in the night.

These patterns, typically observed in an urban background site like this, are influenced by emissions of some sources in specific hours (i.e. road traffic) with a not evident effect of the dynamics of the boundary-layer.

Similarly, weekly pattern of the same size ranges were obtained (Fig. 6.9). Nanoparticles did not show clear trend during the week even if the highest value was on Tuesday (about 8,500 particles/cm^3) and the minimum ones during weekend. Larger particles also had relatively high values in the first part of the week with a rapid decrease during following days.

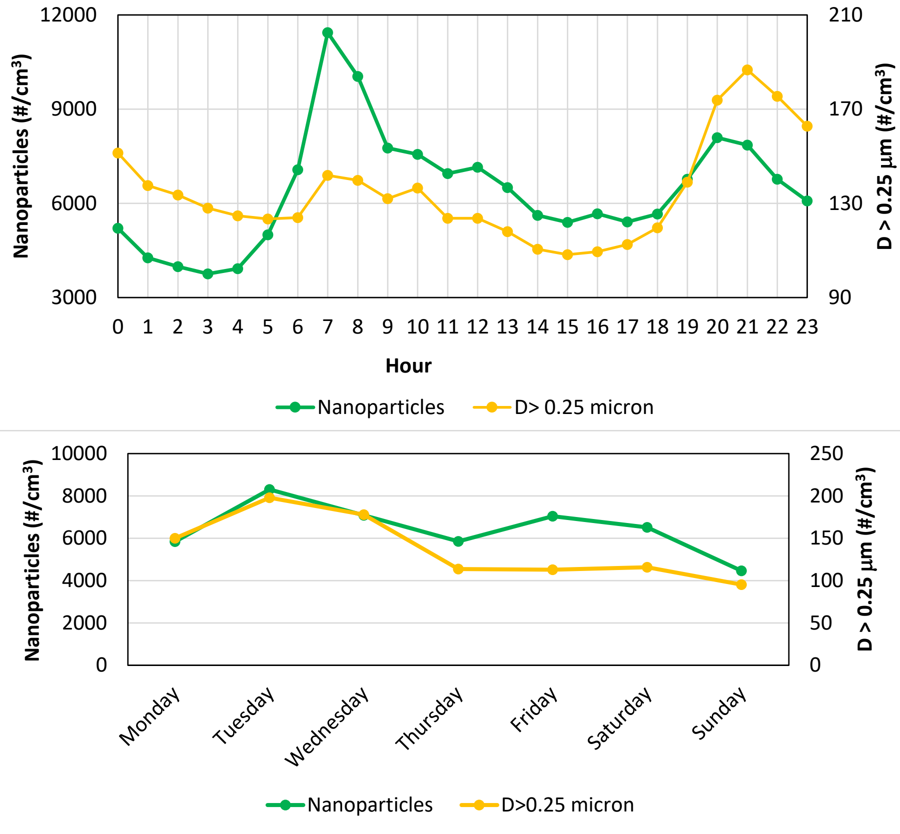


Figure 6.9. Average daily patterns of number concentrations (top) and weekly pattern (bottom) for nanoparticles and larger particles.

Analogously, daily pattern in term of mass concentration for PM1 and for coarse particles (PM10-1) were calculated (Fig. 6.10).

Daily trend of PM_1 was almost superimposable with that in number for large particles with higher values (about 15 µg/m^3) from late afternoon to the night hours. Instead, the pattern of PM_{10-1} (coarse fraction) is different with a broad increase in diurnal hours in the morning (about 4 µg/m^3) and (less evident) in the afternoon.

Weekly patterns for mass concentration in the same size ranges are also reported (Fig. 6.10) showing maximum values (from 4 to 6 µg/m^3) found in the middle of the week for coarse particles. On the other hand, PM_1 increased in the first part of the week (Tuesday and Wednesday) with concentration of about 15 µg/m^3.

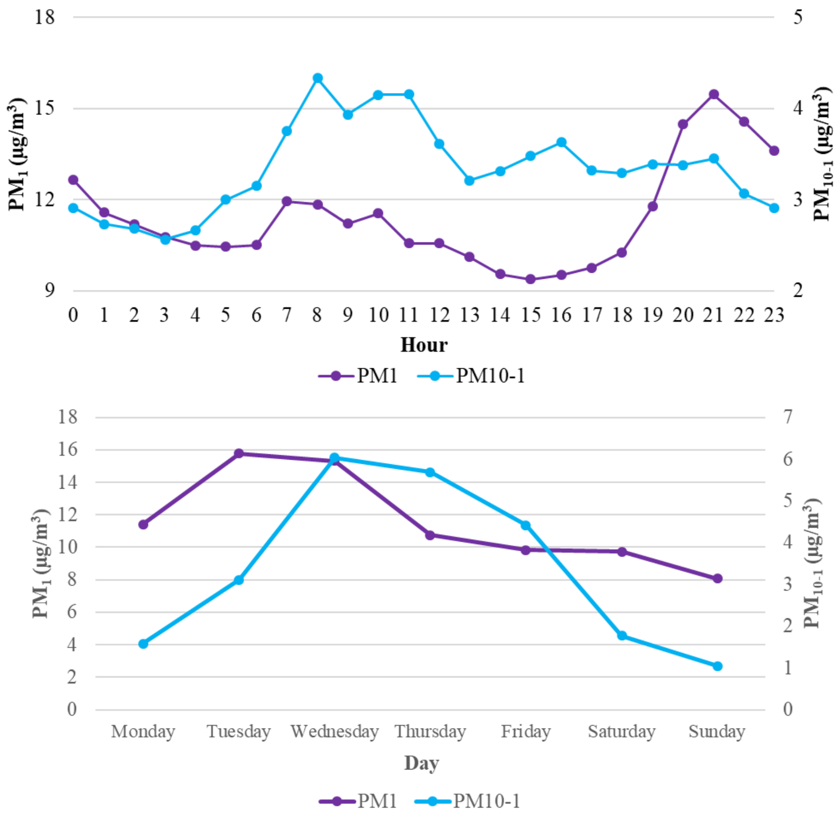


Figure 6.10. Average daily patterns of mass concentrations (top) and weekly pattern (bottom) for PM_{10} and PM_{10-1}.

ECOlogical supporting for traffic Management in cOastal areas By using an InteLlIgenT sYstem

AXIS 4: Maritime Transport
Objective 4.1: Improve the quality, safety and environmental sustainability of marine and coastal transport services and nodes by promoting multimodality in the Programme area

ANNEX

Detailed concentration data

LIST OF TABLES

TABLE A1

Particulate concentration data (µg/m^3)

Sample ID	PM
S01St01	ND
S01St02	2,38
S01St03	8,72
S01St04	18,83
S01St05	8,73
S01St06	24,45
S01St07	22,22
S01St08	19,87
S01St09	17,27
S01St10	19,32
S01St11	15,37
S01	*157,16*
S02St01	1,84
S02St02	1,53
S02St03	3,03
s02St04	5,70
S02St05	3,55
S21St06	ND
S02St07	2,18
S02St08	1,65
S02St09	1,89
S02St10	0,70
S02St11	0,12
S02	*22,19*
S03St01	1,93
S03St02	1,33
S03St03	3,25
S03St04	7,04
S03St05	4,71
S03St06	3,12
S03St07	2,18
S03St08	1,27
S03St09	1,04
S03St10	0,49
S03St11	0,22
S03	*26,58*
S04St01	ND
S04St02	0,34
S04St03	1,65
S04St04	3,25
S04St05	2,11
S04St06	1,37
S04St07	1,85
S04St08	2,69
S04St09	1,23
S04St10	0,46
S04St11	0,01
S04	*14,96*

Sample ID	PM
S05St01	0,47
S05St02	0,50
S05St03	1,20
S05St04	1,69
S05St05	0,94
S05St06	0,79
S04St07	1,84
S04St08	1,83
S04St09	0,55
S04St10	ND
S04St11	ND
S05	*9,82*
S06St01	2,36
S06St02	1,51
S06St03	1,92
S06St04	3,87
S06St05	1,97
S06St06	3,86
S06St07	2,82
S06St08	6,09
S06St09	2,54
S06St10	1,99
S06St11	2,90
S06	*51,46*
S07St01	0,69
S07St02	0,61
S07St03	1,18
S07St04	2,40
S07St05	0,95
S07St06	1,69
S07St07	2,51
S07St08	3,49
S07St09	ND
S07St10	0,07
S07St11	5,20
S07	*18,79*
S08St01	0,37
S08St02	0,09
S08St03	0,59
S08St04	1,56
S08St05	0,83
S08St06	1,28
S08St07	1,71
S08St08	3,25
S87St09	1,27
S08St10	0,67
S08St11	0,33
S08	*11,94*

Sample ID	PM
S09St01	1,73
S09St02	1,30
S09St03	2,06
s09St04	2,33
S09St05	1,17
S09St06	2,83
S09St07	2,27
S09St08	4,83
S09St09	2,78
S09St10	2,08
S09St11	0,76
S09St12	13,28
St9	*37,42*
S10St01	1,68
S10St02	0,78
S10St03	2,64
S10St04	3,56
S10St05	1,78
S10St06	1,75
S10St07	2,55
S10St08	3,13
S10St09	1,88
S10St10	0,66
S10St11	ND
S10St12	3,50
S10	*23,90*
S11St01	0,84
S11St02	0,82
S11St03	0,90
S11St04	1,47
S11St05	1,29
S11St06	1,48
S11St07	2,82
S11St08	2,86
S11St09	1,39
S11St10	0,52
S11St11	ND
S11St12	2,19
S15	*16,58*
S12St01	1,39
S12St02	1,53
S12St03	2,73
S12St04	3,02
S12St05	1,31
S12St06	1,35
S12St07	1,36
S12St08	2,98
S12St09	1,37
S12St10	0,17
S12St11	0,12
S12St12	1,12
S12	*18,44*

Sample ID	PM
S13St01	1,63
S13St02	1,90
S13St03	4,68
S13St04	10,64
S13St05	7,43
S13St06	3,56
S13St07	1,92
S13St08	1,84
S13St09	0,85
S13St10	0,36
S13St11	0,05
S13St12	0,64
S13	*35,50*
S14St01	0,55
S14St02	0,49
S14St03	0,81
S14St04	1,53
S14St05	0,53
S14St06	0,52
S14St07	0,81
S14St08	1,22
S14St09	0,26
S14St10	0,33
S14St11	ND
S14St12	0,55
S15	*7,60*
S15St01	0,32
S15St02	0,26
S15St03	0,88
S15St04	1,51
S15St05	1,03
S15St06	0,49
S15St07	0,61
S15St08	1,89
S15St09	0,92
S15St10	0,38
S15St11	ND
S15St12	0,53
S15	*8,83*
S16St01	0,47
S16St02	0,58
S16St03	1,33
S16St04	1,56
S16St05	1,13
S16St06	0,53
S16St07	0,87
S16St08	1,28
S16St09	0,45
S16St10	0,20
S16St11	ND
S16St12	0,45
S16	*8,84*

TABLE A2
Ion concentration data (ng/m³)

Sample ID	PO43-	CL-	NO3-	SO42-	Na+	NH4+	K+	Ca2+	Mg2+
S01St01	97,884	29,003	36,253	0,000	275,525	ND	18,127	43,504	14,501
S01St02	159,514	29,003	25,377	0,000	116,010	ND	7,251	246,522	10,876
S01St03	94,258	25,377	58,005	14,501	116,010	ND	18,127	427,788	25,377
s01St04	116,010	18,127	97,884	21,752	137,762	ND	21,752	384,284	10,876
S01St05	58,005	29,003	58,005	58,005	79,757	ND	7,251	257,398	ND
S01St06	18,127	36,253	39,879	170,390	61,630	18,127	14,501	184,891	ND
S01St07	61,630	14,501	39,879	424,163	65,256	90,633	14,501	188,517	ND
S01St08	0,000	25,377	43,504	464,041	65,256	112,385	21,752	177,641	ND
S01St09	10,876	29,003	39,879	188,517	61,630	ND	21,752	184,891	ND
S01St10	50,755	21,752	29,003	29,003	47,129	ND	7,251	134,137	ND
S01St11	3,625	21,752	14,501	0,000	68,881	ND	7,251	105,134	ND
S01	*670,685*	*279,150*	*482,168*	*1370,372*	*1094,848*	*221,145*	*159,514*	*2334,708*	*61,630*
S02St01	221,154	61,243	20,414	22,966	ND	ND	5,104	431,251	10,207
S02St02	255,178	91,864	25,518	0,000	280,696	ND	5,104	224,557	ND
S02St03	258,580	188,832	40,828	12,759	347,042	ND	10,207	130,141	ND
s02St04	105,474	533,322	181,176	68,898	505,252	ND	33,173	183,728	10,207
S02St05	282,397	387,871	158,210	61,243	459,320	ND	15,311	173,521	25,518
S21St06	142,900	107,175	53,587	45,932	250,074	ND	15,311	84,209	15,311
S02St07	47,633	15,311	28,070	132,693	155,659	10,207	30,621	56,139	10,207
S02St08	139,497	ND	20,414	160,762	140,348	28,070	17,862	51,036	10,207
S02St09	44,231	25,518	10,207	79,105	117,382	17,862	20,414	15,311	ND
S02St10	27,219	ND	ND	17,862	122,485	7,655	10,207	10,207	ND
S02St11	ND	15,311	ND	ND	173,521	ND	17,862	112,278	20,414
S02	*1524,263*	*1426,445*	*538,426*	*602,220*	*2551,780*	*63,794*	*181,176*	*1472,377*	*102,071*
S03St01	13,335	160,490	48,845	88,386	48,845	ND	ND	81,408	13,956
S03St02	15,816	74,430	9,304	20,933	44,193	ND	13,956	207,009	37,215
S03St03	21,399	267,483	39,541	32,563	109,319	ND	20,933	162,816	13,956
S03St04	6,823	507,055	155,838	109,319	239,572	ND	6,978	437,277	44,193
S03St05	15,506	288,417	134,905	106,993	181,423	ND	11,630	318,654	25,585
S03St06	8,994	109,319	167,468	118,623	53,497	6,978	ND	102,341	11,630
S03St07	9,614	20,933	13,956	200,031	48,845	39,541	74,430	269,809	34,889
S03St08	ND	ND	ND	148,860	ND	37,215	ND	137,231	ND
S03St09	7,443	13,956	ND	76,756	ND	25,585	ND	62,800	ND
S03St10	9,924	ND	ND	20,933	ND	11,630	ND	9,304	ND
S03St11	10,854	13,956	ND	ND	ND	6,978	ND	88,386	ND
S03	*119,708*	*1456,040*	*569,856*	*923,399*	*725,694*	*127,927*	*127,927*	*1877,035*	*181,423*
S04St01	10,488	50,257	30,591	8,740	32,776	ND	6,555	28,406	8,740
S04St02	10,780	3620,671	78,663	491,642	1815,798	ND	58,997	408,609	122,364
S04St03	16,607	139,845	102,699	50,257	150,770	ND	13,110	900,251	310,281
S04St04	12,819	107,069	255,654	54,627	142,030	ND	10,925	257,839	10,925
S04St05	6,992	61,182	131,105	50,257	83,033	ND	8,740	128,919	13,110
S04St06	22,433	30,591	43,702	98,328	80,848	13,100	48,072	371,463	54,627
S04St07	ND	ND	15,296	334,317	13,110	87,400	10,925	174,806	8,740
S04St08	16,315	4,370	13,110	458,866	24,036	135,500	19,666	122,364	8,740
S04St09	19,520	ND	ND	207,582	10,925	61,200	15,296	50,257	4,370
S04St10	13,402	ND	ND	89,588	8,740	28,400	4,370	13,110	2,185
S04St11	12,236	ND	ND	6,555	ND	8,700	ND	ND	ND
S04	*141,593*	*4013,984*	*670,818*	*1850,759*	*2362,067*	*334,300*	*196,657*	*2456,025*	*544,084*

TABLE A2
Ion concentration data (ng/m³)

Sample ID	PO43-	CL-	NO3-	SO42-	Na+	NH4+	K+	Ca2+	Mg2+
S05St01	9,346	13,143	15,334	17,525	19,715	ND	ND	ND	ND
S05St02	13,436	8,762	ND	15,334	10,953	ND	ND	499,452	37,240
S05St03	10,807	8,762	28,478	13,143	ND	ND	ND	155,531	ND
S05St04	3,505	15,334	83,242	26,287	28,478	ND	4,381	131,435	ND
S05St05	9,346	ND	37,240	15,334	ND	ND	ND	41,621	ND
S05St06	ND	ND	19,715	56,955	ND	ND	ND	ND	ND
S05St07	0,876	ND	21,906	210,296	ND	61,336	4,381	ND	ND
S05St08	8,178	ND	28,478	216,867	ND	67,908	6,572	ND	8,762
S05St09	9,346	ND	8,762	83,242	21,906	26,287	8,762	120,482	60,000
S05St10	14,020	ND	ND	35,049	15,334	13,143	28,478	19,715	30,000
S05St11	7,594	ND	ND	ND	ND	ND	ND	ND	30,000
S05	***86,455***	***46,002***	***243,154***	***690,033***	***96,386***	***168,675***	***52,574***	***968,237***	***166,002***
S06St01	ND	8,710	8,710	ND	ND	ND	ND	629,287	34,839
S06St02	3,048	0,000	ND	ND	ND	ND	ND	87,099	ND
S06St03	5,226	13,065	ND	ND	ND	ND	ND	ND	ND
S06St04	3,048	74,034	28,307	30,484	39,194	ND	ND	56,614	ND
S06St05	8,928	32,662	21,775	32,662	26,130	ND	ND	113,228	13,065
S06St06	0,653	ND	ND	43,549	ND	8,710	ND	17,420	ND
S06St07	3,919	ND	ND	148,068	ND	37,017	6,532	8,710	ND
S06St08	2,831	2,177	ND	187,262	13,065	58,792	19,597	324,442	34,839
S06St09	0,871	ND	ND	74,034	ND	23,952	ND	ND	6,532
S06St10	6,750	ND	ND	37,017	ND	15,242	ND	ND	ND
S06St11	3,266	ND	ND	ND	ND	6,532	ND	ND	5,371
S06	***38,541***	***130,648***	***58,792***	***553,076***	***78,389***	***150,245***	***26,130***	***1236,799***	***94,647***
S07St01	1,015	30,444	ND	20,296	126,850	ND	ND	499,789	76,211
S07St02	1,776	32,981	ND	17,759	50,740	ND	2,537	134,461	ND
S07St03	2,030	38,055	73,573	27,907	96,406	ND	0,000	83,721	ND
S07St04	1,776	38,055	32,981	35,518	55,814	ND	7,611	63,425	ND
S07St05	3,044	ND	ND	22,833	32,981	ND	5,074	43,129	ND
S07St06	1,776	ND	12,685	53,277	2,537	ND	ND	15,222	ND
S07St07	1,776	20,296	53,277	218,182	22,833	12,685	ND	45,666	6,532
S07St08	1,015	15,222	32,981	238,478	15,222	35,518	7,611	7,611	ND
S07St09	1,522	40,592	25,370	55,814	43,129	ND	17,759	ND	ND
S07St10	2,283	38,055	15,222	63,425	22,833	ND	58,351	ND	ND
S07St11	2,030	30,444	ND	ND	22,833	ND	ND	ND	ND
S07	***20,042***	***284,144***	***246,089***	***753,488***	***492,178***	***48,203***	***98,943***	***893,023***	***82,744***
S08St01	1,015	30,444	ND	20,296	126,850	ND	ND	499,789	76,211
S08St02	1,776	32,981	ND	17,759	50,740	ND	2,537	134,461	ND
S08St03	2,030	38,055	73,573	27,907	96,406	ND	ND	83,721	ND
S08St04	1,776	38,055	32,981	35,518	55,814	ND	7,611	63,425	ND
S08St05	3,044	ND	ND	22,833	32,981	ND	5,074	43,129	ND
S08St06	1,776	ND	12,685	53,277	2,537	ND	ND	15,222	ND
S08St07	1,776	20,296	53,277	218,182	22,833	12,685	ND	45,666	6,532
S08St08	1,015	15,222	32,981	238,478	15,222	35,518	7,611	7,611	ND
S08St09	1,522	40,592	25,370	55,814	43,129	ND	17,759	ND	ND
S08St10	2,283	38,055	15,222	63,425	22,833	ND	58,351	ND	ND
S08St11	2,030	30,444	ND	ND	22,833	ND	ND	ND	ND
S08	***20,042***	***284,144***	***246,089***	***753,488***	***492,178***	***48,203***	***98,943***	***893,023***	***82,744***

TABLE A2

Ion concentration data (ng/m^3)

Sample ID	PO43-	CL-	NO3-	SO42-	Na+	NH4+	K+	Ca2+	Mg2+
S09St01	0,626	42,230	23,461	16,423	32,846	ND	ND	39,884	7,038
S09St02	5,005	7,038	ND	23,461	35,192	ND	ND	49,269	7,038
S09St03	1,251	25,807	16,423	25,807	35,192	ND	ND	82,115	11,731
s09St04	0,626	37,538	46,923	37,538	39,884	ND	ND	133,730	18,769
S09St05	ND	32,846	60,999	35,192	32,846	ND	ND	98,538	11,731
S09St06	1,564	28,154	25,807	46,923	18,769	ND	ND	60,999	7,038
S09St07	ND	23,461	39,884	150,152	16,423	ND	ND	60,999	7,038
S09St08	1,564	11,731	25,807	253,382	11,731	53,961	14,077	103,230	14,077
S09St09	0,626	7,038	28,154	117,307	ND	16,423	7,038	63,346	7,038
S09St10	1,564	ND	9,385	42,230	21,115	ND	ND	ND	ND
S09St11	1843,435	49,269	9,385	63,346	225,229	ND	ND	1475,718	82,115
S09St12	198,014	14,077	ND	ND	4,692	ND	304,997	86,807	ND
S09	***2054,274***	***279,190***	***286,228***	***811,762***	***473,919***	***70,384***	***326,112***	***2254,634***	***173,614***
S10St01	2,779	27,792	13,896	27,792	23,160	ND	ND	90,323	ND
S10St02	4,014	32,424	6,948	25,476	23,160	ND	ND	39,372	ND
S10St03	4,941	111,167	64,847	41,688	74,111	ND	ND	138,959	13,896
S10St04	5,250	162,118	229,282	69,479	143,591	ND	ND	111,167	13,896
S10St05	7,102	32,424	92,639	34,740	39,372	ND	ND	30,108	6,948
S10St06	7,102	6,948	67,163	148,222	34,740	ND	ND	53,267	11,580
S10St07	4,632	ND	37,056	280,233	9,264	39,372	13,896	20,844	ND
S10St08	1,544	4,632	34,740	324,237	6,948	71,795	6,948	23,160	6,948
S10St09	0,618	4,632	37,056	155,170	2,316	23,160	9,264	ND	ND
S10St10	139,885	60,215	34,740	90,323	162,118	ND	ND	23,160	ND
S10St11	8,029	0,000	11,580	16,212	ND	ND	ND	ND	ND
S10St12	0,926	0,000	ND	ND	ND	ND	ND	9,264	ND
S10	***186,822***	***442,351***	***629,946***	***1213,572***	***518,779***	***134,327***	***30,108***	***539,622***	***53,267***
S11St01	0,943	103,696	18,854	37,708	120,193	ND	ND	1616,717	113,123
S11St02	85,785	160,258	11,784	40,064	299,305	ND	ND	259,240	7,070
S11St03	12,255	47,135	11,784	16,497	70,702	ND	9,427	136,690	ND
S11St04	7,542	25,924	21,211	16,497	30,637	ND	ND	122,550	ND
S11St05	8,484	16,497	18,854	54,205	25,924	ND	ND	641,031	40,064
S11St06	6,285	16,497	18,854	223,889	47,135	28,281	ND	1095,880	21,211
S11St07	10,055	16,497	23,567	381,790	16,497	103,696	4,713	216,819	ND
S11St08	7,542	21,211	21,211	273,381	30,637	82,486	7,070	108,410	ND
S11St09	4,713	ND	14,140	103,696	ND	30,637	ND	63,632	ND
S11St10	6,599	25,924	9,427	23,567	14,140	ND	ND	49,491	ND
S11St11	1,571	16,497	ND	ND	2,357	ND	ND	ND	ND
S11St12	2,828	23,567	9,427	2,357	30,637	ND	21,211	11,784	ND
S11	***154,602***	***473,703***	***179,112***	***1173,652***	***688,165***	***245,100***	***42,421***	***4322,244***	***181,468***
S12St01	2,789	11,620	16,268	9,296	39,507	ND	4,648	148,733	16,268
S12St02	1,239	6,972	2,324	4,648	34,859	ND	0,000	855,217	51,127
S12St03	26,028	51,127	20,916	34,859	190,565	ND	6,972	248,664	ND
S12St04	9,296	30,211	92,958	27,888	67,395	ND	ND	211,480	13,944
S12St05	7,127	25,564	83,663	44,155	65,071	ND	ND	155,705	9,296
S12St06	8,056	2,324	18,592	55,775	25,564	ND	ND	76,691	ND
S12St07	10,225	9,296	23,240	230,072	44,155	51,127	ND	79,015	6,972
S12St08	8,366	13,944	23,240	374,158	32,535	116,198	4,648	58,099	6,972
S12St09	8,986	11,620	18,592	183,593	34,859	60,423	ND	27,888	ND
S12St10	6,817	16,268	13,944	60,423	13,944	13,944	6,972	13,944	ND
S12St11	10,225	ND	6,972	2,324	9,296	ND	4,648	ND	ND
S12St12	6,507	6,972	6,972	ND	6,972	ND	ND	16,268	ND
S12	***105,663***	***185,917***	***327,678***	***1027,190***	***564,722***	***241,692***	***27,888***	***1891,703***	***104,578***

TABLE A2

Ion concentration data (ng/m³)

Sample ID	PO43-	CL-	NO3-	SO42-	Na+	NH4+	K+	Ca2+	Mg2+
S13St01	9,636	37,302	16,320	9,325	6,994	ND	ND	44,155	11,620
S13St02	9,325	32,639	16,320	11,657	32,639	ND	ND	34,859	9,296
S13St03	9,015	202,829	41,965	72,272	118,900	ND	ND	153,381	25,564
S13St04	11,812	755,362	263,444	202,829	442,959	ND	ND	332,326	51,127
S13St05	12,434	398,663	235,468	118,900	296,083	ND	4,663	199,861	39,507
S13St06	2,487	188,841	172,521	114,237	158,533	ND	ND	85,987	20,916
S13St07	268,262	172,521	37,302	454,616	610,818	11,657	11,657	376,482	74,367
S13St08	0,933	81,598	11,657	223,811	51,290	41,965	23,314	81,339	6,972
S13St09	15,853	ND	4,663	100,249	ND	16,320	4,663	ND	ND
S13St10	38,234	93,255	4,663	37,302	121,231	ND	34,970	23,240	6,972
S13St11	13,677	44,296	ND	ND	48,959	ND	ND	ND	ND
S13St12	1,865	212,154	ND	16,320	128,225	ND	ND	297,467	44,155
S13	***393,534***	***2219,459***	***804,321***	***1361,517***	***2016,630***	***69,941***	***79,266***	***1629,096***	***290,495***
S14St01	68,658	87,746	55,419	50,800	284,021	ND	ND	46,182	ND
S14St02	16,318	18,473	4,618	25,400	69,273	ND	ND	20,782	6,927
S14St03	15,702	27,709	23,091	20,782	34,637	ND	ND	36,946	9,236
S14St04	17,857	41,564	71,583	32,328	80,819	ND	ND	399,477	46,182
S14St05	12,931	2,309	41,564	20,782	16,164	ND	6,927	55,419	ND
S14St06	12,007	6,927	16,164	50,800	23,091	ND	ND	57,728	6,927
S14St07	10,776	11,546	11,546	115,456	30,018	ND	ND	57,728	6,927
S14St08	13,547	16,164	9,236	124,692	30,018	13,855	13,855	4,618	ND
S14St09	4,310	ND	4,618	85,437	13,855	ND	4,618	858,990	46,182
S14St10	14,163	ND	4,618	41,564	2,309	ND	ND	69,273	ND
S14St11	14,470	ND	ND	11,546	2,309	ND	ND	ND	ND
S14St12	1,232	ND	ND	ND	ND	ND	ND	4,618	ND
S14	***201,970***	***212,438***	***242,457***	***579,587***	***586,515***	***13,855***	***25,400***	***1611,761***	***122,383***
S15St01	20,335	25,960	58,410	29,205	51,920	ND	ND	ND	ND
S15St02	16,441	29,205	29,205	35,695	42,185	ND	ND	51,920	ND
S15St03	18,605	45,430	38,940	35,695	87,615	ND	ND	38,940	ND
S15St04	19,903	51,920	74,635	35,695	68,145	ND	ND	22,715	ND
S15St05	5,625	42,185	97,350	42,185	81,125	ND	ND	35,695	9,735
S15St06	19,903	22,715	29,205	81,125	64,900	ND	6,490	584,100	48,675
S15St07	22,931	ND	16,225	165,495	100,595	ND	48,675	116,820	ND
S15St08	14,711	22,715	19,470	142,780	51,920	ND	ND	334,235	22,715
S15St09	23,364	22,715	22,715	314,765	55,165	61,655	ND	629,529	16,225
S15St10	20,335	25,960	16,225	84,370	19,470	ND	ND	64,900	ND
S15St11	16,441	22,715	9,735	25,960	12,980	ND	ND	623,039	22,715
S15St12	17,307	22,715	ND	ND	ND	ND	ND	55,165	ND
S15	***215,900***	***334,235***	***412,115***	***992,969***	***636,019***	***61,655***	***55,165***	***2557,058***	***120,065***
S16St01	13,972	ND	21,932	14,622	43,865	ND	ND	399,659	24,369
S16St02	12,997	ND	4,874	7,311	ND	ND	ND	116,973	ND
S16St03	36,717	46,302	21,932	26,806	53,613	ND	ND	138,906	41,428
S16St04	27,944	58,487	46,302	26,806	60,924	ND	ND	107,226	ND
S16St05	28,269	31,680	56,050	31,680	70,671	ND	ND	565,371	29,243
S16St06	33,467	ND	12,185	51,176	19,496	ND	ND	141,343	ND
S16St07	33,467	4,874	4,874	151,091	31,680	26,806	4,874	73,108	ND
S16St08	29,893	ND	4,874	177,897	70,671	41,428	4,874	21,932	ND
S16St09	36,717	4,874	4,874	90,167	73,108	17,059	ND	2,437	ND
S16St10	39,966	26,806	7,311	60,924	190,082	7,311	ND	4,874	ND
S16St11	33,467	2,437	ND	14,622	121,847	ND	17,059	4,874	ND
S16St12	28,918	ND	ND	ND	109,662	ND	7,311	ND	ND
S16	***355,794***	***175,460***	***185,208***	***653,101***	***845,620***	***92,604***	***34,117***	***1576,703***	***95,041***

TABLE A3

Method detection limits (MDL) and quantification limits (MQL) for ions (ng/m^3)

ion	MDL	MQL
Cl^-	7,2	24
SO_4^{2-}	4,8	12
NO_3^-	7,2	24
PO_4^{3-}	0,48	1,58
Na^+	7,2	24
NH_4^+	2,4	9,6
K^+	2,4	7,2
Ca^{2+}	7,2	24
Mg^{2+}	4,8	12

TABLE A4

Distribution parameters (median, 1st and 3rd quartiles) of the concentration of ions in various dimensional classes (ng/m3)

Parameter		St 01 >18 µm	St 02 18-10 µm	St 03 10-5,6 µm	St 04 5,6-3,2 µm	St 05 3,2-1,8 µm	St 06 1,8-1,0 µm	St 07 1,0-0,56 µm	St 08 0,56-0,32µm	St 09 0,32-0,18 µm	St10 0,18-0,1 µm	St11 0,1-0,056µm	St12 <0,056
	Mediane	9,49	11,89	13,98	8,42	8,71	7,58	9,83	5,19	8,21	13,71	9,13	4,67
PO43-	1st quartile	1,01	3,53	5,08	3,28	6,31	1,78	1,78	1,01	1,52	6,67	2,65	1,55
	3rd quartile	17,15	16,38	23,71	18,88	14,22	19,01	28,2	14,13	17,6	32,73	14,07	23,11
	Mediane	33,87	32,53	46,39	57,33	32,75	11,67	13,02	5,85	4,67	13,20	8,20	7,04
Cl-	1st quartile	26,88	13,62	26,76	37,8	22,64	5,43	0,00	1,09	ND	ND	ND	ND
	3rd quartile	74,49	77,18	126,48	163,54	54,35	33,42	20,61	18,54	18,34	32,43	22,23	22,98
	Mediane	21,17	8,19	39,24	78,94	66,14	32,96	25,65	20,66	12,08	8,3	ND	ND
NO3-	1st quartile	14,74	4,75	20,69	39,64	39,4	17,38	14,63	10,45	4,77	1,09	ND	ND
	3rd quartile	33,42	23,49	61,43	205,23	114,23	60,38	38,79	30,73	28,12	17,83	9,56	ND
	Mediane	21,63	19,35	26,31	35,61	35,27	69,04	205,16	220,34	95,21	39,43	3,26	0,00
SO42-	1st quartile	11,97	3,66	13,82	26,55	28,80	48,86	149,11	154,81	77,93	26,12	0,00	0,00
	3rd quartile	33,19	25,7	38,69	69,19	55,73	133,42	309,74	296,91	156,54	73,9	15,42	1,16
Na+	Mediane	38,36	43,19	74,76	74,48	52,44	35,05	26,43	19,63	12,39	20,29	7,61	2,35
	1st quartile	21,44	23,36	41,36	40,46	29,49	14,79	11,27	7,01	0,00	11,34	ND	ND
	3rd quartile	122,69	96,89	112,66	144,85	82,08	63,27	61,79	51,6	49,15	121,86	58,92	69,94
	Mediane	ND	ND	ND	ND	ND	ND	31,91	47,96	20,51	ND	ND	ND
NH4+	1st quartile	ND	ND	ND	ND	ND	ND	5,10	31,79	8,16	ND	ND	ND
	3rd quartile	ND	ND	ND	ND	ND	7,84	50,70	72,43	25,94	9,64	ND	ND
	Mediane	ND	ND	ND	ND	ND	ND	11,29	10,73	7,90	1,09	0,00	ND
K+	1st quartile	ND	ND	ND	ND	ND	ND	4,51	6,76	0,00	0,00	0,00	ND
	3rd quartile	2,55	3,82	9,75	7,29	7,09	4,41	14,32	19,52	16,53	9,42	4,51	14,1
	Mediane	85,87	124,37	136,85	126,14	105,88	72,6	59,36	66,19	32,78	19,58	1,10	10,52
Ca2+	1st quartile	41,69	44,67	77,78	85,33	42,37	39,60	31,13	17,45	ND	7,09	ND	6,94
	3rd quartile	415,45	235,54	154,46	295,08	228,63	163,12	181,66	129,8	91,91	36,19	96,76	70,99
	Mediane	10,91	6,95	12,35	10,9	11,27	9,31	3,27	6,96	ND	ND	ND	ND
Mg2+	1st quartile	ND	ND	ND	ND	3,47	ND	ND	ND	ND	ND	ND	ND
	3rd quartile	29,6	15,16	19,76	16,45	25,55	18,11	9,47	9,48	6,79	ND	10,21	ND

TABLE A5
Metal concentration data (ng/m3)

Sample ID	Cr52	Mn55	Fe54	Cd112	Ni60	Cu63	Zn66	Pb206	Al27	Sb123	V51
S01St01	1,749	1,728	247,730	0,015	0,856	1,183	1,586	0,404	31,697	0,061	0,363
S01St02	3,530	2,993	329,559	0,040	0,992	2,187	4,599	0,489	84,561	0,128	0,661
S01St03	3,483	4,950	451,684	0,060	2,418	3,788	6,894	0,790	145,618	0,231	1,358
S01St04	0,764	4,223	341,224	0,047	0,899	4,186	4,847	0,641	71,847	0,211	1,210
S01St05	1,466	2,727	268,105	0,064	0,731	3,122	5,189	0,751	84,002	0,207	2,274
S01St06	0,000	1,822	128,830	0,202	0,810	2,239	11,601	1,561	28,164	0,210	1,538
S01St07	0,000	1,505	57,616	0,130	1,003	1,008	4,061	2,113	5,912	0,181	2,040
S01St08	0,000	0,735	31,884	0,060	1,625	0,787	6,624	1,330	0,000	0,162	2,052
S01St09	0,643	1,047	79,451	0,071	1,678	1,714	2,352	0,700	15,544	0,122	1,714
S01St10	0,000	0,170	47,151	0,039	3,620	1,316	0,000	0,360	2,054	0,062	1,790
S01St11	0,000	0,103	21,007	0,020	0,891	0,634	0,881	0,102	0,000	0,000	1,109
S01	***11,634***	***22,002***	***2004,242***	***0,749***	***15,523***	***22,165***	***48,634***	***9,242***	***469,400***	***1,576***	***16,110***
S02St01	1,365	1,365	117,090	1,605	1,605	1,946	2,791	0,015	1,111	1,100	0,659
S02St02	1,554	1,554	161,742	0,578	0,578	1,373	2,620	0,317	0,087	0,100	1,716
S02St03	2,654	2,654	266,340	1,003	1,003	3,634	9,833	0,393	32,618	0,200	0,630
S02St04	3,976	3,976	422,274	1,271	1,271	8,526	5,835	0,998	55,191	0,200	10,517
S02St05	2,091	2,091	236,822	1,552	1,552	4,123	2,988	0,294	32,657	0,000	2,543
S02St06	2,740	2,740	467,791	1,577	1,577	2,425	3,286	0,634	33,268	0,300	2,127
S02St07	0,988	0,988	74,474	1,363	1,363	0,971	8,456	0,851	0,165	0,200	2,088
S02St08	0,383	0,383	39,723	1,072	1,072	0,347	3,123	0,701	0,139	0,200	2,857
S02St09	0,142	0,142	0,984	1,052	1,052	0,228	1,888	0,489	0,115	0,100	2,539
S02St10	0,000	0,009	0,000	0,563	0,563	0,000	0,000	0,230	0,000	0,100	0,006
S02St11	0,000	0,000	0,001	0,575	0,575	0,272	3,283	0,186	0,000	1,930	1,120
S02	***15,894***	***15,903***	***1787,240***	***12,209***	***12,209***	***23,844***	***44,102***	***5,107***	***155,353***	***4,430***	***26,801***
S03St01	1,244	7,927	117,090	0,009	1,230	3,265	4,772	0,294	56,732	0,034	0,000
S03St02	0,547	1,257	73,110	0,000	0,476	1,592	0,591	0,101	31,817	0,033	0,622
S03St03	1,705	3,353	226,128	0,026	1,153	4,404	2,575	0,230	63,480	0,143	2,500
S03St04	1,421	5,006	330,865	0,057	1,292	9,548	2,225	0,296	89,606	0,228	0,933
S03St05	0,859	3,809	282,991	0,032	0,827	5,858	1,860	0,263	45,587	0,119	1,244
S03St06	0,456	1,891	129,437	0,013	0,633	3,240	0,610	0,389	102,504	0,073	0,052
S03St07	0,000	0,661	24,588	0,017	1,164	2,506	0,383	0,749	29,893	0,089	0,725
S03St08	0,000	0,262	8,701	0,017	0,994	0,906	0,937	0,976	2,039	0,114	0,449
S03St09	0,511	0,120	0,000	0,000	1,219	0,281	0,000	0,627	0,000	0,068	0,414
S03St10	0,000	0,071	4,309	0,009	0,744	0,231	0,000	0,660	0,000	0,051	0,208
S32St11	0,169	0,100	7,616	0,013	1,005	0,180	7,031	0,284	0,000	0,000	0,000
S03	***6,912***	***24,455***	***1204,835***	***0,194***	***10,735***	***32,011***	***20,984***	***4,868***	***421,658***	***0,951***	***7,147***

TABLE A5
Metal concentration data (ng/m3)

Sample ID	Cr 52	Mn55	Fe54	Cd112	Ni60	Cu63	Zn66	Pb206	Al27	Sb123	V51
S04St01	0,358	0,388	76,658	0,000	0,283	0,504	1,022	0,000	39,930	0,000	1,116
S04St02	0,866	0,606	80,934	0,109	0,358	0,878	1,663	0,000	15,081	0,000	0,817
S04St03	0,240	1,343	124,128	0,014	0,346	3,210	3,014	0,037	18,408	0,149	1,184
S04St04	1,561	3,617	321,614	0,135	0,985	10,366	4,569	0,243	105,375	0,444	2,245
S04St05	1,935	3,397	297,180	0,075	1,164	9,777	5,391	0,318	101,249	0,373	0,411
S04St06	0,974	2,512	155,308	0,031	0,956	6,065	3,617	0,692	62,076	0,194	2,632
S04St07	0,000	0,621	39,123	0,025	0,558	2,359	2,345	1,054	6,777	0,140	2,034
S04St08	0,686	0,363	59,329	0,044	1,100	6,388	1,902	1,347	0,589	0,176	2,341
S04St09	0,000	0,000	0,000	0,058	0,991	0,269	1,215	0,459	0,000	0,090	0,832
S04St10	0,000	0,000	0,000	0,016	0,497	0,021	0,304	0,111	0,000	0,000	1,075
S04St11	0,387	0,167	66,470	0,000	0,500	1,943	3,038	0,031	5,746	0,000	0,604
S04	***7,006***	***13,014***	***1220,744***	***0,507***	***7,739***	***41,780***	***28,083***	***4,292***	***355,232***	***1,566***	***15,290***
S05St01	0,563	0,544	60,809	0,149	0,601	0,114	0,747	0,000	24,103	0,000	0,867
S05St02	0,643	1,427	122,399	0,008	0,449	1,141	1,435	0,145	29,866	0,060	0,828
S05St03	0,683	2,944	259,605	0,022	1,142	3,714	2,727	0,173	46,144	0,186	0,511
S05St04	2,466	4,325	343,692	0,065	1,688	8,429	4,169	0,373	88,687	0,398	1,694
S05St05	2,021	2,669	198,966	0,054	1,036	6,110	3,801	0,426	72,075	0,254	1,082
S05St06	1,093	1,181	73,353	0,034	0,750	3,540	3,584	0,818	26,521	0,196	0,302
S05St07	0,706	0,368	0,000	0,025	0,707	1,050	1,907	0,902	13,433	0,159	2,461
S05St08	0,194	0,071	0,000	0,032	0,533	1,050	1,382	0,609	0,000	0,100	0,000
S05St09	0,285	0,277	56,584	0,074	1,684	0,325	2,403	0,521	5,652	0,097	0,513
S05St10	1,074	0,000	0,000	0,000	0,844	0,000	1,653	0,091	3,179	0,000	0,040
S05St11	0,078	0,077	39,820	0,043	0,640	6,189	1,876	0,048	8,268	0,000	1,070
S05	***9,805***	***13,882***	***1155,228***	***0,506***	***10,074***	***31,663***	***25,684***	***4,106***	***317,928***	***1,450***	***9,368***
S06St01	1,222	0,720	123,326	0,015	1,186	1,187	4,414	0,039	26,657	0,000	3,741
S06St02	0,577	0,321	78,262	0,000	0,401	0,584	1,625	0,000	13,698	0,000	1,203
S06St03	0,300	0,446	56,170	0,000	0,191	1,508	0,000	0,000	8,818	0,063	0,000
S06St04	1,562	2,265	259,425	0,048	1,093	7,984	2,877	0,106	27,414	0,291	2,061
S06St05	0,471	0,586	73,653	0,000	0,311	2,931	0,618	0,000	13,769	0,103	0,577
S06St06	0,000	0,000	0,000	0,000	0,000	0,918	0,000	0,019	1,436	0,000	0,000
S06St07	0,133	0,093	6,146	0,000	0,193	0,755	0,432	0,336	4,057	0,075	0,278
S06St08	1,336	0,894	99,392	0,087	1,753	1,790	5,901	1,572	17,295	0,460	4,160
S06St09	0,142	0,000	0,000	0,000	0,226	0,055	0,745	0,187	1,857	0,053	1,054
S06St10	0,135	0,000	0,000	0,000	0,221	0,043	0,532	0,000	0,773	3,209	0,000
S06St11	0,315	0,000	19,179	0,000	0,409	0,397	0,532	0,031	2,542	0,000	0,973
S06	***6,192***	***5,324***	***715,552***	***0,150***	***5,985***	***18,152***	***17,674***	***2,290***	***118,316***	***4,253***	***14,046***

TABLE A5

Metal concentration data (ng/m3)

Sample ID	Cr 52	Mn55	Fe54	Cd112	Ni60	Cu63	Zn66	Pb206	Al27	Sb123	V51
S07St01	1,965	0,000	68,234	0,000	0,906	0,670	1,185	0,167	43,261	0,053	2,465
S07St02	1,730	1,487	77,337	0,005	2,267	1,219	1,348	0,083	30,698	0,000	4,349
S07St03	1,314	2,371	118,108	0,045	0,859	4,033	0,817	0,187	19,300	0,135	2,616
S07St04	2,244	3,836	277,785	0,091	1,800	10,201	3,161	0,316	54,185	0,347	3,888
S07St05	1,664	1,977	131,530	0,107	0,815	8,139	2,455	0,321	17,695	0,209	1,788
S07St06	1,976	1,464	80,624	0,034	0,972	5,280	2,929	0,736	11,517	0,149	3,687
S07St07	0,758	1,177	26,188	0,049	0,839	3,118	4,791	1,334	8,686	0,192	0,000
S07St08	0,740	0,992	30,359	0,063	1,741	1,388	6,250	1,684	18,278	0,287	2,050
S07St09	0,224	0,214	2,267	0,026	1,164	0,244	3,441	0,607	15,055	0,101	1,636
S07St10	0,000	0,000	0,000	0,000	0,902	0,000	0,000	0,181	2,969	0,000	0,236
S07St11	0,127	0,000	0,000	0,059	0,444	1,639	2,003	0,180	21,939	0,000	0,000
S07	***12,743***	***13,518***	***812,433***	***0,479***	***12,709***	***35,931***	***28,380***	***5,793***	***243,582***	***1,474***	***22,716***
S08St01	0,165	0,092	35,445	0,000	0,262	0,298	0,988	0,038	5,197	0,000	0,147
S08St02	0,170	0,000	55,653	0,000	0,231	0,397	1,180	0,061	9,087	0,000	0,148
S08St03	0,147	0,279	74,706	0,000	0,291	1,096	1,033	0,056	7,197	0,051	0,227
S08St04	0,330	1,258	145,851	0,029	0,530	3,834	2,140	0,131	8,937	0,173	0,267
S08St05	0,225	0,773	97,597	0,025	0,389	3,276	1,634	0,166	6,010	0,127	0,144
S08St06	0,221	0,405	62,765	0,014	0,363	2,324	2,284	0,281	13,676	0,071	0,258
S08St07	0,213	0,428	58,841	0,020	0,616	1,499	3,239	0,663	9,899	0,068	0,404
S08St08	0,095	0,011	11,122	0,022	0,639	0,643	2,272	0,679	4,930	0,078	0,431
S08St09	0,163	0,000	43,819	0,905	0,633	0,438	1,950	0,287	7,149	0,000	0,243
S08St10	0,162	0,000	40,526	0,012	0,449	0,063	2,046	0,146	5,398	0,000	0,092
S08St11	0,092	0,000	19,013	0,000	0,263	0,000	0,379	0,108	2,363	0,000	0,000
S08	***1,981***	***3,248***	***645,339***	***1,028***	***4,665***	***13,868***	***19,146***	***2,616***	***79,844***	***0,569***	***2,360***
S09St01	0,464	0,776	104,472	0,063	2,367	0,941	1,031	0,202	10,308	0,000	0,405
S09St02	0,758	1,229	165,953	0,566	0,550	1,046	1,493	0,271	36,832	0,000	0,670
S09St03	0,022	1,331	114,420	7,258	0,438	1,249	1,647	0,253	18,767	0,000	0,865
S09St04	0,369	2,251	206,237	0,300	1,588	3,271	3,167	0,408	36,914	0,125	1,496
S09St05	0,487	1,579	180,942	0,456	0,994	2,067	1,772	0,360	38,428	0,062	0,735
S09St06	0,000	0,898	94,668	0,007	0,383	0,883	2,091	0,656	15,632	0,000	0,000
S09St07	1,272	2,398	252,329	0,027	1,437	2,406	2,466	2,986	31,333	0,226	0,551
S09St08	1,108	1,256	188,995	0,000	1,551	1,548	1,962	3,059	25,176	0,243	0,363
S09St09	1,226	0,899	220,289	0,375	2,025	1,825	1,367	1,641	37,569	0,118	0,696
S09St10	0,943	0,609	216,918	0,000	1,591	0,523	0,135	0,493	24,661	0,000	0,781
S09St11	0,427	0,382	151,425	0,035	0,835	0,232	0,000	0,049	22,702	0,000	0,000
S09st12	1,216	0,726	237,943	0,103	0,981	0,841	0,000	0,431	40,742	0,000	0,000

TABLE A5

Metal concentration data (ng/m3)

Sample ID	Cr 52	Mn55	Fe54	Cd112	Ni60	Cu63	Zn66	Pb206	Al27	Sb123	V51
S10St01	0,000	0,749	27,633	0,021	0,338	0,377	3,234	0,079	6,609	0,000	1,078
S10St02	0,333	1,672	107,481	0,039	0,363	0,751	0,000	0,210	32,080	0,000	1,151
S10St03	0,920	3,570	242,182	0,053	0,845	2,556	1,297	0,692	87,996	0,108	1,348
S10St04	1,129	3,983	255,064	0,080	0,717	3,501	2,457	0,496	88,486	0,172	0,000
S10St05	0,570	1,845	99,103	0,000	0,525	1,913	0,000	0,331	35,294	0,134	0,000
S10St06	0,461	0,994	55,179	0,092	0,665	1,089	0,445	0,463	17,460	0,063	2,520
S10St07	1,207	1,112	119,129	0,017	0,725	1,301	0,983	1,029	36,967	0,073	2,074
S10St08	0,904	0,728	82,637	0,037	0,849	0,763	2,014	1,499	19,407	0,104	2,198
S10St09	0,214	0,164	21,140	0,056	1,030	0,752	0,525	0,598	3,622	0,062	3,097
S10St10	0,445	0,167	47,518	0,031	0,554	0,330	0,000	0,244	11,458	0,000	2,409
S10St11	1,182	0,467	121,328	0,016	0,491	0,960	0,930	1,020	35,846	0,000	2,422
S10St12	0,285	0,000	16,299	0,663	0,319	0,602	0,000	0,000	0,000	0,000	2,309
S10	***7,648***	***15,450***	***1194,692***	***1,105***	***7,421***	***14,895***	***11,884***	***6,661***	***375,226***	***0,716***	***20,606***
S11St01	0,464	0,776	104,472	0,063	2,367	0,941	2,952	0,202	10,308	0,000	2,660
S11St02	0,758	1,229	165,953	0,566	0,550	1,046	1,893	0,271	36,832	0,000	4,579
S11St03	0,022	1,331	114,420	7,258	0,438	1,249	1,256	0,253	18,767	0,000	0,741
S11St04	0,369	2,251	206,237	0,300	1,588	3,271	2,377	0,408	36,914	0,125	2,000
S11St05	0,487	1,579	180,942	0,456	0,994	2,067	1,485	0,360	38,428	0,062	2,011
S11St06	0,000	0,898	94,668	0,007	0,383	0,883	1,399	0,656	15,632	0,000	1,648
S11St07	1,272	2,398	252,329	0,027	1,437	2,406	4,959	2,986	31,333	0,226	3,391
S11St08	1,108	1,256	188,995	0,000	1,551	1,548	3,435	3,059	25,176	0,243	2,351
S11St09	1,226	0,899	220,289	0,375	2,025	1,825	3,421	1,641	37,569	0,118	3,804
S11St10	0,943	0,609	216,918	0,000	1,591	0,523	1,296	0,493	24,661	0,000	0,019
S11St11	0,427	0,382	151,425	0,035	0,835	0,232	1,042	0,049	22,702	0,000	1,860
S11St12	1,216	0,726	237,943	0,103	0,981	0,841	0,000	0,431	40,742	0,000	2,719
S11	***8,292***	***14,336***	***2134,591***	***9,191***	***14,741***	***16,831***	***25,516***	***10,809***	***339,062***	***0,774***	***27,784***
S12St01	0,000	0,749	27,633	0,021	0,338	0,377	3,234	0,079	6,609	0,000	0,000
S12St02	0,333	1,672	107,481	0,039	0,363	0,751	0,000	0,210	32,080	0,000	0,816
S12St03	0,920	3,570	242,182	0,053	0,845	2,556	1,297	0,692	87,996	0,108	2,037
S12St04	1,129	3,983	255,064	0,080	0,717	3,501	2,457	0,496	88,486	0,172	3,578
S12St05	0,570	1,845	99,103	0,000	0,525	1,913	0,000	0,331	35,294	0,134	2,473
S12St06	0,461	0,994	55,179	0,092	0,665	1,089	0,445	0,463	17,460	0,063	3,656
S12St07	1,207	1,112	119,129	0,017	0,725	1,301	0,983	1,029	36,967	0,073	4,736
S12St08	0,904	0,728	82,637	0,037	0,849	0,763	2,014	1,499	19,407	0,104	6,607
S12St09	0,214	0,164	21,140	0,056	1,030	0,752	0,525	0,598	3,622	0,062	3,702
S12St10	0,445	0,167	47,518	0,031	0,554	0,330	0,000	0,244	11,458	0,000	4,092
S12St11	1,182	0,467	121,328	0,016	0,491	0,960	0,930	1,020	35,846	0,000	5,445
S12St12	0,285	0,000	16,299	0,663	0,319	0,602	0,000	0,000	0,000	0,000	4,071
S12	***7,648***	***15,450***	***1194,692***	***1,105***	***7,421***	***14,895***	***11,884***	***6,661***	***375,226***	***0,716***	***41,213***

TABLE A5

Metal concentration data (ng/m3)

Sample ID	Cr52	Mn55	Fe54	Cd112	Ni60	Cu63	Zn66	Pb206	Al27	Sb123	V51
S13St01	0,687	1,889	211,410	0,047	4,503	0,841	2,511	0,943	103,798	0,000	0,847
S13St02	0,894	2,492	263,116	0,013	1,406	1,721	1,300	0,421	148,902	0,000	1,515
S13St03	1,021	4,057	358,474	0,023	0,992	4,948	1,537	0,353	261,738	0,102	1,595
S13St04	1,827	6,959	575,863	0,075	1,221	8,588	2,323	0,511	514,264	0,201	4,614
S13St05	1,824	5,867	471,677	0,242	0,902	6,995	3,217	0,551	517,802	0,141	4,824
S13St06	0,455	1,881	142,814	0,009	0,388	2,719	0,597	0,379	160,505	0,059	4,180
S13St07	0,331	0,825	66,029	0,033	1,126	2,188	1,236	0,772	48,748	0,097	4,018
S13St08	0,341	0,301	58,263	0,060	1,867	1,228	0,367	0,960	0,000	0,118	1,393
S13St09	0,000	0,004	0,000	0,019	0,922	0,346	0,599	0,438	5,994	0,051	0,762
S13St10	0,000	0,025	18,076	0,000	1,520	0,192	0,347	0,113	0,000	0,000	0,856
S13St11	0,071	0,000	19,487	0,004	0,433	0,275	0,393	0,000	2,809	0,000	0,607
S13St12	0,025	0,000	31,807	0,719	0,174	0,114	0,000	0,000	4,146	0,000	0,090
S13	***7,474***	***24,300***	***2217,017***	***1,243***	***15,455***	***30,154***	***14,428***	***5,440***	***1768,705***	***0,770***	***25,301***
S14St01	0,000	0,338	31,718	0,029	0,300	0,254	0,395	0,028	4,934	0,000	0,000
S14St02	0,329	0,747	55,134	0,007	4,541	1,040	10,024	0,151	13,369	0,000	0,886
S14St03	0,941	1,600	148,898	0,022	1,073	2,518	1,924	0,297	41,922	0,065	1,756
S14St04	1,456	2,595	242,015	0,047	1,860	5,671	2,487	0,423	45,942	0,177	1,735
S14St05	1,854	1,663	166,423	0,037	1,538	4,992	2,403	0,305	64,114	0,158	2,582
S14St06	0,587	0,613	44,136	0,535	0,738	2,415	1,579	0,235	30,570	0,092	2,040
S14St07	0,000	0,372	0,455	0,016	1,144	1,345	1,759	0,325	0,000	0,180	0,727
S14St08	0,419	0,257	6,514	0,049	0,808	1,270	1,426	0,581	7,754	0,256	1,386
S14St09	0,221	0,106	6,825	0,061	0,767	0,642	1,564	0,326	8,713	0,114	1,328
S14St10	0,377	0,071	9,000	0,023	0,485	0,159	0,439	0,147	10,919	0,000	0,702
S14St11	0,162	0,000	0,000	0,000	0,000	0,135	0,670	0,000	2,472	0,000	1,519
S14St12	0,739	0,138	21,095	0,199	0,156	0,412	1,696	0,069	18,837	0,000	1,940
S14	***7,084***	***8,499***	***732,211***	***1,026***	***13,409***	***20,853***	***26,368***	***2,888***	***249,546***	***1,042***	***16,603***
S15St01	0,799	0,439	53,935	0,092	0,299	0,304	1,463	0,336	13,359	0,000	0,000
S15St02	1,137	0,566	78,958	0,041	1,644	0,562	0,664	0,142	24,785	0,000	0,065
S15St03	0,434	1,151	97,376	0,008	2,644	1,922	2,864	0,246	18,153	0,090	0,000
S15St04	1,094	1,927	200,113	0,056	3,350	5,265	4,072	0,394	27,907	0,173	0,190
S15St05	0,937	1,151	128,795	0,052	3,182	4,946	8,381	0,278	23,426	0,104	0,830
515St06	0,526	0,548	40,137	0,136	0,514	3,139	1,545	0,161	28,570	0,074	0,970
S15St07	0,162	0,430	13,803	0,000	1,027	3,008	4,753	0,323	10,896	0,080	0,000
S15St08	0,296	0,288	24,371	0,000	2,470	1,120	1,171	0,419	11,861	0,085	0,354
S15St09	0,715	0,254	45,819	0,400	5,830	0,422	1,477	0,278	25,806	0,167	1,420
S15St10	0,170	0,074	26,590	0,000	0,814	0,260	4,682	0,112	4,723	0,056	0,000
S15St11	0,000	0,000	0,000	0,020	1,198	0,043	0,000	0,000	0,000	0,000	0,000
S15St12	0,192	0,000	0,000	0,000	1,509	0,273	1,469	0,032	9,989	0,000	0,000
S15	***6,462***	***6,828***	***709,896***	***0,805***	***24,481***	***21,263***	***32,542***	***2,721***	***199,475***	***0,829***	***3,829***

Table A5
Metal concentration data (ng/m3)

Sample ID	Cr 52	Mn55	Fe54	Cd112	Ni60	Cu63	Zn66	Pb206	Al27	Sb123	V51
S16St01	0,333	0,424	36,867	0,000	1,955	0,460	1,553	0,078	8,563	0,000	1,878
S16St02	0,893	0,978	81,764	0,019	0,436	1,247	2,115	0,224	33,383	0,000	2,734
S16St03	0,973	1,323	99,999	0,021	0,588	1,951	1,253	0,283	34,583	0,000	2,840
S16St04	1,078	1,659	136,740	0,036	0,731	3,883	1,467	0,353	39,114	0,106	3,271
S16St05	0,838	1,162	97,578	0,016	0,685	3,648	1,088	0,266	24,625	0,112	3,030
516St06	0,715	0,574	54,048	0,005	0,535	2,381	1,287	0,278	28,170	0,053	3,812
S16St07	0,566	0,479	47,484	0,375	0,746	1,964	0,426	0,595	17,713	0,091	0,672
S16St08	0,542	0,336	34,550	0,026	0,864	1,225	3,120	0,908	16,749	0,110	1,929
S16St09	0,231	0,088	9,647	0,014	0,632	0,173	0,266	0,316	11,207	0,000	1,680
S16St10	0,000	0,000	6,613	0,003	0,490	0,031	0,000	0,110	0,000	0,000	1,139
S16St11	0,020	0,000	0,000	0,064	0,000	0,000	0,000	0,000	0,882	0,000	1,308
S65St12	0,467	0,084	31,145	0,000	0,073	0,166	0,566	0,022	19,215	0,000	3,536
S16	***6,656***	***7,107***	***636,434***	***0,577***	***7,734***	***17,131***	***13,142***	***3,432***	***234,204***	***0,471***	***27,829***

TABLE A6

Detection limits for metals (ng/m3)

metal	MDL	MQL
Cr	0,0156	0,048
Mn	0,06	0,18
Fe	0,36	1,2
Cd	0,00072	0,0024
Ni	0,0312	0,096
Cu	0,012	0,036
Zn	0,072	0,24
Pb	0,012	0,036
Al	0,72	2,4
Sb	0,048	0,144
V	0,036	0,12

TABLE A7

Distribution parameters (median, 1st and 3rd quartiles) of the concentration of metals in various dimensional classes (ng/m3)

Parameter		St 01 >18 µm	St 02 18-10 µm	St 03 10-5,6 µm	St 04 5,6-3,2 µm	St 05 3,2-1,8 µm	St 06 1,8-1,0 µm	St 07 1,0-0,56 µm	St 08 0,56-0,32µm	St 09 0,32-0,18 µm	St10 0,18-0,10 µm	St11 0,10-0,056µm	St12 <0,056
Cr	Mediane	0,51	0,76	0,92	1,28	0,90	0,46	0,46	0,48	0,22	0,17	0,14	0,38
	1st quartile	0,25	0,44	0,27	0,92	0,53	0,28	0,15	0,25	0,15	0,00	0,05	0,24
	3rd quartile	1,23	1,02	1,17	1,69	1,84	0,84	1,10	0,90	0,58	0,44	0,41	0,98
Mn	Mediane	0,73	1,24	1,99	3,73	1,84	0,99	0,74	0,55	0,16	0,06	0,07	0,06
	1st quartile	0,41	0,68	1,33	2,25	1,37	0,59	0,43	0,27	0,04	ND	ND	ND
	3rd quartile	1,25	1,67	3,57	4,27	3,06	1,85	1,34	0,94	0,59	0,17	0,27	0,43
Fe	Mediane	64,52	81,35	121,12	255,06	148,98	68,06	43,30	33,22	15,39	13,54	19,33	26,12
	1st quartile	33,58	75,22	98,69	203,18	98,35	49,09	9,97	9,91	0,33	0,00	0,00	16,3
	3rd quartile	110,78	144,18	242,18	326,24	233,54	111,75	92,58	82,64	51,20	47,33	93,90	134,88
Cd	Mediane	0,02	0,03	0,02	0,07	0,05	0,03	0,02	0,04	0,06	0,01	0,02	0,15
	1st quartile	0,00	0,01	0,02	0,05	0,02	0,01	0,02	0,02	0,02	0,00	0,00	0,05
	3rd quartile	0,06	0,08	0,06	0,11	0,17	0,11	0,04	0,06	0,38	0,03	0,04	0,66
Ni	Mediane	0,98	0,51	0,85	1,44	0,95	0,65	0,91	1,05	1,03	0,65	0,49	0,32
	1st quartile	0,32	0,38	0,44	0,86	0,61	0,39	0,72	0,83	0,70	0,49	0,34	0,17
	3rd quartile	1,95	1,53	1,15	1,77	1,35	0,85	1,15	1,75	1,85	1,21	0,84	0,98
Cu	Mediane	0,59	1,05	2,56	5,47	3,46	2,40	2,08	1,23	0,43	0,18	0,34	0,51
	1st quartile	0,34	0,75	1,72	3,67	2,50	1,09	1,30	0,85	0,28	0,03	0,16	0,22
	3rd quartile	1,06	1,42	3,91	8,21	5,55	3,26	2,46	1,55	1,23	0,33	1,30	0,72
Zn	Mediane	1,57	1,39	1,30	2,46	1,56	1,40	1,83	2,01	1,35	0,33	0,91	1,02
	1st quartile	1,01	0,63	1,14	2,35	0,68	0,60	0,92	1,28	0,52	0,00	0,39	ND
	3rd quartile	3,09	1,89	2,65	3,62	3,51	2,61	4,41	3,44	2,38	1,30	1,46	136,12
Pb	Mediane	0,12	0,18	0,25	0,40	0,33	0,46	0,84	1,19	0,56	0,15	0,05	0,03
	1st quartile	0,04	0,09	0,18	0,31	0,27	0,28	0,63	0,79	0,32	0,11	0,02	ND
	3rd quartile	0,31	0,27	0,52	0,50	0,39	0,71	1,72	1,63	0,66	0,30	0,23	0,25
Al	Mediane	11,83	31,26	26,94	50,06	36,86	21,99	12,16	9,81	6,57	3,07	2,68	14,41
	1st quartile	6,61	14,39	18,28	35,78	20,56	14,65	6,34	0,29	2,74	0,00	0,44	2,07
	3rd quartile	35,81	35,11	88,00	88,59	78,04	29,57	31,33	18,84	15,3	11,19	22,32	29,98
Sb	Mediane	ND	0,01	0,11	0,19	0,13	0,07	0,12	0,14	0,09	0,03	ND	ND
	1st quartile	ND	ND	0,06	0,17	0,11	0,06	0,08	0,10	0,06	0,01	ND	ND
	3rd quartile	0,04	0,04	0,15	0,32	0,21	0,17	0,19	0,25	0,12	0,04	ND	ND
V	Mediane	0,75	0,86	1,27	1,87	1,52	1,84	1,38	1,99	1,37	0,47	1,02	2,12
	1st quartile	0,07	1,62	0,57	1,07	0,66	0,28	0,48	0,44	0,73	0,03	ND	0,05
	3rd quartile	1,50	4,58	1,90	3,42	2,51	3,14	2,27	2,35	2,13	1,07	1,41	3,13

TABLE A8
Concentration of carbonaceous species: EC, EC and TC (μg/m³)

sample ID	OC	EC	TC	ΔOC	ΔEC	ΔTC	OptEC
S01St01	0,05	0,00	0,05	0,01	0,00	0,01	0,00
S01St02	0,05	0,01	0,06	0,01	0,00	0,01	0,00
S01St03	0,23	0,05	0,28	0,02	0,00	0,02	0,00
S01St04	0,32	0,04	0,36	0,02	0,00	0,03	0,00
S01St05	0,21	0,05	0,26	0,02	0,00	0,02	0,00
S01St06	0,42	0,07	0,49	0,03	0,01	0,03	0,02
S01St07	0,70	0,10	0,80	0,04	0,01	0,05	0,01
S01St08	0,73	0,09	0,82	0,05	0,01	0,05	0,01
S01St09	0,39	0,06	0,45	0,03	0,01	0,03	0,01
S01St10	0,26	0,06	0,32	0,02	0,01	0,03	0,01
S01St11	0,17	0,03	0,21	0,02	0,00	0,02	0,00
S01	***3,54***	***0,56***	***4,10***				
S02St01	0,03	0,00	0,03	0,01	0,00	0,01	0,00
S02St02	0,13	0,01	0,14	0,01	0,00	0,01	0,00
S02St03	0,16	0,01	0,17	0,01	0,00	0,02	0,00
s02St04	0,27	0,02	0,29	0,02	0,00	0,02	0,01
S02St05	0,14	0,02	0,16	0,01	0,00	0,02	0,00
S21St06	0,19	0,03	0,22	0,02	0,00	0,02	0,01
S02St07	0,25	0,06	0,31	0,02	0,00	0,03	0,02
S02St08	0,33	0,07	0,40	0,03	0,01	0,03	0,01
S02St09	0,27	0,06	0,32	0,02	0,00	0,03	0,01
S02St10	0,15	0,06	0,20	0,02	0,00	0,02	0,01
S02St11	0,08	0,03	0,11	0,01	0,00	0,02	0,00
S02	***1,99***	***0,36***	***2,35***				
S03St01	0,09	0,00	0,09	0,01	0,00	0,01	0,00
S03St02	0,08	0,00	0,09	0,01	0,00	0,01	0,00
S03St03	0,34	0,00	0,34	0,02	0,00	0,02	0,00
S03St04	0,51	0,00	0,51	0,03	0,00	0,03	0,00
S03St05	0,24	0,02	0,27	0,02	0,00	0,02	0,01
S03St06	0,18	0,06	0,24	0,02	0,00	0,02	0,01
S03St07	0,28	0,05	0,33	0,02	0,00	0,03	0,01
S03St08	0,35	0,05	0,40	0,03	0,00	0,03	0,01
S03St09	0,27	0,05	0,31	0,02	0,00	0,03	0,01
S03St10	0,28	0,08	0,36	0,02	0,01	0,03	0,01
S03St11	0,06	0,05	0,11	0,01	0,00	0,02	0,00
S03	***2,69***	***0,37***	***3,06***				
S04St01	0,09	0,02	0,11	0,01	0,00	0,01	0,00
S04St02	0,06	0,02	0,07	0,01	0,00	0,01	0,00
S04St03	0,44	0,06	0,50	0,03	0,00	0,03	0,00
S04St04	0,36	0,03	0,39	0,02	0,00	0,03	0,00
S04St05	0,25	0,04	0,29	0,02	0,00	0,02	0,01
S04St06	0,27	0,05	0,33	0,02	0,00	0,02	0,01
S04St07	0,38	0,05	0,43	0,03	0,00	0,03	0,02
S04St08	0,54	0,04	0,58	0,03	0,00	0,04	0,02
S04St09	0,38	0,05	0,43	0,03	0,00	0,03	0,01
S04St10	0,31	0,06	0,37	0,02	0,00	0,03	0,01
S04St11	0,22	0,05	0,27	0,02	0,00	0,02	0,01
S04	***3,29***	***0,47***	***3,76***				

TABLE A8

Concentration of carbonaceous species: EC, EC and TC (μg/m^3)

sample ID	OC	EC	TC	ΔOC	ΔEC	ΔTC	OptEC
S05St01	0,09	0,02	0,11	0,01	0,00	0,01	0,00
S05St02	0,04	0,02	0,06	0,01	0,00	0,01	0,00
S05St03	0,17	0,06	0,22	0,01	0,00	0,02	0,00
S05St04	0,15	0,03	0,19	0,01	0,00	0,02	0,00
S05St05	0,13	0,03	0,15	0,01	0,00	0,01	0,00
S05St06	0,17	0,03	0,21	0,01	0,00	0,02	0,01
S05St07	0,38	0,05	0,43	0,03	0,00	0,03	0,02
S05St08	0,46	0,07	0,52	0,03	0,01	0,04	0,02
S05St09	0,29	0,06	0,35	0,02	0,00	0,03	0,01
S05St10	0,09	0,03	0,12	0,01	0,00	0,02	0,01
S05St11	0,08	0,03	0,12	0,01	0,00	0,02	0,00
S05	***2,05***	***0,42***	***2,48***				
S06St01	0,12	0,03	0,15	0,01	0,00	0,01	0,00
S06St02	0,04	0,01	0,05	0,01	0,00	0,01	0,00
S06St03	0,18	0,03	0,21	0,01	0,00	0,02	0,00
S06St04	0,13	0,03	0,16	0,01	0,00	0,01	0,00
S06St05	0,13	0,02	0,15	0,01	0,00	0,01	0,00
S06St06	0,27	0,05	0,32	0,02	0,00	0,02	0,01
S06St07	0,36	0,03	0,38	0,03	0,00	0,03	0,01
S06St08	0,67	0,04	0,71	0,04	0,00	0,04	0,02
S06St09	0,43	0,03	0,47	0,03	0,00	0,03	0,01
S06St10	0,27	0,04	0,31	0,02	0,00	0,02	0,01
S06St11	0,03	0,04	0,07	0,01	0,00	0,01	0,00
S06	***2,64***	***0,35***	***2,99***				
S07St01	0,07	0,02	0,09	0,01	0,00	0,01	0,00
S07St02	0,07	0,01	0,09	0,01	0,00	0,01	0,00
S07St03	0,08	0,02	0,10	0,01	0,00	0,01	0,00
S07St04	0,26	0,06	0,32	0,02	0,00	0,02	0,00
S07St05	0,10	0,02	0,13	0,01	0,00	0,01	0,00
S07St06	0,20	0,03	0,23	0,02	0,00	0,02	0,01
S07St07	0,68	0,05	0,73	0,04	0,00	0,05	0,03
S07St08	0,94	0,03	0,97	0,06	0,00	0,06	0,02
S07St09	0,43	0,03	0,46	0,03	0,00	0,03	0,01
S07St10	0,27	0,04	0,30	0,02	0,00	0,03	0,01
S07St11	0,13	0,04	0,17	0,02	0,00	0,02	0,01
S07	***3,23***	***0,34***	***3,58***				
S08St01	0,07	0,02	0,09	0,01	0,00	0,01	0,00
S08St02	0,02	0,01	0,03	0,01	0,00	0,01	0,00
S08St03	0,07	0,02	0,08	0,01	0,00	0,01	0,00
S08St04	0,10	0,03	0,13	0,01	0,00	0,01	0,00
S08St05	0,10	0,03	0,14	0,01	0,00	0,01	0,00
S08St06	0,27	0,04	0,31	0,02	0,00	0,02	0,01
S08St07	0,67	0,04	0,67	0,04	0,00	0,04	0,02
S08St08	0,89	0,03	0,92	0,05	0,00	0,06	0,02
S87St09	0,31	0,02	0,33	0,02	0,00	0,03	0,01
S08St10	0,14	0,03	0,17	0,01	0,00	0,02	0,01
S08St11	0,06	0,03	0,08	0,01	0,00	0,01	0,01
S08	***2,70***	***0,28***	***2,95***				

TABLE A8
Concentration of carbonaceous species: EC, EC and TC (μg/m³)

sample ID	OC	EC	TC	ΔOC	ΔEC	ΔTC	OptEC
S09St01	0,05	0,00	0,05	0,01	0,00	0,01	0,00
S09St02	0,06	0,03	0,09	0,01	0,00	0,01	0,00
S09St03	0,08	0,03	0,11	0,01	0,00	0,01	0,00
s09St04	0,11	0,03	0,14	0,01	0,00	0,01	0,00
S09St05	0,09	0,03	0,12	0,01	0,00	0,01	0,00
S09St06	0,19	0,03	0,22	0,01	0,00	0,02	0,01
S09St07	0,38	0,03	0,41	0,02	0,00	0,03	0,01
S09St08	0,58	0,05	0,62	0,03	0,00	0,04	0,01
S09St09	0,38	0,08	0,46	0,02	0,01	0,03	0,01
S09St10	0,20	0,07	0,26	0,01	0,01	0,02	0,01
S09St11	0,09	0,04	0,13	0,01	0,00	0,01	0,00
S09St12	0,72	0,04	0,76	0,04	0,00	0,04	0,00
S09	***2,92***	***0,44***	***3,36***				
S10St01	0,07	0,01	0,08	0,01	0,00	0,01	0,00
S10St02	0,09	0,01	0,10	0,01	0,00	0,01	0,00
S10St03	0,13	0,03	0,16	0,01	0,00	0,01	0,00
S10St04	0,17	0,02	0,20	0,01	0,00	0,01	0,00
S10St05	0,14	0,03	0,17	0,01	0,00	0,01	0,01
S10St06	0,27	0,04	0,31	0,02	0,00	0,02	0,01
S10St07	0,56	0,04	0,59	0,03	0,00	0,04	0,02
S10St08	0,68	0,03	0,72	0,04	0,00	0,04	0,02
S10St09	0,44	0,04	0,48	0,03	0,00	0,03	0,01
S10St10	0,23	0,06	0,28	0,02	0,00	0,02	0,01
S10St11	0,08	0,03	0,11	0,01	0,00	0,01	0,00
S10St12	0,23	0,04	0,27	0,02	0,00	0,02	0,01
S10	***3,08***	***0,39***	***3,47***				
S11St01	0,06	0,01	0,07	0,01	0,00	0,01	0,00
S11St02	0,08	0,00	0,08	0,01	0,00	0,01	0,00
S11St03	0,09	0,02	0,10	0,01	0,00	0,01	0,00
S11St04	0,11	0,03	0,15	0,01	0,00	0,01	0,00
S11St05	0,11	0,02	0,12	0,01	0,00	0,01	0,00
S11St06	0,28	0,03	0,31	0,02	0,00	0,02	0,01
S11St07	0,59	0,04	0,63	0,03	0,00	0,04	0,02
S11St08	0,59	0,03	0,63	0,03	0,00	0,04	0,02
S11St09	0,31	0,03	0,35	0,02	0,00	0,02	0,01
S11St10	0,16	0,05	0,20	0,01	0,00	0,02	0,01
S11St11	0,05	0,03	0,08	0,01	0,00	0,01	0,00
S11St12	0,09	0,03	0,12	0,01	0,00	0,01	0,00
S11	***2,52***	***0,33***	***2,85***				
S12St01	0,03	0,00	0,03	0,00	0,00	0,01	0,00
S12St02	0,15	0,01	0,15	0,01	0,00	0,01	0,00
S12St03	0,16	0,03	0,19	0,01	0,00	0,01	0,00
S12St04	0,18	0,03	0,21	0,01	0,00	0,01	0,00
S12St05	0,15	0,02	0,17	0,01	0,00	0,01	0,00
S12St06	0,19	0,03	0,22	0,01	0,00	0,02	0,01
S12St07	0,27	0,03	0,30	0,02	0,00	0,02	0,01
S12St08	0,58	0,05	0,63	0,03	0,00	0,04	0,01
S12St09	0,35	0,05	0,40	0,02	0,00	0,03	0,01
S12St10	0,18	0,06	0,25	0,01	0,00	0,02	0,01
S12St11	0,06	0,03	0,09	0,01	0,00	0,01	0,00
S12St12	0,11	0,04	0,14	0,01	0,00	0,01	0,00

TABLE A8
Concentration of carbonaceous species: EC, EC and TC (μg/m³)

sample ID	OC	EC	TC	ΔOC	ΔEC	ΔTC	OptEC
S13St01	0,06	0,00	0,06	0,01	0,00	0,01	0,00
S13St02	0,08	0,00	0,08	0,01	0,00	0,01	0,00
S13St03	0,18	0,01	0,19	0,01	0,00	0,01	0,00
S13St04	0,24	0,01	0,24	0,01	0,00	0,02	0,00
S13St05	0,15	0,04	0,19	0,01	0,00	0,01	0,01
S13St06	0,12	0,03	0,14	0,01	0,00	0,01	0,01
S13St07	0,31	0,06	0,37	0,02	0,00	0,02	0,02
S13St08	0,34	0,03	0,37	0,02	0,00	0,02	0,02
S13St09	0,21	0,03	0,24	0,01	0,00	0,02	0,01
S13St10	0,12	0,05	0,18	0,01	0,00	0,02	0,00
S13St11	0,05	0,02	0,08	0,01	0,00	0,01	0,00
S13St12	0,08	0,03	0,11	0,01	0,00	0,01	0,00
S13	***1,93***	***0,32***	***2,25***				
S14St01	0,02	0,00	0,02	0,00	0,00	0,01	0,00
S14St02	0,03	0,00	0,03	0,00	0,00	0,01	0,00
S14St03	0,07	0,01	0,09	0,01	0,00	0,01	0,00
S14St04	0,16	0,03	0,19	0,01	0,00	0,01	0,00
S14St05	0,09	0,02	0,11	0,01	0,00	0,01	0,00
S14St06	0,13	0,03	0,17	0,01	0,00	0,01	0,01
S14St07	0,21	0,03	0,24	0,01	0,00	0,02	0,01
S14St08	0,32	0,02	0,34	0,02	0,00	0,02	0,01
S14St09	0,21	0,02	0,23	0,01	0,00	0,02	0,01
S14St10	0,12	0,04	0,15	0,01	0,00	0,01	0,01
S14St11	0,04	0,02	0,06	0,01	0,00	0,01	0,00
S14St12	0,06	0,02	0,08	0,01	0,00	0,01	0,00
S14	***1,48***	***0,24***	***1,71***				
S15St01	0,02	0,00	0,02	0,01	0,00	0,01	0,00
S15St02	0,04	0,01	0,05	0,01	0,00	0,01	0,00
S15St03	0,09	0,02	0,11	0,01	0,00	0,01	0,00
S15St04	0,15	0,02	0,18	0,01	0,00	0,01	0,00
S15St05	0,12	0,02	0,14	0,01	0,00	0,01	0,00
S15St06	0,15	0,04	0,19	0,01	0,00	0,02	0,01
S15St07	0,26	0,04	0,30	0,02	0,00	0,02	0,01
S15St08	0,42	0,06	0,48	0,03	0,01	0,03	0,02
S15St09	0,26	0,06	0,32	0,02	0,01	0,02	0,01
S15St10	0,15	0,05	0,20	0,01	0,01	0,02	0,01
S15St11	0,06	0,03	0,09	0,01	0,00	0,01	0,00
S15St12	0,08	0,03	0,11	0,01	0,00	0,01	0,00
S15	***1,81***	***0,37***	***2,19***				
S16St01	0,03	0,01	0,03	0,00	0,00	0,01	0,00
S16St02	0,03	0,02	0,04	0,00	0,00	0,01	0,00
S16St03	0,08	0,03	0,09	0,01	0,00	0,01	0,00
S16St04	0,14	0,02	0,17	0,01	0,00	0,01	0,00
S16St05	0,11	0,03	0,13	0,01	0,00	0,01	0,00
S16St06	0,09	0,03	0,12	0,01	0,00	0,01	0,01
S16St07	0,21	0,03	0,24	0,02	0,00	0,02	0,01
S16St08	0,34	0,02	0,37	0,02	0,00	0,02	0,01
S16St09	0,16	0,04	0,18	0,01	0,00	0,02	0,01
S16St10	0,11	0,02	0,15	0,01	0,00	0,01	0,01
S16St11	0,04	0,02	0,06	0,01	0,00	0,01	0,00
S16St12	0,06	0,02	0,08	0,01	0,00	0,01	0,00
S16	***1,39***	***0,28***	***1,65***				

TABLE A9
Concentration of WSOC ($\mu g/m^3$)

Sample ID	WSOC	Sample ID	WSOC	Sample ID	WSOC	Sample ID	WSOC
S01St01	0,11	S05St01	0,07	S09St01	0,05	S13St01	0,05
S01St02	0,17	S05St02	0,04	S09St02	0,00	S13St02	0,01
S01St03	0,05	S05St03	0,03	S09St03	0,00	S13St03	0,04
S01St04	0,05	S05St04	0,10	s09St04	0,06	S13St04	0,07
S01St05	0,08	S05St05	0,00	S09St05	0,11	S13St05	0,03
S01St06	0,22	S05St06	0,02	S09St06	0,07	S13St06	0,05
S01St07	0,38	S05St07	0,16	S09St07	0,21	S13St07	0,21
S01St08	0,44	S05St08	0,17	S09St08	0,23	S13St08	0,15
S01St09	0,45	S05St09	0,09	S09St09	0,13	S13St09	0,06
S01St10	0,11	S05St10	0,08	S09St10	0,03	S13St10	0,10
S01St11	0,11	S05St11	0,04	S09St11	0,05	S13St11	0,02
				S09St12	0,03	S13St12	0,00
S01	*2,15*	*S05*	*0,80*	*St9*	*0,97*	*S13*	*0,79*
S02St01	0,13	S06St01	0,05	S10St01	0,08	S14St01	0,60
S02St02	0,05	S06St02	0,00	S10St02	0,04	S14St02	0,11
S02St03	0,02	S06St03	0,00	S10St03	0,03	S14St03	0,03
s02St04	0,06	S06St04	0,02	S10St04	0,04	S14St04	0,05
S02St05	0,10	S06St05	0,02	S10St05	0,01	S14St05	0,01
S21St06	0,06	S06St06	0,03	S10St06	0,10	S14St06	0,05
S02St07	0,14	S06St07	0,16	S10St07	0,23	S14St07	0,08
S02St08	0,09	S06St08	0,19	S10St08	0,25	S14St08	0,12
S02St09	0,10	S06St09	0,09	S10St09	0,15	S14St09	0,06
S02St10	-0,01	S06St10	0,06	S10St10	0,27	S14St10	0,05
S02St11	0,00	S06St11	0,02	S10St11	0,01	S14St11	0,00
				S10St12	0,00	S14St12	0,00
S02	*0,75*	*S06*	*2,26*	*S10*	*1,22*	*S15*	*1,17*
S03St01	0,09	S07St01	0,05	S11St01	0,24	S15St01	0,03
S03St02	0,03	S07St02	0,03	S11St02	0,45	S15St02	0,08
S03St03	0,05	S07St03	0,21	S11St03	0,08	S15St03	0,02
S03St04	0,10	S07St04	0,03	S11St04	0,05	S15St04	0,04
S03St05	0,06	S07St05	0,03	S11St05	0,04	S15St05	0,05
S03St06	0,05	S07St06	0,10	S11St06	0,12	S15St06	0,06
S03St07	0,11	S07St07	0,35	S11St07	0,22	S15St07	0,11
S03St08	0,02	S07St08	0,29	S11St08	0,21	S15St08	0,07
S03St09	0,02	S07St09	0,10	S11St09	0,10	S15St09	0,17
S03St10	0,00	S07St10	0,10	S11St10	0,04	S15St10	0,04
S03St11	0,05	S07St11	0,00	S11St11	0,00	S15St11	0,02
				S11St12	0,00	S15St12	0,00
S03	*0,59*	*S07*	*1,28*	*S15*	*1,54*	*S15*	*0,69*
S04St01	0,03	S08St01	0,03	S12St01	0,10	S16St01	0,06
S04St02	0,42	S08St02	0,05	S12St02	0,08	S16St02	0,03
S04St03	0,12	S08St03	0,03	S12St03	0,27	S16St03	0,10
S04St04	0,12	S08St04	0,00	S12St04	0,09	S16St04	0,05
S04St05	0,07	S08St05	0,04	S12St05	0,12	S16St05	0,05
S04St06	0,07	S08St06	0,11	S12St06	0,07	S16St06	0,05
S04St07	0,17	S08St07	0,14	S12St07	0,20	S16St07	0,12
S04St08	0,21	S08St08	0,26	S12St08	0,29	S16St08	0,12
S04St09	0,12	S87St09	0,09	S12St09	0,17	S16St09	0,07
S04St10	0,07	S08St10	0,08	S12St10	0,10	S16St10	0,21
S04St11	0,02	S08St11	0,02	S12St11	0,05	S16St11	0,04
			0,05	S12St12	0,00	S16St12	0,00
S04	*1,41*	*S08*	*0,86*	*S12*	*1,53*	*S16*	*0,91*

TABLE A10

Distribution parameters (median, 1st and 3rd quartiles) of the concentration of metals in various dimensional classes (ng/m^3)

Parameter		St 01 >18 µm	St 02 18-10 µm	St 03 10-5,6 µm	St 04 5,6-3,2 µm	St 05 3,2-1,8 µm	St 06 1,8-1,0 µm	St 07 1,0-0,56 µm	St 08 0,56-0,32µm	St 09 0,32-1,8 µm	St10 1,8-1,0 µm	St11 1,0-0,56µm	St12 <0,056
OC	Mediane	0,061	0,059	0,142	0,164	0,129	0,188	0,366	0,558	0,310	0,171	0,064	0,084
	1st quartile	0,031	0,039	0,082	0,136	0,105	0,162	0,267	0,348	0,263	0,133	0,053	0,068
	3rd quartile	0,078	0,082	0,179	0,263	0,147	0,274	0,576	0,679	0,387	0,266	0,087	0,167
EC	Mediane	0,001	0,010	0,020	0,027	0,025	0,034	0,040	0,037	0,043	0,511	0,029	0,034
	1st quartile	0,000	0,005	0,014	0,024	0,020	0,030	0,030	0,033	0,029	0,039	0,026	0,025
	3rd quartile	0,019	0,015	0,032	0,030	0,030	0,045	0,049	0,055	0,057	0,059	0,038	0,038
WSOC	Mediane	0,0513	0,3674	0,0312	0,0426	0,0349	0,0573	0,1425	0,1346	0,0965	0,0426	6,9400	ND
	1st quartile	0,0148	0,0591	0,0049	0,0036	0,0091	0,0264	0,0433	0,0737	0,0603	0,0054	0,1500	ND
	3rd quartile	0,1014	0,0491	0,0648	0,0549	0,0661	0,1005	0,2219	0,2592	0,1421	0,1010	0,0336	ND

TABLE A11

Daily average meteorological conditions.

Date	WS_aver	RH (%)_aver	T (°C)_aver	WS_err	RH (%)_err	T (°C)_err	Prevalent Direction
28/03/2019	0.977	32.379	13.345	0.099	0.693	0.487	ENE
29/03/2019	0.819	40.694	15.029	0.126	1.836	0.497	E
30/03/2019	0.318	34.871	15.771	0.023	1.798	0.783	N.D.
31/03/2019	0.346	36.739	16.179	0.029	1.504	0.611	ESE
01/04/2019	0.949	30.171	19.713	0.144	1.474	0.648	NE
02/04/2019	0.402	40.845	19.025	0.071	2.613	0.705	SSE
03/04/2019	0.483	70.241	14.976	0.060	1.475	0.272	ESE
04/04/2019	0.795	68.576	15.028	0.113	1.005	0.202	N.D.
05/04/2019	0.496	80.577	10.958	0.078	1.376	0.353	ESE
06/04/2019	0.239	63.534	13.020	0.020	2.862	0.552	N.D.
07/04/2019	0.329	75.585	12.052	0.052	1.638	0.222	ESE
08/04/2019	0.391	77.348	13.531	0.054	1.814	0.423	ESE
09/04/2019	0.245	59.808	16.432	0.030	2.507	0.534	SE
10/04/2019	0.586	79.791	13.978	0.106	2.467	0.354	ESE
11/04/2019	0.604	78.167	12.796	0.090	3.901	0.294	ESE
12/04/2019	0.439	66.141	10.625	0.064	1.748	0.150	E
13/04/2019	0.964	50.196	11.522	0.150	1.712	0.384	N.D.
14/04/2019	0.911	51.437	12.051	0.132	0.930	0.386	ENE
15/04/2019	0.686	42.334	15.332	0.093	2.528	0.614	NE
16/04/2019	0.340	36.913	15.601	0.032	2.017	0.765	N.D.
17/04/2019	0.385	33.611	17.457	0.050	1.237	0.787	N.D.
18/04/2019	0.820	38.073	17.753	0.131	1.288	0.632	ENE
19/04/2019	0.346	39.837	18.632	0.034	1.832	0.797	S
20/04/2019	0.333	32.874	20.050	0.027	1.933	0.854	SSE
21/04/2019	0.316	32.799	18.440	0.028	1.831	0.682	S
22/04/2019	0.348	52.764	17.091	0.036	1.429	0.535	N.D.
23/04/2019	0.225	82.654	14.920	0.017	1.567	0.125	SE
24/04/2019	0.313	84.114	16.473	0.034	1.451	0.308	SE
25/04/2019	0.348	72.000	18.387	0.046	1.797	0.517	N.D.
26/04/2019	0.555	70.739	18.089	0.074	2.807	0.533	ESE
27/04/2019	0.529	70.960	15.251	0.067	1.917	0.385	E
28/04/2019	0.337	68.257	14.237	0.025	2.444	0.512	SSE
29/04/2019	0.527	60.072	12.806	0.086	1.066	0.263	ESE
30/04/2019	0.477	60.390	14.562	0.064	0.835	0.345	ESE
01/05/2019	0.379	57.725	16.599	0.027	1.701	0.616	SSE
02/05/2019	0.354	59.769	17.006	0.027	1.238	0.554	S
03/05/2019	0.625	78.056	15.191	0.072	1.441	0.163	SE
04/05/2019	0.460	70.325	16.234	0.053	2.529	0.352	SSE
05/05/2019	0.677	60.262	12.202	0.077	3.178	0.298	NE
06/05/2019	0.548	48.160	11.277	0.100	1.408	0.279	ENE
07/05/2019	0.375	44.854	13.277	0.038	2.145	0.664	S
08/05/2019	0.480	61.021	14.645	0.044	2.297	0.564	N.D.
09/05/2019	0.797	86.936	13.159	0.148	1.061	0.242	ESE
10/05/2019	0.341	68.253	15.915	0.028	2.215	0.701	N.D.
11/05/2019	0.471	67.099	17.279	0.074	2.166	0.587	N.D.
12/05/2019	0.326	70.460	15.264	0.065	2.274	0.177	ENE

WS: wind speed (m/s); RH: relative humidity; T: temperature
N.D.: not defined prevalent wind direction

Table A12

Daily average particle concentrations in number and in mass

Date	Conc D<0.25 µm (#/cm^3)	Conc D>0.25 µm (#/cm^3)	PM_{10} (µg/m^3)	$PM_{2.5}$ (µg/m^3)	PM_1 (µg/m^3)
28/03/2019	3408.79	75.31	7.11	6.66	6.09
29/03/2019	4904.29	143.46	12.67	11.91	11.21
30/03/2019	5988.28	183.74	16.40	15.43	14.53
31/03/2019	6412.76	180.82	16.17	15.34	14.49
01/04/2019	4507.08	183.22	17.58	15.91	14.76
02/04/2019	7911.58	286.61	27.54	24.48	22.45
03/04/2019	6296.46	301.32	27.59	26.31	23.97
04/04/2019	4058.58	185.30	21.72	19.87	16.34
05/04/2019	8494.85	86.80	9.93	9.27	7.70
06/04/2019	9000.12	88.70	9.61	8.87	7.65
07/04/2019	7994.10	163.38	15.66	14.93	13.78
08/04/2019	10000.30	228.52	19.89	19.07	18.01
09/04/2019	7612.92	269.90	22.26	21.39	20.47
10/04/2019	7290.17	255.54	20.65	19.99	19.05
11/04/2019	5639.81	78.23	6.71	6.56	6.35
12/04/2019	6677.57	162.95	13.73	13.32	12.89
13/04/2019	3702.41	243.25	19.51	19.03	18.46
14/04/2019	2072.76	129.36	10.67	10.50	10.27
15/04/2019	5039.03	207.21	18.36	17.12	16.30
16/04/2019	7188.61	242.03	23.41	20.81	19.27
17/04/2019	7819.23	188.61	19.68	16.99	15.41
18/04/2019	6324.13	118.33	12.33	10.98	10.03
19/04/2019	6611.46	103.72	10.59	9.68	8.97
20/04/2019	6147.92	106.05	10.27	9.64	9.02
21/04/2019	5217.46	106.60	10.41	9.87	9.24
22/04/2019	4671.76	166.55	16.75	15.93	14.40
23/04/2019	9218.28	237.79	25.30	23.30	19.94
24/04/2019	5557.00	173.82	44.39	36.36	19.47
25/04/2019	6627.52	143.31	41.48	32.20	16.57
26/04/2019	7508.75	100.26	31.98	23.60	11.81
27/04/2019	7094.47	50.22	8.38	7.09	4.97
28/04/2019	4366.19	37.59	4.60	4.26	3.55
29/04/2019	6905.40	76.85	7.32	7.04	6.61
30/04/2019	7055.75	71.22	7.42	6.80	6.12
01/05/2019	7422.99	78.77	7.76	7.31	6.73
02/05/2019	7648.42	96.72	9.66	8.91	8.07
03/05/2019	6427.37	113.44	12.35	11.35	9.72
04/05/2019	6384.65	62.88	6.79	6.47	5.70
05/05/2019	2018.55	18.11	2.02	1.86	1.64
06/05/2019	3897.49	37.37	3.70	3.54	3.33
07/05/2019	10850.23	80.01	8.19	7.62	7.01
08/05/2019	8080.43	68.38	7.57	6.93	6.14
09/05/2019	7256.04	97.40	10.61	10.06	8.68
10/05/2019	8673.97	79.84	8.12	7.59	6.98
11/05/2019	7311.79	74.43	7.98	7.49	6.70
12/05/2019	3150.68	31.00	3.49	3.27	2.85

Table A13
Average size distributions in number and in mass

Diameter min (µm)	Diameter max (µm)	Diameter average (µm)	**Average Particle Number ($\#/cm^3$)**	**Average Particle mass ($\mu g/m^3$)**
0.009	0.25	0.1295	**6417**	**0.59**
0.25	0.28	0.265	**55.44**	**1.12**
0.28	0.3	0.29	**36.92**	**1.00**
0.3	0.35	0.325	**21.50**	**0.80**
0.35	0.4	0.375	**11.31**	**0.65**
0.4	0.45	0.425	**4.72**	**0.39**
0.45	0.5	0.475	**1.72**	**0.20**
0.5	0.58	0.54	**1.80**	**0.31**
0.58	0.65	0.615	**0.73**	**0.19**
0.65	0.7	0.675	**0.25**	**0.09**
0.7	0.8	0.75	**0.32**	**0.16**
0.8	1	0.9	**0.20**	**0.17**
1	1.3	1.15	**0.17**	**0.31**
1.3	1.6	1.45	**0.09**	**0.33**
1.6	2	1.8	**0.10**	**0.71**
2	2.5	2.25	**0.05**	**0.71**
2.5	3	2.75	**0.02**	**0.55**
3	3.5	3.25	**0.01**	**0.27**
3.5	4	3.75	**0.00**	**0.20**
4	5	4.5	**0.00**	**0.28**
5	6.5	5.75	**0.00**	**0.11**
6.5	7.5	7	**0.00**	**0.03**
7.5	8.5	8	**0.00**	**0.02**
8.5	10	9.25	**0.00**	**0.02**
10	12.5	11.25	**0.00**	**0.02**
12.5	15	13.75	**0.00**	**0.02**
15	17.5	16.25	**0.00**	**0.01**
17.5	20	18.75	**0.00**	**0.01**
20	25	22.5	**0.00**	**0.03**
25	30	27.5	**0.00**	**0.04**
30	32	31	**0.00**	**0.02**